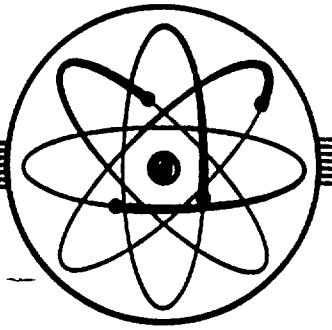
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Radiation Uses in INDUSTRY *and* SCIENCE



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By

LLOYD E. BROWNELL

THE UNIVERSITY OF MICHIGAN

1961

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PREFACE

This book has been prepared as a source of information for industrial management on some of the existing and potential applications of radioisotopes and nuclear radiations in industry and research. Also it is intended for use by scientific and technical personnel not familiar with this field, but seeking information over a broad spectrum of possible applications. Training in physics, chemistry, mathematics, and the biological sciences is not necessary, but some familiarity with these fields will be helpful to the reader.

Background for the preparation of the text has involved several years (1951 to 1960) research study, consulting work, and teaching a university course on possible uses of nuclear radiations. Considerable text material has been taken from reports and technical papers from these research studies and from the lecture notes for this course. A bulk of information and illustrations came from the periodical literature, proceedings of both Geneva Conferences on Peaceful Uses of Atomic Energy, AEC reports, QMC publications, and papers from various technical meetings. Different parts of this text were written at different times; therefore, some chapters contain more recent information than others.

Many individuals assisted in the preparation of this manuscript by writing particular sections, reviewing the literature, editing the original drafts, checking references, and typing. The names of all individuals who assisted in this work will not be given, but I wish to express my thanks and sincere appreciation to all who helped. In particular, I wish to thank Dr. John V. Nehemias, former health physicist at the Fission Products Laboratory and Dr. Ardath Emmons, former supervisor of the Phoenix Radiation Laboratory, at the University of Michigan; Dr. Roland Hannan, former Head of Radiation Division at Low Temperature Research Station, and Ian Scarisbrick, former physicist at Cavendish Laboratory, at Cambridge University, England; and Dr. E. Watson Williams, editor in biological sciences, Cambridge, England.

Readers interested in the more technical problems of the use of radioisotopes are referred to a companion text (by L. E. Brownell) entitled "Laboratory Manual for the 1961 Summer Institute on Radioisotope Technology Sponsored by AEC and NSF at The University of Michigan." This manual contains information on: (1) safety in the use of radioisotopes and radiation; (2) design of radioisotope laboratories; (3) film, glass, and chemical dosimetry; (4) procedures for gamma shielding calculations; (5) problems and procedures in counting nuclear radiations; and (6) experiments designed for teaching radioisotope technology.

Lloyd Brownell
Consultant
Professor of Nuclear and Chemical Engineering, University of Michigan

April 28, 1961

CONTENTS

Preface iii

Chapter 1 INTRODUCTION

The Nature of Atomic Energy 1
Nuclear Reactors and Their Importance 2
Growth of Radioisotope Use 2

Chapter 2 PROPERTIES OF IONIZING RADIATIONS

Natural Radioactivity 3
Early Observations 3
Radioactive Decay and Half-Life 5
Natural Radioactive Series 7
Natural Sources of Radioisotopes and Fissionable Materials 9
Artificial Radioactivity 9
Discovery 9
Transuranic Elements, Discovery of Fission and the Neptunium Series . . . 10
The Bohr Compound-Nucleus Theory 11
The Binding-Energy Curve 11
Fission 12
Fusion 12
Alpha-Particle Emission 12
Beta-Particle Emission 12
Production of Artificial Radioisotopes 13
Alpha-Particle Emission 14
Absorption of Alpha-Particle Radiation 23
Beta-Particle Emission 28
Absorption of Beta-Particle Radiation 30
Gamma Photon Emission 31
Interaction of Gamma Radiation and Matter 33
Photoelectric Effect 34
Compton Scattering 35
Pair Production 36
Nuclear Radiations as Waves and as Particles 37
Nature of a Particle 39
Nature of a Wave 39
Absorption of Gamma Radiation 40
Absorption Coefficients for Narrow Beams 40
Half-Value and Tenth-Value Thicknesses 42
Different Energies 42
Build-Up Factors 43
Broad-Beam Coefficients 46
References 46

Chapter 3 CONTROLLED FISSION AND NUCLEAR REACTORS

The Neutron 52
Methods of Detection 52
Charge 53
Mass 53
Neutron Capture Cross Sections 53
The Discovery of Nuclear Fission 54
The Fission Process 56
The Fission Chain Reaction 59
Controlled Fission 61
The Energy of Fission 63
Reactor Types 65
Neutron Energy 65
Moderators 68
Coolants 69
Water-Cooled Reactors 70
Organic Liquid-Cooled Reactors 71
Boiling Water Reactors 71
Liquid-Metal-Cooled Reactors 71
Gas-Cooled Reactors 71
References 72

Chapter 4 REACTORS AS INDUSTRIAL SOURCES OF RADIATION

Radiations from the Reactor Core 78
Reactor Core Promotion of Chemical Reactions 79
Advantages of Converting Fission Energy to Chemical Energy 79
Systems Suitable for Chemical Reaction in a Reactor Core 79
Oxidation of Nitrogen in a Reactor Core 80
Reactor Core Processing of Foods 81
Types of Applications 81
Problems of Using the Reactor Core 81
Radioactivity of Gaseous Fission Products from Fuel Solutions 82
Theoretical Yield Relationships 83
Decay Scheme 84
Calculated Fission Gas Build-Up 84
Fission Gases Continuously Produced in a Multipurpose Reactor 84
Fission Gas Radiation Source 87
Fission Product Gas Used Outside the Reactor Core 89
Fission Gas Irradiator Using a Mobile Reactor 89
Fission Gases and Reflector Solutions as Food Irradiation Sources 91
Radioactivity from Mixed Fission Products in Fuel Solution 93
Reactor Liquid Fuels as Food Irradiation Sources 93
Induced Radioactivity 94
Use of a Shielded Nuclear Fuel 94
Reactor Fuel Elements as Sources of Radiation 94
MTR Fuel Elements 96
Use of Fuel Elements in Mobile Food Irradiation Facility 97
Radioisotopes Produced by Neutron Capture as Food Irradiation Sources 98
Production of Gamma Emitter in Reactor Core 100
Production of Gamma Emitter in Reactor Blanket 100
Ordinary Water (H_2O) Blanket 101
Heavy Water (D_2O) Blanket 101
Beryllium Oxide (BeO) Blanket 102
Production of Gamma Emitter in Sodium Coolant 102
Beryllium Oxide-Moderated Sodium-Cooled Reactor 102

Graphite-Moderated Sodium-Cooled Reactor 102
Combined Production of Gamma Emitter in Core and Blanket 103
Reactors with Removable Solids as Gamma Radiation Sources 103
Multipurpose Reactor Types 103
Liquid Metal-Cooled Heterogeneous Reactors 104
Aqueous Homogeneous Reactor 106
Liquid Metal Fuel Homogeneous Reactors 106
Characteristics of Carrier Metal 106
Reactor Core 107
Flow Diagram 108
Multipurpose Use of the LMFR in a Tire Plant 110
Vulcanization of Tires with Gamma Radiation 110
Proposed Commercial Irradiation of Tires 112
Summary 113
Research Reactors as Sources of Radiation 113
X-10 113
BEPO 113
NRX 113
MTR 114
Access Openings in MTR 115
References 116

Chapter 5 USES OF RADIATION IN INDUSTRY

Industrial Radiography 120
Use of Radiography 121
Selection of Radioisotopes 122
Cobalt-60 125
Cesium-137 126
Iridium-192 128
Cerium-144 129
Thulium-170 129
Beta-Particle Excited X-Ray Sources 129
Industrial Radiation Gauges 131
Advantages of Radiation Gauges 132
Sources for Radiation Gauges 133
Radiation Detectors 134
Absorption Gauges 135
Gamma Absorption Gauges 135
X-ray (radioisotope) Gauges 137
Beta-Particle Absorption Gauges 137
Scattering or Reflection Gauges 140
Beta-Particle Scattering 140
Gamma Scattering 141
Neutron Scattering 141
Alpha-Particle Gauges 141
Luminescent Compounds, Paints, and Light Sources 142
Mechanism of Alpha-Particle Activation 142
Mechanism of Beta-Particle Activation 142
Advantages of Beta-Particle Emitters 143
Typical Uses 143
Problems Requiring Additional Study 144
Static Elimination 144
Alpha-Ray Static Eliminators 144
Beta-Particle Static Eliminators 144

Transforming Radiation Energy Atomic Batteries and Electrostatic Generators . . . 145
Direct Charging Battery . . . 146
Contact Potential Battery . . . 147
Thermopile Battery . . . 148
Thermionic Converters . . . 148
Scintillator-Photocell Battery . . . 149
Semiconductor Battery . . . 149
Irradiated Electrolytic Cell . . . 150
Electrostatic Generator . . . 151
High-Pressure Hydrogen-Oxygen Cell . . . 151
References . . . 153

Chapter 6 APPLICATIONS OF TRACER STUDIES AND AUTORADIOGRAPHY

Tracer Methods of Analysis . . . 157
Isotope Dilution Method . . . 158
Reverse Isotope Dilution Method . . . 159
Isotope Derivative Method . . . 159
Radiometric Analysis Method . . . 160
Activation Analysis Method . . . 160
Autoradiographic Method . . . 161
Counting Nuclear Radiations in Tracer Experiments . . . 161
Geiger-Müller Counters . . . 162
Scintillation Counters . . . 163
Auxiliary and Counting Equipment . . . 165
Effect of Alpha-Particle Radiation on Counting . . . 165
Effect of Beta-Particle Radiation on Counting . . . 165
Effect of Gamma Radiation on Counting . . . 166
Industrial Applications . . . 167
Wear Studies . . . 168
Mass Transfer . . . 170
Metal Industry . . . 171
Oil Industry . . . 172
Other Industries . . . 177
Research . . . 179
Chemical Reactions . . . 179
Water Analysis . . . 180
Diffusion . . . 180
Vulcanization . . . 183
Agriculture . . . 183
Photosynthesis . . . 184
Plant Nutrients . . . 186
Animal Husbandry . . . 187
References . . . 189

Chapter 7 PROMOTION OF CHEMICAL REACTIONS BY RADIATION

Scope of Radiation Chemistry . . . 195
Photochemistry . . . 195
Radiation Chemistry . . . 196
Types of Chemical Bonds . . . 196
Covalent Bonds . . . 196
Ionic Bonds . . . 198
Semipolar or Dative Bonds . . . 199
Metallic Bonds . . . 199

Interaction of Ionizing Radiation with Matter 199
Effect of Ionizing Radiation Upon Molecules 199
Gases 200
Liquids 200
Solids 202
Reaction Processes in Radiation Chemistry 202
Impact Processes 202
Electron Capture 203
Ionization and Excitation Transfer 204
Neutralization of Positive Ions 204
Dissociation of Excited Molecules 204
Thermodynamic and Kinetic Considerations for Reaction Promoted by Ionizing Radiation 205
Chemical Reactions of Possible Industrial Interest 206
Inorganic Reactions 207
Organic Reactions 208
Synthesis 209
Oxidation 210
Halogenation 210
Reduction 211
Polymerization of Ethylene 211
Cross-linking Polyethylene 213
Vulcanization of Rubber 215
Copolymerization of Various Unsaturated Hydrocarbons with Sulfur Dioxide 218
Polymerization of Acetylene 219
Chlorination of Benzene 219
Chlorination of Toluene, Xylene, and Mesitylene 220
Oxidation of Ethylene 222
Cracking of Hydrocarbons 224
Radiation Damage to Materials of Construction 224
Organic Materials 224
Inorganic Materials 224
References 226

Chapter 8 EFFECTS OF RADIATION ON BIOLOGICAL MATERIALS AND SIMPLE ORGANISMS

Effect of Ionizing Radiations on Biochemicals 231
Direct and Indirect Action of Radiation 231
Proteins 232
Protein Protection by Complexing 237
Amino Acids 239
Carbohydrates 240
Lipids 243
Vitamins 244
Water-Soluble Vitamins 244
Oil-Soluble Vitamins 246
Nutrilites 249
Enzymes 251
Radiation Effects Within a Biological Cell 253
Effect of Ionizing Radiation on Microorganisms 255
Effect of Low Doses 255
Target Theory 256
Method of Expressing Dose 258
Specific Microorganisms in Aqueous Media at Room Temperature 259
Bacteria 259
Molds and Yeasts 263

Protozoa 267
Viruses and Bacteriophages 267
Chemical Composition of Media 269
Temperature 271
Pretreatment with Heat 271
Other Factors 273
Effect of Ionizing Radiation on Some Insects and Parasites 273
Mexican Fruit Fly 273
The Parasitic Nematode, *Trichinella Spiralis* 278
References 278

Chapter 9 EFFECTS OF RADIATION ON PLANTS, FOODS, AND PHARMACEUTICALS

Dose Ranges for Irradiation 284
Low-Dosage Irradiation of Plants and Plant Products 286
Production of Mutants 286
Irradiation of Potatoes 291
Weight Loss 292
Suberization and Periderm Formation 293
Carbohydrate Analyses 294
Respiration 294
Problem of Potato Rot 295
Later Observations 295
Studies of Potato Irradiation in Europe 295
Irradiation of Onions 295
Irradiation of Other Root Crops 296
Irradiation of Seeds 297
Irradiation of Fruit 298
Apples 298
Cherries and Peaches 298
Plums 299
Strawberries 300
Other Fruits 300
Irradiation of Cereal Products 301
Radiopasteurization of Milk and Milk Products 302
Radiopasteurization of Egg Products 305
Radiopasteurization of Meat, Poultry, and Seafood 305
Taste Panel Tests on Pork 305
Taste Panel Tests on Beef 307
Seafoods 307
Microbiological Tests on Beef 309
Microbiological Tests on Fish 309
Microbiological Tests on Poultry 311
Odor and Flavor Problems of High-Dosage Irradiation of Meat 313
Fresh Meat 313
Canned Meats 314
Ground Beef 314
Beef Tenderloin 317
Pork Luncheon Meat 317
Cured Ham 317
Meat Fats 317
Program of the Office of the Quartermaster General (U. S. Army) on Food Irradiation 317
Effect of Gamma Irradiation on the Potency of Pharmaceutical Products 322
Commercial Radiosterilization of Surgical Sutures 324

Wholesomeness of Irradiated Foods and Food Constituents 324
Procedures Recommended by Food and Drug Administration 324
Possibility of Induced Radioactivity 324
Possibility of Toxicity 326
Short-term Experiments with Rats 326
Long-term Experiments with Rats 329
Other Short-term Feeding Experiments 329
Long-term Experiments with Chickens 330
Long-term Experiments with Dogs and Monkeys 330
Possible Carcinogenicity of Irradiated Foods 330
Feeding Irradiated Foods to Human Beings 331
General Comments on Wholesomeness of Irradiated Food 333
Status of Wholesomeness of Irradiated Food as of 1960 334
References 335

Chapter 10 POSSIBLE PRACTICAL APPLICATIONS OF THE BIOLOGICAL EFFECTS OF GAMMA RADIATION

Control of Insect Populations 341
Screwworm (*Callitroga hominivorax*) 341
The Curacao Experiment 342
Eradication of the Screwworm in Southeastern United States 343
Raising the Quarantine on Nonedible Agricultural Imports and Exports 345
Sterilization of Animal Products for Control of Disease 346
Control of Pink Bollworm in Imported Cotton 346
Design for a Cotton-Bale Irradiator 347
Fumigation Costs Compared to Irradiation Costs for Cotton Bales 349
Irradiation of Tobacco and Other Nonedible Products 349
Insect Control in Edible Products 350
Control of Fruit Flies 351
Design for a Mobile Product Irradiator 351
Control of Insects in Flour, Grain, and Meal 352
Irradiator for 100-lb Bags of Grain, Flour, or Meal 356
Gravity-flow Grain Irradiator 356
Irradiation of Foods as a Public Health Measure 360
Trichinosis 360
Design for a Pork Irradiator 361
Beef Tapeworm 361
Tuberculosis 363
Brucellosis 363
Ornithosis 363
Infections from Seafood 363
Radiopasteurization of Fresh Meats, Poultry, Eggs, and Seafoods 363
Design for Radiopasteurization Facility for Fresh Meat 365
High Radiopasteurization in Preserving Food 365
Design for High-Radiopasteurizing Facility 368
Potato Irradiation for Sprout Inhibition 370
Design for a Potato Irradiator 370
Radiosterilization of Foods 373
Designs for Food Radiosterilizers 374
U. S. Army Ionizing Radiation Center 377
Radiosterilization of Surgical Items 377
Advantages of Radiosterilization of Bone-bank Bone 378
Comparative Studies on Bone-bank Bone 379
Use of Radiosterilized Human Bone-bank Bone 379

Design for a Gamma-Radiosterilizer for Surgical Products 379
Radiosterilization of Bulk Medical Supplies 381
Design for a Radiosterilizer for Bulk Medical Supplies 382
Radiosterilization of Hospital Mattresses and Surgical Items 384
Design for a Hospital Mattress Irradiator 384
Cost Estimate for a Hospital Mattress Irradiator 387
References 388

Chapter 11 ELECTRONIC DETECTION AND MEASUREMENT OF RADIATION

Ionization of Gases 391
Principle of Operation of Ionization Chamber 392
Effect of Varying the Electric Field 393
Electroscopes and Condenser Type Dosimeters 394
Condenser Type Meters 396
Ion-Chamber Dose-Rate Meters 399
Measurements with Dose-Rate Meters 401
Thimble Chambers 402
Proportional Counters 404
Geiger-Müller Counters 406
Quenching G-M Tubes 407
G-M Meters 408
Geiger-tube Designs 409
Neutron Counters 410
Counters for Fast Neutrons 410
Counters for Slow Neutrons 411
Neutron Detectors for Various Flux Ranges 412
Scintillation Counters 413
Phosphors 413
Photomultiplier Tubes 415
Applications of Scintillation Counters 416
Use of Transistors in Radiation Detectors 417
References 417

Chapter 1
INTRODUCTION

Many people consider the "peaceful atom" to be synonymous with production of electric power from nuclear fuel. This is understandable because nuclear power is a major objective of many countries of this energy-hungry world and is of vital interest to all others. There is, however, another important side of the peaceful atom story—nuclear radiations and radioisotopes and their use in industry, agriculture, medicine, and research.

The purpose of this book is to survey some of the nonpower uses of atomic energy (exclusive of medical uses) that have already proved practical, and to suggest other applications. The volume cuts across a number of disciplines. It is written for engineers, scientists, and interested laymen—management people in particular. Those not satisfied with the extent of the coverage of their specialties or with the technical level will, it is hoped, make use of the considerable number of references.

This book will serve its purpose if its panorama of radiation applications and potentialities improves the understanding and judgement of industrial managers, helps nuclear specialists to extend their horizons, or challenges nonnuclear scientists and engineers to contribute their talents in developing new uses for radiation and radioactive materials.

THE NATURE OF ATOMIC ENERGY

Atomic energy (more correctly nuclear energy) is the basic form of energy of the universe. Nuclear reactions in the sun release energy which reaches the earth as light. Through photosynthesis plant life converts the light to chemical energy. Thus, when we burn coal, oil or gas, we are releasing energy which had its origin in the solar fires. Research is in progress in several countries to duplicate under controlled conditions the nuclear reactions that take place in the sun and stars. Called thermonuclear reactions because they occur in significant numbers only at temperatures of millions of degrees centrigrade, they involve the combining of nuclei of light atoms to form heavier nuclei. In one solar process, for example, a deuteron (heavy hydrogen nucleus) combines with a proton (hydrogen nucleus.) The mass of the resulting helium nucleus is less than the sum of the masses of the deuteron and the proton. The mass difference is converted to energy and the amount is given by Einstein's celebrated equation: $E = mc^2$ where E is energy, m is mass, and c is the velocity of light.

The principal nuclear processes described in this book—nuclear fission and radioactive decay—differ from thermonuclear processes in that they involve the breakdown rather than the fusion of atomic nuclei. Nevertheless, they are similar to fusion in a very fundamental respect: they too are accompanied by the conversion of mass to energy and the amount of energy released is governed by Einstein's equation. The energy released in a nuclear reaction is enormous compared to that liberated in a chemical reaction involving the same amount of matter. To illustrate the point, the complete fissioning of one kilogram (2.2 lb) of uranium would yield as much energy as the exploding of 20,000 tons of TNT! The total mass loss in the fissioning of the uranium would be approximately one gram. Prevailing scientific opinion is that ordinary chemical reactions are also accompanied by mass losses in accord with the

Einstein equation. In the case in point the mass lost by the TNT would also be one gram or two parts in ten billion! Direct confirmation of mass loss in a chemical reaction is impossible at present, because the quantity of mass changed to energy is too small to be measured by even the most sensitive instruments available today.

NUCLEAR REACTORS AND THEIR IMPORTANCE

Fission may proceed at an extremely rapid rate, as in an atom bomb, or at a much slower rate under controlled conditions in a nuclear reactor. The importance of the development of nuclear reactors is an accomplishment in the application of science that can hardly be overstated. Not only do these machines permit the practical use of the heat from fission, they also produce radiation and radioactive material in prodigious amounts. Before the advent of controlled fission the amounts of radiation and radioactive material at man's disposal were sharply limited. True, he had the natural radioisotopes such as radium and some radioisotopes produced artificially by bombarding materials with high speed particles (protons, deuterons) from accelerators. Nevertheless, the scarcity and resulting high cost of radium (only a few pounds had been isolated up to 1940) and the high cost and generally short life of cyclotron-produced isotopes impeded use, particularly by industry.

As nuclear reactors became available in the 1940's the picture radically changed. A large power or test reactor produces as much radiation as many tons of radium. Nonradioactive materials can be made radioactive by subjecting them to the neutron flux present in a reactor. Moreover, the fission process results in a host of radioactive fission products, some of intense activity. The activity of fission products built up is enormous, particularly in a high power reactor after extended operation, so great that it is a major impediment to the recovery of the "unburned" nuclear fuel. For the most part, fission products have been stored as by-product wastes from fuel reprocessing operations. Because of their radioactivity they cannot be discarded in the manner common for industrial waste. Storing them is costly, a fact that weighs heavily in the economics of nuclear power. Turning these radioactive wastes into economic assets is one of the most challenging problems of the Nuclear Age.

GROWTH OF RADIOISOTOPE USE

A few statistics will show the phenomenal growth in radioisotope use during the past decade and a half. In 1946 (the first year the AEC made radioisotopes available to the civilian economy) there were only 86 users of radioisotopes. The Oak Ridge National Laboratory, the AEC's principal isotope distribution center, made 281 shipments totaling 8 curies. By the end of 1959 there were 5,419 organizations and individuals licensed to use radioisotopes. In 1959 the Oak Ridge National Laboratory made 3,741 shipments totaling 245,000 curies bringing the cumulative number of shipments to 14,002 and the number of curies shipped to 860,000. While the growth in the use of radioisotopes as indicated by these numbers is impressive indeed, informed opinion is that the surface of radioisotope applications has just been scratched. Myriad uses for radioisotopes and radiation are still in the research stage of development and others have not been explored at all. The saturation point for tried and proven uses has certainly not been reached. It is to encourage wide consideration of both the existing and potential applications of radioisotopes and radiation that this book is offered.

Chapters 2 and 3 treat the physical principles of radiation, radioisotopes, controlled nuclear fission, and reactors, some knowledge of which is required for understanding the applications discussed in the remaining 8 chapters. Reactors as industrial sources of radiation are covered in Chapter 4. Chapter 5 deals with industrial applications of radiation such as gauging and radiographic inspection. Uses of radioisotopes in tracer studies and autoradiography are treated in Chapter 6. A variety of applications in industry and agriculture are discussed. The effects of ionizing radiations on the promotion of chemical reactions are covered in Chapter 7. Chapters 8 and 9 deal with the effects of radiation on biological materials, simple organisms, plants, foods, and pharmaceuticals. Possible practical applications of the biological effects of gamma radiation are treated in Chapter 10. A survey of instruments and techniques used in radiation detection and measurement is given in Chapter 11.

Chapter 2
PROPERTIES OF IONIZING RADIATIONS

Ionizing radiations are those which interact with the electrons of an atom and eject one or more of them from their orbits. The remaining atom, charged in the process, is called an ion. The energy required to cause ionization depends on two things: the atom or molecule involved, and the orbit from which the electron is expelled. The ionizing radiation loses energy not only when the electron escapes from its orbit but also in giving kinetic energy to the electron and the ion produced. There is no sharp energy threshold for the ionization process: radiations with energies of 50 ev or more are generally classed as ionizing, although energies down to a few electron volts will produce ionization in certain systems.

Three types of nuclear radiations, alpha, beta, and gamma are capable of producing ionization but neutrons interact only very weakly with electrons and do not cause ionization. However, if fast neutrons pass through hydrogenous matter, fast "knock-out" protons are formed and these cause intense ionization.

This chapter discusses natural and artificial radioactivity, the three natural series of radioisotopes, decay, half-life, and binding energy of nuclei. Interactions of ionizing radiations with matter are discussed in some detail. The behavior of these radiations as matter and as wave motion is mentioned. The chapter ends with some data on narrow and broad beam absorption coefficients for common materials.

NATURAL RADIOACTIVITY

The following paragraphs describe naturally occurring radioisotopes and discuss their properties.

EARLY OBSERVATIONS

Henri Becquerel accidentally discovered natural radioactivity in 1896.[1] Soon after this, the radiations from radioactive substances were separated into three distinct components according to their behavior in a magnetic field.

Alpha radiation was found to carry a positive electric charge and to have a mass of approximately 4 atomic mass units. It was then demonstrated spectrographically that the gas produced as a result of alpha decay was identical with helium gas. This type of radiation was, as a result, considered to be a particle emission, and the use of the term, alpha ray, became anomalous.

Beta radiation was found to carry a negative electric charge of one-half the magnitude of the charge found on alpha particles, and to possess a much smaller mass. Subsequent work has shown beta radiation to be physically identical with electrons. It then became common to speak of "beta particles" rather than beta rays.

Gamma radiation was the third type of radiation and was more difficult to identify. It was apparently not affected by a magnetic field and was very penetrating. It was eventually shown to be electromagnetic radiation of very short wavelength, shorter even than X-radiation, and was considered to be a continuous transverse electromagnetic wave phenomenon.

Figure 2.1 is drawn from a sketch prepared originally by Marie Curie for inclusion in her doctoral thesis[2] and is her representation of the behavior of alpha, beta, and gamma radiation in a magnetic field.

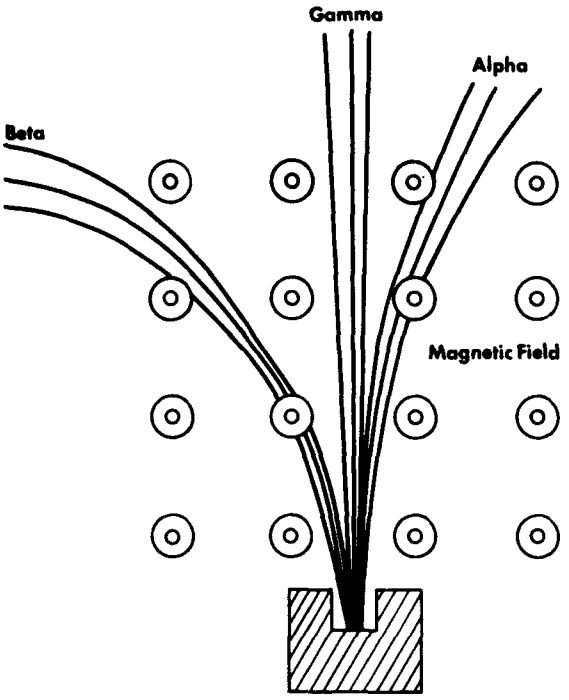


Fig. 2.1 — Marie Curie's representation of the behavior of alpha, beta, and gamma radiation in a magnetic field.

These observations of natural radioactivity were the first evidence of the divisibility of the atomic nucleus and the transformation of one element into another. Some of the early experiments by Becquerel[1] and Rutherford and Soddy[3] gave clues regarding the nature of radioactive disintegration. In 1900 it was found that if ammonium carbonate was used to precipitate a uranium salt and the precipitate was redissolved in an excess of reagent, a small residue of highly radioactive salt was left undissolved. However, the solution of uranium salt when concentrated by evaporation showed very little radioactivity. Thus it appeared that most of the radioactive ingredient in natural uranium could be separated out by the procedure described. The uranium in the insoluble and highly radioactive precipitate was named uranium X (UX) to distinguish it from natural uranium (U). Becquerel[1] allowed fractions of U and UX to stand for some time and observed an increase in the activity of U and a decrease in the activity of UX so that the relative activities were equal in about 20 days and were completely reversed after 140 days, as is shown in Fig. 2.2. Rutherford and Soddy[3] observed a similar phenomenon with thorium (Th) and thorium X (ThX), as shown in Fig. 2.3.

The shapes of the curves in Figs. 2.2 and 2.3 are essentially the same, and the major difference is that the change in the activities of Th and ThX occurs much more rapidly than in the case of U and UX. The decay curve of UX can be expressed as

$$A_X = A_{X_0} e^{-\lambda t} \tag{2.1}$$

where A_{X_0} = activity of UX at zero time

A_X = activity of UX at any time t

λ = disintegration constant

t = time

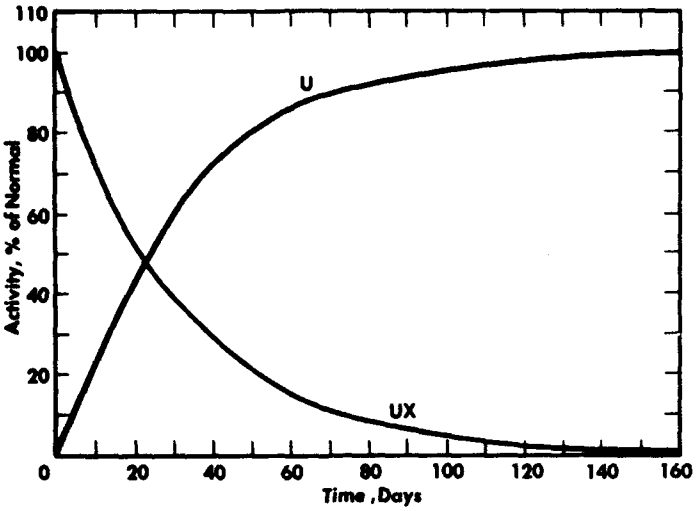


Fig. 2.2— Increase and decrease in activity of U and UX fractions.

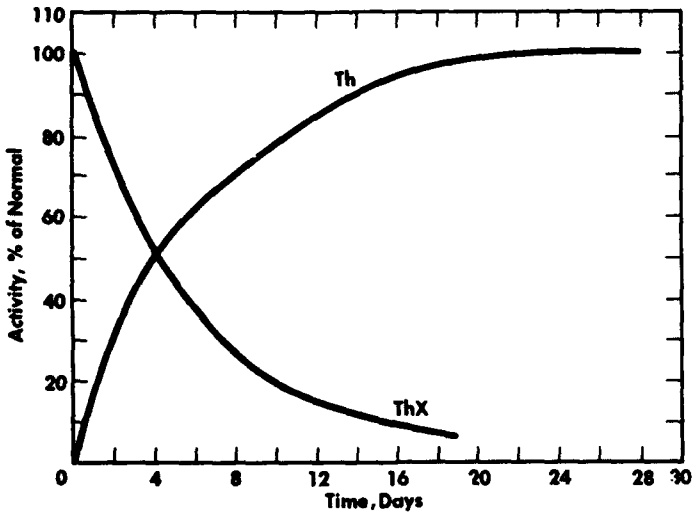


Fig. 2.3— Increase and decrease in activity of Th and ThX.

The recovery curve of U can be expressed as

$$A = A_0(1 - e^{-\lambda t}) \tag{2.2}$$

where A_0 = activity of U at zero time
A = activity of U at any time t

RADIOACTIVE DECAY AND HALF-LIFE

As a result of these relationships, Rutherford and Soddy[3] proposed the theory that the atoms of radioactive elements undergo spontaneous disintegration into atoms of a new element with the emission of alpha and beta particles. Furthermore, the intensity of the radioactivity is proportional to the number of atoms disintegrating at a given time. Replacing A_x, the relative radioactivity, with N, the number of atoms disintegrating, Eq. 2.1 becomes

$$N = N_0 e^{-\lambda t} \tag{2.3}$$

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where N_0 is the number of atoms at time zero. Differentiating

$$\frac{-dN}{dt} = \lambda N \tag{2.4}$$

$\frac{-dN}{dt}$ = rate of disintegration of atoms (negative because number of atoms decreases with time)

Taking logarithms of Eq. 2.3

$$\ln \frac{N_t}{N_0} = -\lambda t \tag{2.5}$$

where N_t is the number of atoms at time t.

As the number of atoms of the radioisotope is constantly decreasing with it becomes convenient to speak of the time necessary to reduce the amount of the radioisotope to half its original value, i.e., the time t*, such that $e^{-\lambda t} = \frac{1}{2}$. This time is called the half-life. Thus

$$\ln \tfrac{1}{2} = -\lambda t^*$$

or

$$\ln 2 = \lambda t^*$$

Substituting 0.693 for ln 2:

$$\text{Half-life} = t^* = \frac{0.693}{\lambda} \tag{2.6}$$

Thus, with elapsed time measured in half-lives, whose magnitude depends on the particular radioisotope, the relationship between relative activity and time is shown in Fig. 2.4, plotted on a linear graph and also on a semilogarithmic graph.

Equation 2.3 is the fundamental equation for the decay of a radioactive element. If the element is being produced from decay of a parent element as well as being removed by its own decay, Eq. 2.3 must be modified. Let B represent the number of atoms of the radioactive element produced per second.

The decay equation then becomes

$$\frac{dN}{dt} = B - \lambda N \tag{2.7}$$

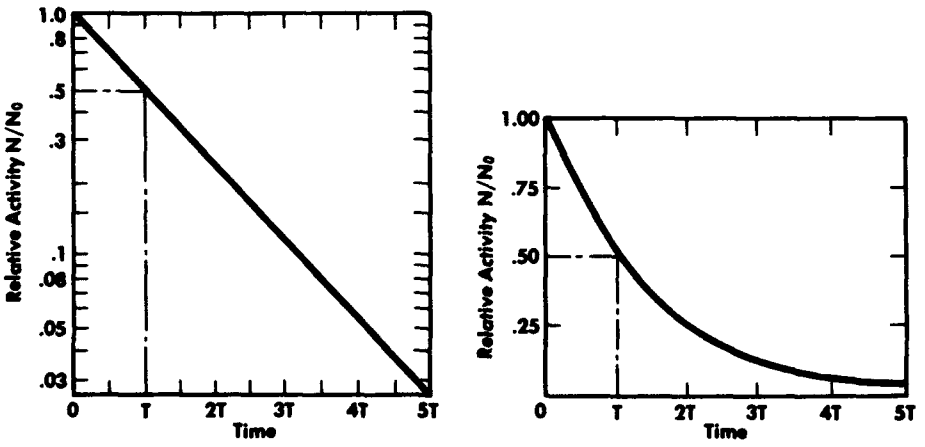


Fig. 2.4—Relation between relative activity and time.

where the first term on the right-hand side is the production term and the second term is the decay term. Rearranging Eq. 2.7

$$\frac{dN}{dt} + \lambda N = B \tag{2.8}$$

Equation 2.8 is solved by the use of an integrating factor and gives

$$Nt = e^{-\lambda t}\left[\frac{Be^{\lambda t}}{\lambda} + c\right] \tag{2.9}$$

If no atoms of the radioactive element are present at zero time, then $N_t = 0$ at $t = 0$, and

$$c = \frac{-B}{\lambda} \tag{2.10}$$

so

$$N_t = \frac{B}{\lambda}(1 - e^{-\lambda t}) \tag{2.11}$$

Defining N_∞ as the number of atoms present when $t = \infty$, we have

$$N_\infty = \frac{B}{\lambda} \tag{2.12}$$

or

$$N = N_\infty (1 - e^{-\lambda t}) \tag{2.13}$$

Equation 2.13 can be used to describe the growth of activity in U or Th fractions, as shown in Figs. 2.2 and 2.4 if all the UX and ThX are initially removed.

NATURAL RADIOACTIVE SERIES

Extensive physical and chemical research on the naturally occurring radioisotopes has established that most of these are members of one of three long radioactive series which run from either uranium, actinium, or thorium through progressive decay down to a stable isotope of lead. The three series are named after the isotope heading the series and are:

1. Thorium (4n) series
2. Uranium (4n + 2) series
3. Actinium (4n + 3) series

The mass number of any member of one of the series is given by the corresponding relationship, 4n, 4n + 2, or 4n + 3, where n is an integer. These three series of natural radioisotopes, thorium (4n), uranium (4n + 2), and actinium (4n + 3) are listed in Tables 2.1 to 2.3, respectively.[4]

As an example of the decay of one important natural radioisotope in the uranium (4n + 2) series, consider Ra^{226}. From Table 2.2, Ra^{226} has an atomic charge of 88 and a mass of 226 and decays by emitting an alpha particle (having a mass of 4 and a charge of 2) to become a gas, radon, with a charge of 86 and a mass of 222, i.e.,

$$\begin{aligned} {}_{88}Ra^{226} &\rightarrow {}_{86}Rn^{222} + {}_{2}He^{4} \\ 226 &= (4 \times 56) + 2 \\ 222 &= (4 \times 55) + 2 \end{aligned} \tag{2.14}$$

Table 2.1—THE THORIUM SERIES[4]

Radioactive species	Nuclide	Type of disintegration	Half-life	Disintegration constant, sec^{-1}	Particle energy, Mev
Thorium (Th)	$_{90}Th^{232}$	α	1.39×10^{10}y	1.58×10^{-18}	3.98
Mesothorium 1 (MsTh1)	$_{88}Ra^{228}$	β	6.7y	3.28×10^{-9}	0.04
Mesothorium 2 (MsTh2)	$_{89}Ac^{228}$	β	6.13h	3.14×10^{-5}	2.18
Radiothorium (RdTh)	$_{90}Th^{228}$	α	1.90y	1.16×10^{-8}	5.423
Thorium X (ThX)	$_{88}Ra^{224}$	α	3.64d	2.20×10^{-6}	5.681
Th emanation (Tn)	$_{86}Em^{220}$	α	54.5s	1.27×10^{-2}	6.282
Thorium A (ThA)	$_{84}Po^{216}$	α,β	0.16s	4.33	6.774
Thorium B (ThB)	$_{82}Pb^{212}$	β	10.6h	1.82×10^{-5}	0.58
Astatine 216 (At^{216})	$_{85}At^{216}$	α	3×10^{-4}s	2.3×10^{3}	7.79
Thorium C (ThC)	$_{83}Bi^{212}$	α,β	47m	2.46×10^{-4}	α: 6.086 β: 2.25
Thorium C′ (ThC′)	$_{84}Po^{212}$	α	3.0×10^{-7}s	2.31×10^{6}	8.776
Thorium C″ (ThC″)	$_{81}Tl^{208}$	β	2.1m	3.73×10^{-3}	1.79
Thorium D (ThD)	$_{82}Pb^{208}$	Stable			

Table 2.2—THE URANIUM SERIES[4]

Radioactive species	Nuclide	Type of disintegration	Half-life	Disintegration constant, sec^{-1}	Particle energy, Mev
Uranium I (UI)	$_{92}U^{238}$	α	4.50×10^{9}y	4.88×10^{-18}	4.20
Uranium X_1 (UX_1)	$_{90}Th^{234}$	β	24.1d	3.33×10^{-7}	0.20
Uranium X_2 (UX_2)	$_{91}Pa^{234}$	β	1.18m	9.77×10^{-3}	2.32
Uranium Z (UZ)	$_{91}Pa^{234}$	β	6.7h	2.88×10^{-5}	1.2
Uranium II (UII)	$_{92}U^{234}$	α	2.50×10^{5}y	8.80×10^{-14}	4.763
Ionium (Io)	$_{90}Th^{230}$	α	8.0×10^{4}y	2.75×10^{-13}	4.68
Radium (Ra)	$_{88}Ra^{226}$	α	1620y	1.36×10^{-11}	4.777
Ra Emanation (Rn)	$_{86}Em^{222}$	α	3.82d	2.10×10^{-6}	5.486
Radium A (RaA)	$_{84}Po^{218}$	α,β	3.05m	3.78×10^{-3}	α: 6.998 β: ?
Radium B (RaB)	$_{82}Pb^{214}$	β	26.8m	4.31×10^{-4}	0.7
Astatine-218 (At^{218})	$_{85}At^{218}$	α	1.5–2.0s	0.4	6.63
Radium C (RaC)	$_{83}Bi^{214}$	α,β	19.7m	5.86×10^{-4}	α: 5.51 β: 3.17
Radium C′ (RaC′)	$_{84}Po^{214}$	α	1.64×10^{-4}s	4.23×10^{3}	7.680
Radium C″ (RaC″)	$_{81}Tl^{210}$	β	1.32m	8.75×10^{-4}	1.9
Radium D (RaD)	$_{82}Pb^{210}$	β	22y	1.00×10^{-9}	0.018
Radium E (RaE)	$_{83}Bi^{210}$	β	5.0d	1.60×10^{-6}	1.17
Radium F (RaF)	$_{84}Po^{210}$	α	138.3d	5.80×10^{-8}	5.300
Thallium-206 (Tl^{206})	$_{81}Tl^{206}$	β	4.2m	2.75×10^{-3}	1.51
Radium G (RaG)	$_{82}Pb^{206}$	Stable			

Table 2.3— THE ACTINIUM SERIES[4]

Radioactive species	Nuclide	Type of disintegration	Half-life	Disintegration constant, sec^{-1}	Particle energy, Mev
Actinouranium (AcU)	$_{92}U^{235}$	α	7.10×10^{8}y	3.09×10^{-17}	4.58
Uranium Y (UY)	$_{90}Th^{231}$	β	24.6h	7.82×10^{-6}	0.30
Protoactinium (Pa)	$_{91}Pa^{231}$	α	3.43×10^{4}y	6.40×10^{-13}	5.042
Actinium (Ac)	$_{89}Ac^{227}$	α,β	22.0y	1.00×10^{-9}	α: 4.94 β: 0.04
Radioactinium (RdAc)	$_{90}Th^{227}$	α	18.6d	4.31×10^{-7}	6.05
Actinium K (AcK)	$_{87}Fr^{223}$	β	21m	5.50×10^{-4}	1.2
Actinium X (AcX)	$_{88}Ra^{223}$	α	11.2d	7.16×10^{-7}	5.75
Ac Emanation (An)	$_{86}Em^{219}$	α	3.92s	0.177	6.824
Actinium A (AcA)	$_{84}Po^{215}$	α,β	1.83×10^{-3}s	3.79×10^{2}	α: 7.365
Actinium B (AcB)	$_{82}Pb^{211}$	β	36.1m	3.20×10^{-4}	1.39
Astatine-215 (At^{215})	$_{85}At^{215}$	α	10^{-4}s	7×10^{3}	8.00
Actinium C (AcC)	$_{83}Bi^{211}$	α,β	2.16m	5.25×10^{-3}	α: 6.621
Actinium C′ (AcC′)	$_{84}Po^{211}$	α	0.52s	1.33	7.434
Actinium C″ (AcC″)	$_{81}Tl^{207}$	β	4.79m	2.41×10^{-3}	1.44
Actinium D (AcD)	$_{82}Pb^{207}$	Stable			

Most natural radioisotopes have such short half-lives in comparison with the more stable members of a series that they are only found in very small concentrations in the natural state. Thus, although Ra^{226} has a half-life of 1620 years, this half-life is so small compared to the half-life of 4.5×10^{9} years for U^{238} that the Curies were able to recover only $\frac{1}{10}$ g of impure radium chloride in extracting 1 ton of uranium ore. Thus, the natural radioisotopes, except those with extremely long half-lives, are found in such small concentrations that they are very expensive (about $100 per mg for radium).

NATURAL SOURCES OF RADIOISOTOPES AND FISSIONABLE MATERIALS

All the fission products and reactor-produced radioisotopes that might be used as sources of nuclear radiations must be derived either directly or indirectly from one of the two natural sources of fissionable materials, uranium or thorium. Uranium has been used as the principal reactor fuel because at the present stage of reactor technology, thorium must be converted to uranium before it can be used as a fuel. There also has been a limited use of plutonium made from U^{238} for this purpose. However, thorium has been converted to U^{233} in research experiments, and when large convertor or breeder reactors are put into operation for the production of nuclear power, appreciable quantities of U^{233} will be produced from the action of the surplus neutrons on the thorium reactor blanket. Unlike a convertor reactor, which produces less fuel than it consumes, a breeder reactor produces more fuel than it consumes.

ARTIFICIAL RADIOACTIVITY

The following paragraphs describe the production of artificial radioisotopes and discuss their properties.

DISCOVERY

Artificial radioactivity was discovered in 1934 by the Joliots[29] during their study of alpha-particle (α) bombardment of light elements. When boron, magnesium, and aluminum were bombarded with alpha particles, emissions of positrons (which had been identified 2 years be-

fore) were detected besides neutrons and protons. Emission of positrons occurs from artificial radioisotopes only. The nuclear reaction for $_5B^{10}(\alpha n)_6C^{13}$ is

$$_5B^{10} + _2He^4 \rightarrow _7N^{14} \rightarrow _7N^{13*} + _0n^1 \quad (2.15)$$

$$_7N^{13*} \rightarrow _6C^{13} + _{+1}e^0 \ (t^* = 14 \text{ min}) \quad (2.16)$$

where * represents an unstable nucleus and $_{+1}e^0$ represents a positron.

It was later found that radioactivity can be induced by bombarding an element with protons, deuterons, neutrons, and photons, as well as with alpha particles. About 1000 artificially radioactive nuclei have been identified. These nuclei lie either below or above the stability curve. Besides the emission of electrons, positrons, and photons, orbital electron capture has been reported.

TRANSURANIC ELEMENTS, DISCOVERY OF FISSION AND THE NEPTUNIUM SERIES

After the discovery of neutrons in 1932, Fermi[30] suggested the possibility of the production of elements greater than 92 in atomic number, termed the transuranic elements, by bombarding uranium by neutrons. A new era began with the discovery of nuclear fission[31] in 1939. Other reports of nuclear fission followed shortly thereafter.[32–34] By 1940 the existence of transuranic elements of atomic number 93 and 94 was proved beyond doubt.[35]

$$_{92}U^{238} + _0n^1 \rightarrow _{92}U^{239*} + \gamma \quad (2.17)$$

$$_{92}U^{239*} \rightarrow _{93}Np^{239*} + _{-1}e^0 \ (t^* = 23 \text{ min}) \quad (2.18)$$

$$_{93}Np^{239*} \rightarrow _{94}Pu^{239} + _{-1}e^0 \ (t^* = 2.3 \text{ days}) \quad (2.19)$$

$$_{94}Pu^{239} \rightarrow _2He^4 + _{92}U^{235} \ (t^* = 2400 \text{ years}) \quad (2.20)$$

These transuranic elements decay into a new artificial radioactive series. The series has been named the Neptunium Series (4n + 1) and is the missing link in the chain of series from 4n to 4n + 3. The members of the Neptunium Series are listed in Table 2.4.

Table 2.4—THE NEPTUNIUM SERIES[4]

Nuclide	Type of disintegration	Half-life	Disintegration constant, sec^{-1}	Particle energy, Mev
$_{94}Pu^{241}$	β	14y	1.57×10^{-9}	4.91
$_{95}Am^{241}$	α	470y	4.68×10^{-11}	5.546
$_{93}Np^{237}$	α	2.20×10^6y	9.99×10^{-15}	4.77
$_{91}Pa^{233}$	β	27.4d	2.93×10^{-7}	0.53
$_{92}U^{233}$	α	1.62×10^5y	1.36×10^{-13}	4.823
$_{90}Th^{229}$	α	7340y	3×10^{-12}	5.02
$_{88}Ra^{225}$	β	14.8d	5.42×10^{-7}	~0.3
$_{89}Ac^{225}$	α	10.0d	8.02×10^{-7}	5.80
$_{87}Fr^{221}$	α	4.8m	2.41×10^{-3}	6.30
$_{85}At^{217}$	α	0.018s	3.85×10^1	7.02
$_{83}Bi^{213}$	β, α	47m	2.46×10^{-4}	α: 5.86 β: 1.39
$_{84}Po^{213}$	α	4.2×10^{-6}s	1.65×10^5	8.336
$_{81}Th^{209}$	β	2.2m	5.25×10^{-3}	1.99
$_{82}Pb^{209}$	β	3.2h	6.02×10^{-5}	0.635
$_{83}Bi^{209}$	Stable			

THE BOHR COMPOUND-NUCLEUS THEORY

The compound-nucleus theory of Bohr[36] is useful in explaining nuclear reactions, induced radioactivity, and fission. Consider first a nucleus of a medium or a heavy atom that may be highly excited by interaction with nuclear radiation such as a neutron or proton. The amount of energy required for emission per nucleon varies with the nucleus but on the average is about 8 Mev. The nuclear reaction involved can be explained in three steps, according to Bohr's theory. In the first step, the nucleus is left in an excited state after interaction with nuclear radiation. This nuclear radiation might be neutron, proton, deuteron, alpha particle, or energetic gamma photon. In the second stage, energy of excitation becomes distributed throughout the nucleus and a compound nucleus is formed. The time required for the formation of the compound nucleus may involve many nuclear periods and is considered to be of the order of 10^{-15} sec. In the third stage a proton or neutron may be emitted if the excitation energy exceeds 8 Mev. The possibility of neutron emission is several hundred times that of proton emission because the potential barrier tends to retain the charged protons. If the excitation energy is high, more than 1 neutron or possibly a neutron and a proton or a deuteron may be emitted. The emission of nuclear radiations in the form of neutrons, protons, or deuterons occurs at certain energy levels, and the remaining energy is released as gamma radiation.

Fission may accompany such nuclear reactions. Some nuclei will fission spontaneously, but, in general, the fission reaction requires initiation by outside energy in the form of nuclear radiation. This energy is termed the activation energy, and in the case of fission, U^{235}, U^{233}, and Pu^{239}, capture of a slow neutron of only 0.025 ev energy is required.

THE BINDING-ENERGY CURVE

If two or more neutrons or protons are in close proximity, a force exists that tends to hold the particles together. The amount of energy necessary to disintegrate a cluster of neutrons and protons into widely separated particles is termed the binding energy of the nucleus or cluster. The Einstein[37] relation between binding energy per nucleon and mass number is shown in Fig. 2.5. The curve in Fig. 2.5 shows that the binding energy per nucleon is about 8 Mev on the central portion of the curve but is less for lighter and heavier nuclei. Any nuclear reaction

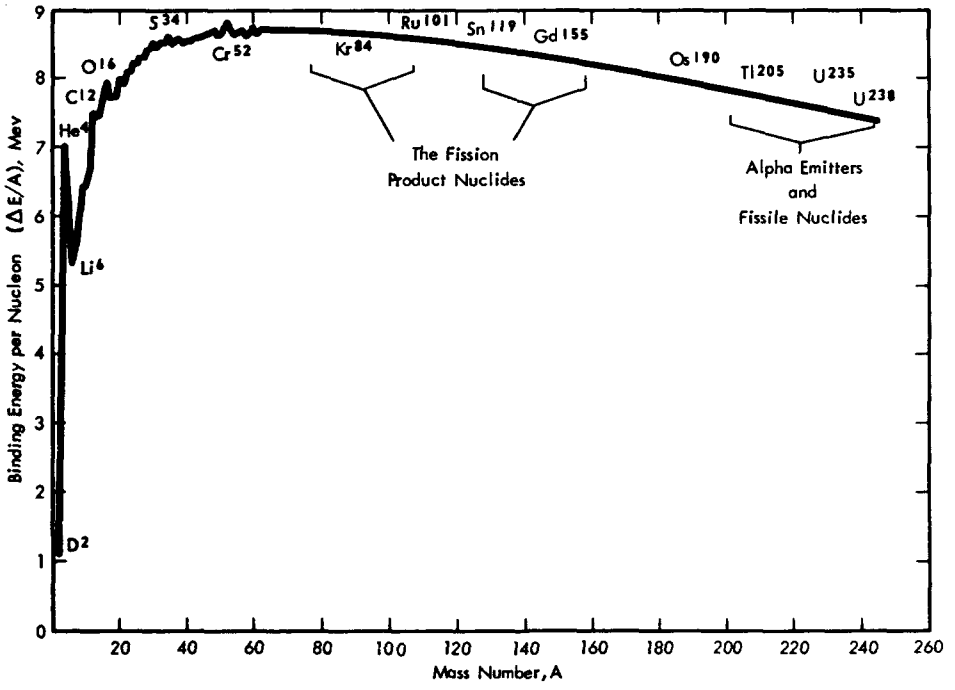


Fig. 2.5—Relation between binding energy per nucleon and mass number.

that shifts the nucleus from either end toward the central portion of the curve would release energy. The binding-energy curve can be used to predict the change in energy during a nuclear reaction. Four examples, fission, fusion, alpha emission, and beta decay are discussed with reference to the binding-energy relationship.

Fission

In the fission reaction a heavy nucleus splits into two lighter nuclei and energy is released. In this case the nuclear reaction involves a shift from the right end of the curve to the central portion. The difference in binding energy in these two positions appears as energy of fission.

Fusion

In the fusion reaction lighter nuclei fuse together to form a heavier nucleus. This reaction involves shifting from the left end of the curve to the central portion. The difference in binding energy in these two positions appears as energy of fusion.

Alpha-Particle Emission

If the daughter nucleus and the alpha particle emitted have a combined binding energy less than that of the parent nucleus, alpha decay is a possible natural process. If it occurs, the excess energy will be dissipated as kinetic energy of the alpha particles or possibly by the emission of one or more gamma photons.

Beta-Particle Emission

In the case of beta-particle emission the parent nucleus must have a binding energy in excess of that of the daughter nucleus by at least 0.51 Mev, the energy equivalent of the mass of one beta particle.

Binding energy can be expressed in terms of mass units as the difference between the mass of the same number of protons and neutrons and the actual mass of the atom under consideration. This is called the mass defect.

$$m = ZM_H + (A-Z)M_N - M({}_ZX^A) \tag{2.21}$$

where m = mass defect
Z = number of protons in the nucleus
A = mass number of the atom
$(A-Z)$ = number of neutrons in the nucleus
M_H = mass of proton (hydrogen nucleus)
M_N = mass of neutron
$M({}_ZX^A)$ = mass of atom of atomic number Z and mass number A

Einstein[37] has shown the following relationship between change in mass and change in energy

$$\Delta E = \Delta mc^2 \tag{2.22}$$

where ΔE = change in energy
Δm = change in mass
c = velocity of light

One mass unit is equivalent to 931 Mev. One Mev is the kinetic energy given to an electron when it passes through an electrical potential of 1,000,000 volts. It is equivalent to 1.6×10^{-12} erg and is the unit of energy also used for systems and particles other than electrons. The difference in mass can be measured by means of a mass spectrograph and is called the mass defect. The mass defect is the binding energy expressed in mass units.

PRODUCTION OF ARTIFICIAL RADIOISOTOPES

The nucleus of any element can be made radioactive by bombarding a small quantity of the element that will yield the desired isotope in a nuclear reactor or particle accelerator. The atoms of the element can be bombarded by a variety of particles in the accelerator or by neutrons and energetic gamma and beta radiations in a reactor.

In a nuclear reactor, radioisotopes are produced by a number of different processes.[38] The processes that produce appreciable quantities of radioisotopes are the n,γ, the n,p, and the n,α reactions, of which the n,γ is the most common and is caused by thermal neutron capture. In this process a neutron is captured by a target nucleus and a gamma photon is emitted. The neutron carries no charge and there is, therefore, no change in the atomic number. The element produced is a radioisotope of the original element in the target material. Since the radioisotope has the same chemical properties as the target material, it cannot be chemically separated in order to increase the specific activity. A typical example is the activation of sodium shown by the following equation

$$_{11}Na^{23} + {}_{0}n^{1} \rightarrow {}_{11}Na^{24} + \gamma \qquad (2.23)$$

Radioisotopes produced by the n,γ reaction may decay by beta-particle emission to a daughter product with a higher atomic number (nuclear charge). The daughter gains one charge and so increases 1 in atomic number for each beta particle emitted but keeps the same mass number. In this case, the daughter product can be chemically separated from the target material to increase the specific activity. Equations 2.24 and 2.25 illustrate these reactions.

$$_{32}Ge^{76} + {}_{0}n^{1} \rightarrow {}_{32}Ge^{77} + \gamma \qquad (2.24)$$

$$_{32}Ge^{77} \xrightarrow{12\ hr} {}_{33}As^{77} + \beta^{-} \qquad (2.25)$$

The n,p process requires neutrons with energies higher than that of thermal neutrons and in this process a neutron enters the nucleus of the target element with sufficient energy to eject a proton. This reduces the atomic number by 1, thereby producing a new element (transmutation), which can be chemically separated from the target element and permits the production of radioisotopes of high specific activity. The production of radioactive phosphorus from sulfur is an example of this process.

$$_{16}S^{32} + {}_{0}n^{1} \rightarrow {}_{15}P^{32} + {}_{1}p^{1} \qquad (2.26)$$

Another reaction that requires high-energy neutrons is the n,α reaction. In this process a neutron of very high energy enters the nucleus of a target atom with sufficient energy to eject an alpha particle. The atomic number is changed and, as in the previous case, a chemical separation is possible, permitting an increase in the specific activity. A typical example of this reaction is the production of radioactive sodium from aluminum.

$$_{13}Al^{27} + {}_{0}n^{1} \rightarrow {}_{11}Na^{24} + {}_{2}He^{4} \qquad (2.27)$$

Radioisotopes are also produced as a result of fission. The atomic numbers of most of these fission products range from 30 to 64. The fission products of different atomic number can be separated chemically and concentrated to high activity. Since a number of isotopes of each element may be present, the isotopic purity and activity are frequently not as high as those of radioisotopes produced by either the n,p or the n,α reactions.

By the procedure described it is possible to produce selected radioisotopes with desired chemical properties having a long half-life and low activity or having a relatively short half-life and intense radioactivity. About 800 different radioisotopes have been produced by this technique but many of these have limited usefulness because of an inconveniently short half-life, undesirable chemical properties, or limited availability of the parent isotope. However, about 150 are sufficiently useful and stable to be kept in stock for sale by the Isotopes Division at Oak

Ridge National Laboratory and comparable organizations in England, France, Canada, and the U.S.S.R. The more important radioisotopes available from Oak Ridge are listed in Table 2.5 according to half-life.[38]

Between 1946 and the autumn of 1957, the USAEC made more than 90,000 shipments of radioisotopes from Oak Ridge National Laboratory alone, as listed in Table 2.6. In 1955 radioisotopes were used in about 2700 United States institutions and laboratories. Also, about 4000 shipments were made to about 660 institutions and laboratories in 46 countries abroad.[39]

Figure 2.6 shows a photograph and Fig. 2.7 a cutaway view of the Oak Ridge National Laboratory air-cooled graphite reactor where most of these radioisotopes are produced.

With reference to Table 2.6, the top five radioisotopes are both beta-particle and gamma emitters. These radioisotopes are used chiefly as gamma sources in radiation therapy, medical diagnosis, or as gamma sources for research. Polonium-210 is at present the most widely used alpha-emitting radioisotope. The next four radioisotopes are pure beta-particle emitters and have been used widely in tracer studies and in therapy. The pure beta-particle emitters considered most important by Aebersold[39] are listed with their properties in Table 2.7. A similar list of the more important beta-particle and gamma-ray emitters is given in Table 2.8.

A radioisotope may be shipped as a solid, liquid, or gas, depending on its natural state and the requirements of the user. If a large quantity of the radioisotope is to be used as a radiation source, it must be suitably contained and shielded and yet accessible for use as a source. In the case of a beta-particle source such as Sr^{90}, one portion of the container must consist of a window thin enough to transmit most of the beta particles. A metal container about $\frac{1}{8}$-in. thick will completely stop the beta particles, allowing only the secondary (Bremsstrahlung) radiation to escape. Figure 2.8 shows a design of a container for a Sr^{90} beta-particle source with a 2-mil stainless-steel window as the far end. The source itself consists of Sr^{90} fluoride ($Sr^{90}F_2$) fused at 1200°C to a platinum cup. In the case of gamma sources, the thickness of the window is not as significant. Figure 2.9 shows a design for a Co^{60} teletherapy source. High-specific-activity Co^{60} metal wafers about the size of a penny can be stacked in such a container to provide a very powerful "point" source such as is desired in most teletherapy applications. Similar, but not quite as compact, "point" gamma sources can be prepared by pelleting $Cs^{137}Cl$ into stainless-steel tubes, as shown in Fig. 2.10.

Point gamma sources are not always desired and in many research applications a cylindrical source has advantages.[41] Figure 2.11 shows a photograph of the 3000-curie (activity in 1952) Co^{60} source at the Fission Products Laboratory, The University of Michigan.[41–44] This source consists of 100 rods of cobalt, 10 in. long and $\frac{1}{4}$ in. in diameter, each enclosed in 3 S aluminum pipe. The 100 rods are held in a 3 S aluminum holder in two concentric rings so as to produce a cylindrical source with a nearly uniform radiation field in the center of the cylinder.

Separated radioisotopes that do not require extensive shielding are shipped in disposable containers. A sketch of one of the standard types of disposable containers is shown in Fig. 2.12.

If the radioisotope shipment requires shielding to an extent not possible in a disposable container, a returnable container of the wooden-box type is used.[38] For larger quantities of gamma emitters, the isotopes may be shipped in a lead container with a stainless-steel jacket and two steel handles, as shown in Fig. 2.13.

Kilocurie amounts of Co^{60} or Cs^{137} require several inches of lead shielding and the shipping containers may weigh a number of tons. A 3-ton lead container with a steel shell was used to ship 3000 curies of Co^{60} from the NRX reactor in Canada to the Fission Products Laboratory, The University of Michigan.[42]

ALPHA-PARTICLE EMISSION

Alpha particles are simply fast moving helium nuclei. Heavy nuclei are more likely to decay by alpha-particle emission than lighter nuclei. Figure 2.14 and Tables 2.1 through 2.4 show that the region of possible alpha-particle emitters includes nuclides with atomic number of 84 and greater.[45]

Some alpha emitters, such as radium, occur naturally. Natural radioisotopes that are alpha emitters usually have very long half-lives, which explains why they are still found in the

Table 2.5—SOME RADIOISOTOPES AVAILABLE FROM OAK RIDGE NATIONAL LABORATORY, LISTED ACCORDING TO HALF-LIFE[38]

Half-life	Max.* energy, Mev	Radioisotope†	Radiation
24.2s	1.516	Ag^{110}	β, γ
30s	1.045	$Rh^{106}(Ru^{106})$	β, γ
72s	1.30	In^{114}	EC,‡ β^-, β^+, γ
2.6m	0.662	$Ba^{137m}(Cs^{137})$	IT,§ γ
17.5m	0.134	$Pr^{144}(Ce^{144})$	β, γ
12.44h	1.51	K^{42}	β, γ
12.8h	1.34	Cu^{64}	EC, β^-, β^+, γ
13.6h	0.961	Pd^{109}	β, γ
14.3h	2.51	Ga^{72}	β, γ
15.06h	2.754	Na^{24}	β, γ
19h	2.1	Ir^{194}	β, γ
19.2h	1.59	Pr^{142}	β, γ
23h	0.164	$Hg^{194}m_2$	EC, IT, γ
24.1h	0.78	W^{187}	β, γ
26.8h	1.7	As^{76}	β, γ
35.87h	1.312	Br^{82}	β, γ
38h	0.524	As^{77}	β, γ
40h	2.50	La^{140}	β, γ
47h	0.548	Sm^{153}	β, γ
53h	0.525	Cd^{115}	β, γ
2.54d	2.18	Y^{90}	β
2.69d	0.411	Au^{198}	β, γ
2.71d	0.191	Hg^{197}	EC, γ
2.79d	0.780	Mo^{99}	β, γ
2.8d	1.9	Sb^{122}	β, γ
2.8d	0.217	Ru^{97}	EC, γ
3.15d	0.209	Au^{199}	β, γ
3.87d	0.764	Re^{186}	EC, β, γ
5.02d	1.17	Bi^{210}	β
7.6d	1.04	Ag^{111}	β, γ
8.08d	0.637	I^{131}	β, γ
11.3d	0.510	Nd^{147}	β, γ
12d	0.494	Ba^{131}	EC, γ
13.7d	0.932	Pr^{143}	β
14.3d	1.701	P^{32}	β
16d	0.129	Os^{191}	β, γ
19.5d	1.08	Rb^{86}	β, γ
27.8d	0.32	Cr^{51}	EC, γ
32.5d	0.145	Ce^{141}	β, γ
35d	0.0026	A^{37}	EC
35d	0.745	Nb^{95}	β, γ
39.8d	0.498	Ru^{103}	β, γ
43d	1.30	Cd^{115m}	β, γ
45d	0.611	Hf^{181}	β, γ
45.1d	1.289	Fe^{59}	β, γ
45.4d	0.279	Hg^{203}	β, γ
49d	0.190	In^{114m}	IT, γ
53d	1.463	Sr^{89}	β
60d	2.04	Sb^{124}	β, γ
61d	1.537	Y^{91}	β, γ
65d	0.754	Zr^{95}	β, γ
72d	0.81	Co^{58}	EC, β^+, γ

(Continued)

Table 2.5—(Continued)

Half-life	Max.* energy, Mev	Radioisotope†	Radiation
73.2d	0.428	W^{185}	β
74.37d	0.613	Ir^{192}	EC, β, γ
85d	1.12	Sc^{46}	β, γ
87.1d	0.167	S^{35}	β
112d	0.393	Sn^{113}	EC, γ
115d	1.223	Ta^{182}	β, γ
127d	0.405	Se^{75}	EC, γ
163d	0.254	Ca^{45}	β
250d	1.12	Zn^{65}	EC, β^+, γ
270d	1.516	Ag^{110m}	IT, β, γ
282d	0.134	Ce^{144}	β, γ
1.0y	2.41	Ru^{106}	β
2.3y	0.794	Cs^{134}	β, γ
2.6y	0.223	Pm^{147}	β
2.7y	0.637	Sb^{125}	β, γ
2.94y	0.0059	Fe^{55}	EC
4.0y	0.765	Tl^{204}	EC, β
5.27y	1.33	Co^{60}	β, γ
9.5y	0.357	Ba^{133}	EC, γ
10.27y	0.54	Kr^{85}	β, γ
12.46y	0.01795	H^3	β
13y	1.40	Eu^{152}	EC, β, γ
16y	1.40	Eu^{154}	β, γ
25y	2.18	Sr^{90}	β
30y	0.662	Cs^{137}	β
85y	0.067	Ni^{63}	β
5568y	0.155	C^{14}	β
2.12×10^5y	0.29	Tc^{99}	β
3.08×10^5y	0.714	Cl^{36}	β
1.72×10^7y	0.039	I^{129}	β, γ

*Max/energy (5 per cent or more) for β, γ emitters.

†The radioisotope in parentheses is the one under which the listed radioisotope is cataloged.

‡EC, electron capture (K capture) process.

§IT, isomeric transition.

Table 2.6—SHIPMENTS AND CURIES OF PRINCIPAL RADIOISOTOPES IN 11-YEAR PERIOD (Aug. 2, 1946–Nov. 30, 1957, from ORNL Only)[40]

Isotope	Shipments	Curies
C^{14}	2,658	50
Co^{60}	1,429	190,007
Cs^{137}	763	5,272
I^{131}	33,320	3,975
Ir^{192}	318	10,481
P^{32}	19,410	1,083
$Sr^{89,90}$	1,084	416
H^3	451	6,948
Others	17,649	965
Irradiated units	13,316	
Total	90,398	219,197

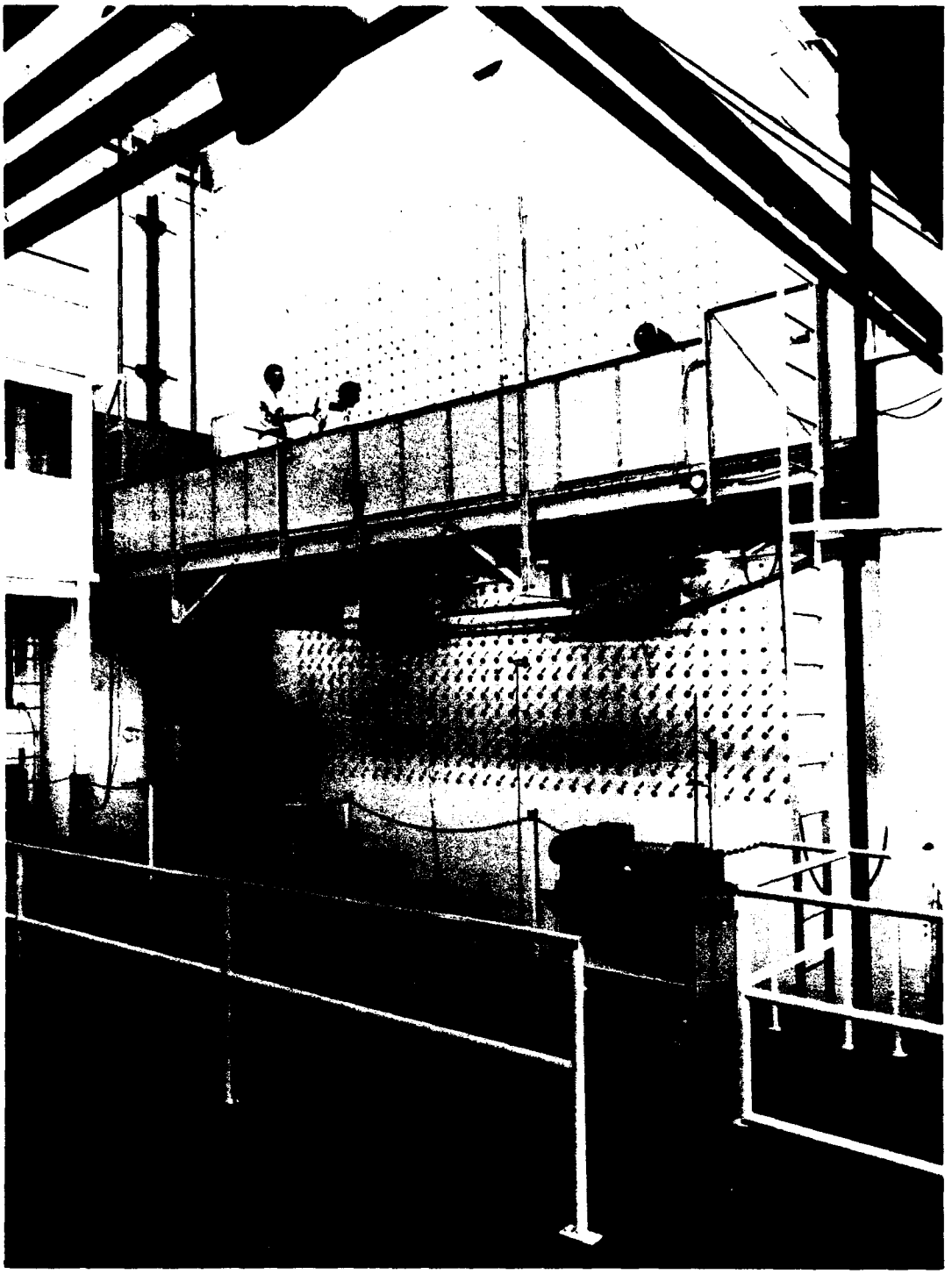


Fig. 2.6 — Photograph of the Oak Ridge National Laboratory air-cooled graphite reactor.

crust of the earth. One method of calculating the age of the earth is by determining the extent of decay of these naturally occurring radioisotopes, knowing their half-lives.

An alpha particle may be ejected from the nucleus as a result of a specific nuclear transition and usually has an energy in the range of 3 to 10 Mev. Two simple laboratory experiments demonstrate that alpha particles are identical with helium nuclei.

In the first experiment the total number of alpha particles ejected by a radioactive source is determined by observing the rate at which these particles strike a fluorescent screen of known geometry. If these same particles are allowed to strike a conductor leading to an elec-

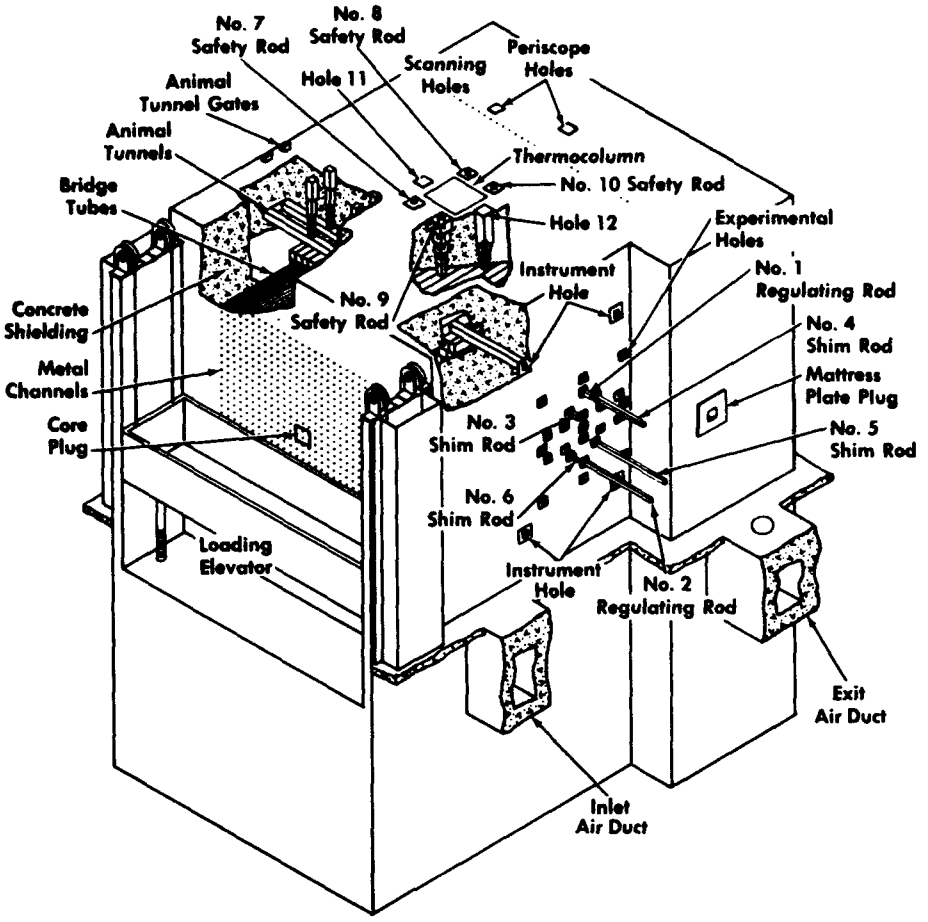


Fig. 2.7 — A cutaway view of the Oak Ridge National Laboratory air-cooled graphite reactor.

Table 2.7 — IMPORTANT REACTOR-PRODUCED PURE BETA-PARTICLE EMITTING RADIOISOTOPES[39]

Element	Isotope	Half-life	Energy of radiation, Mev
Hydrogen	H^3	12.5y	0.019
Carbon	C^{14}	5700y	0.155
Phosphorus	P^{32}	14.3d	1.712
Sulfur	S^{35}	87.1d	0.166
Calcium	Ca^{45}	164d	0.25
Strontium	Sr^{90}	28y	2.24*, 0.54
Thallium	Tl^{204}	4.0y	0.77

*From daughter Y^{90}.

troscope, the total charge collected may be determined. Experiments of this type have shown that the alpha particle carries a positive electric charge equivalent in magnitude to that of two electrons.[46,47]

In the second experiment these particles are deflected in a magnetic field. When a charged particle moves in a magnetic field, the radius of its circular trajectory can be obtained by equating magnetic and centrifugal forces, from which

$$Hqv = \frac{mv^2}{R} \qquad (2.28)$$

Table 2.8—IMPORTANT REACTOR-PRODUCED BETA-PARTICLE AND GAMMA-RAY EMITTING RADIOISOTOPES[39]

Element	Isotope	Half-life	Energy of radiations, Mev	
			Beta	Gamma
Sodium	Na^{24}	14.9h	1.39	2.76, 138
Iron	Fe^{55}	2.91y	None	K (6.2 kev X-rays)
Iron	Fe^{59}	46.3d	0.46, 0.27 1.56	1.10, 1.29, 0.19
Cobalt	Co^{60}	5.2y	0.32	1.33, 1.17
Iodine	I^{131}	8.05d	0.61, 0.34, 0.25, 0.81	0.36, 0.28, 0.64, 0.08
Cesium	Cs^{137}	33y	0.518, 1.18	0.662*
Thulium	Tm^{170}	127d	0.97, 0.88	0.084
Iridium	Ir^{192}	74.5d	0.67, 0.54, 0.24	0.32, 0.31, 0.30, 0.20
Gold	Au^{198}	2.69d	0.97	0.411

*From daughter Ba^{137}.
Note: Radiations are listed in order of abundance.

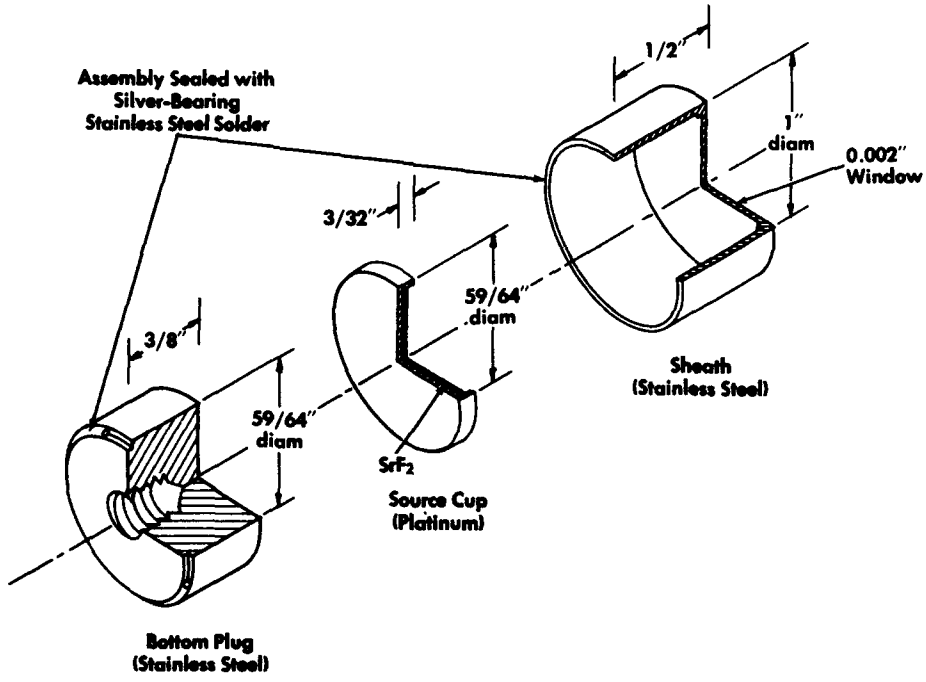


Fig. 2.8—Design of a container for a Sr^{90} beta-particle source with a 2-mil stainless-steel window.

where H = magnetic field strength
q = charge of particle
m = mass of particle
v = velocity of particle
R = radius of curved path

The velocity of the particle may be determined by sending the beam of particles through a collimator which is in both an electric and magnetic field,[48] as shown in Fig. 2.15. The magnetic field is vertically upward out of the paper in this figure.

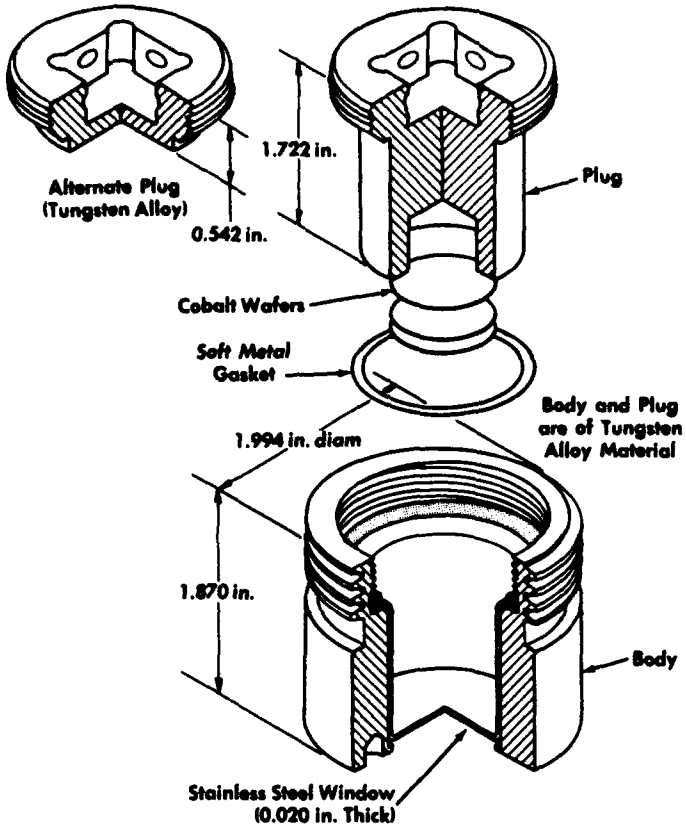


Fig. 2.9—Design for a Co^{60} teletherapy source.

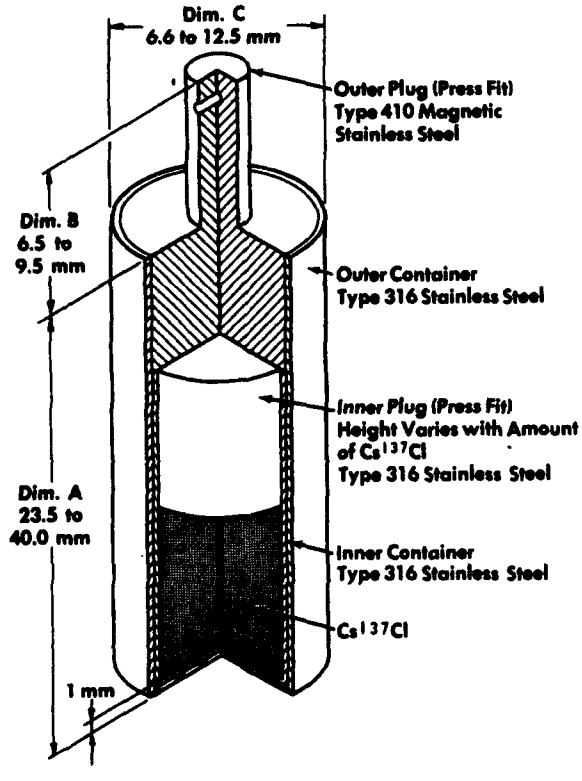


Fig. 2.10—Gamma sources prepared by pelleting $Cs^{137}Cl$ into stainless-steel tubes.

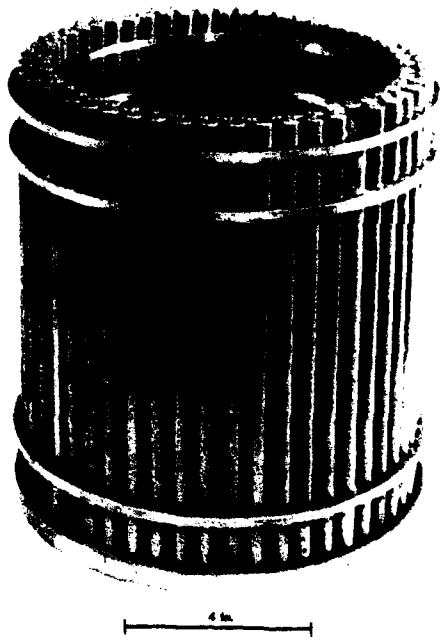


Fig. 2.11 — Photograph of the 3000-curie Co^{60} source at the Fission Products Laboratory, University of Michigan.

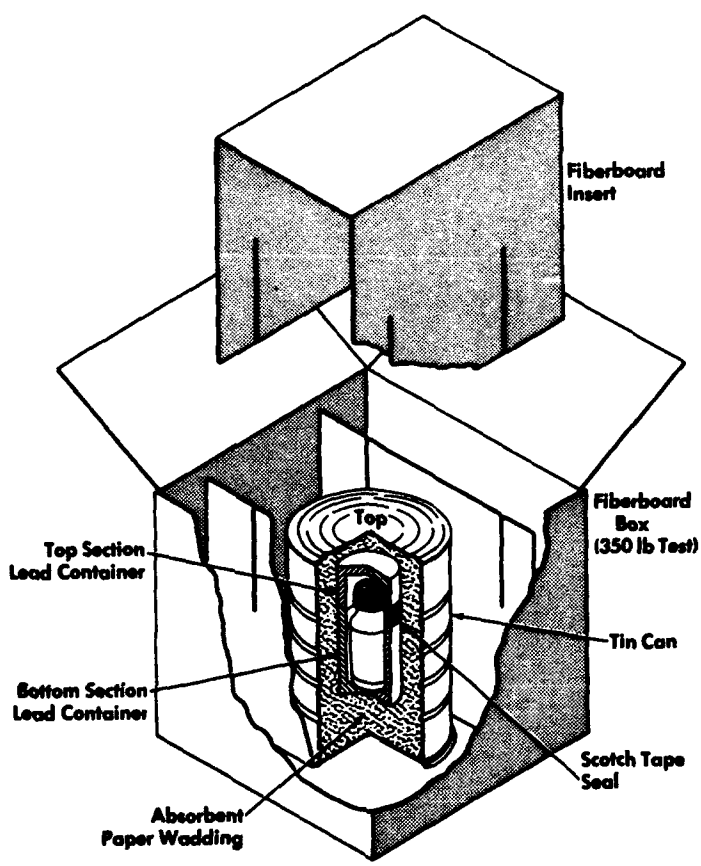


Fig. 2.12 — Sketch of one of the standard types of disposable containers.

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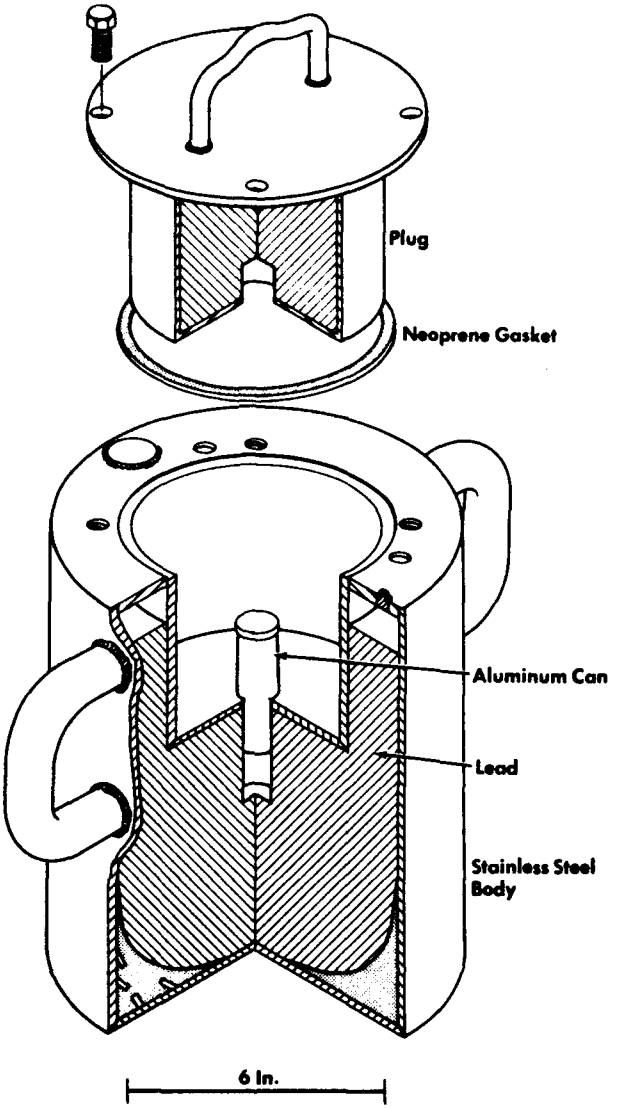


Fig. 2.13— Lead container with a stainless-steel jacket and two steel handles for shipping isotopes.

Particles proceeding through the collimator indicated in Fig. 2.15 are subjected to balancing electric and magnetic forces such that

$$Eq = Hqv \tag{2.29}$$

$$v = \frac{R}{H} \tag{2.30}$$

From Eq. 2.29 the quantity q/m can be calculated from the experimental measurements

$$q/m = \frac{v}{HR} \tag{2.31}$$

Knowing the charge and the charge-to-mass ratio, the mass of the alpha particle is easily calculated and has been shown to be equal to that of the helium nucleus.[49]

Further proof of the nature of alpha-particle radiation may be obtained by directing the particles into an evacuated chamber through very thin glass walls. Under such conditions the specific build-up of helium gas within the chamber can be spectrographically determined.

Alpha particles are evidently helium nuclei and become ordinary helium atoms by acquiring two planetary electrons.

Table 2.9 lists some of the alpha-emitting radioisotopes.[4] Polonium-210 is often used in research experiments on alpha particles. Plutonium-239 is fissionable and may be used as a reactor fuel as an alternative to U^{235}. These and other alpha-particle emitters are particularly toxic if allowed to enter the body.

ABSORPTION OF ALPHA-PARTICLE RADIATION

Alpha particles are very easily absorbed and those commonly encountered in work with radioisotopes can, in general, be totally absorbed by a sheet of heavy paper, a few mils of aluminum, or a few centimeters of air. Ranges in air for different alpha particles[4] are given in Table 2.9.

As the thickness of absorbing material interposed between the source and a detector, such as a scintillation screen, is increased, the number of observed counts is seen to remain approximately constant until a critical thickness, the range, is reached. At that point, the count falls rapidly to zero, as shown in Fig. 2.16.

If the range of alpha particles in absorbing material is plotted against energy, where energy is determined by magnetic deflection measurements, a range vs. energy relationship may be determined.[50] A plot of alpha-particle energy vs. range in air at 760 mm, 15°C is given in Fig. 2.17. When such a relationship has been established, energy determinations may be made by measuring the range.

Figure 2.16 shows that nearly all alpha particles from a given radioactive substance penetrate the same thicknesses of air. The slight differences in penetration are due partly to the statistical nature of energy absorption and partly to changes in direction experienced by some of the particles near the end of their paths, which prevent their reaching the detector. In addition to these phenomena, however, careful experiments have revealed that normal alpha particles emitted by some nuclides fall into several closely spaced energy groups.[51] These closely spaced "lines" are referred to as "fine structure" and have led to considerable advances in the knowledge of nuclear structure.

The range of alpha particles in most gases is of the order of several centimeters: in liquids and solids, very much shorter. The degree of "stopping power" is commonly used as a measure of the efficiency of various solid materials as alpha absorbers. The stopping power of an absorbing medium is defined as the number of centimeters of air required to absorb as much alpha radiation as 1 cm of the medium. Translating this concept to the atomic level, one can define the "atomic stopping power" as the number of atoms of air equivalent to one atom of the absorber. As alpha particles lose energy by interaction with electron fields, the atomic stopping power increases with the atomic number of the absorber, as shown in Fig. 2.18.

An empirical relation between the alpha-particle range and the disintegration constant was first stated as the Geiger-Nuttall rule.[52]

$$\log \lambda = A \log R + B \qquad (2.32)$$

where λ = decay constant

R = range of alpha particles

A, B = constants

A plot of $\log \lambda$ vs. $\log R$ yields straight lines for the three naturally radioactive series. The slopes are approximately equal, the intercepts somewhat different. In the early days, one of the puzzling aspects of alpha decay was why there should be this dependence of half-life on particle energy.

Another anomaly came to light with the comparison of emission and scattering data for the same nucleus. In the case of U^{238}, the fastest natural alpha particles available (nearly 9 Mev from ThC′) were not able to penetrate the potential barrier owing to coulomb repulsion by the nucleus. On the other hand, the same nuclide emits an alpha particle of only 4 Mev.

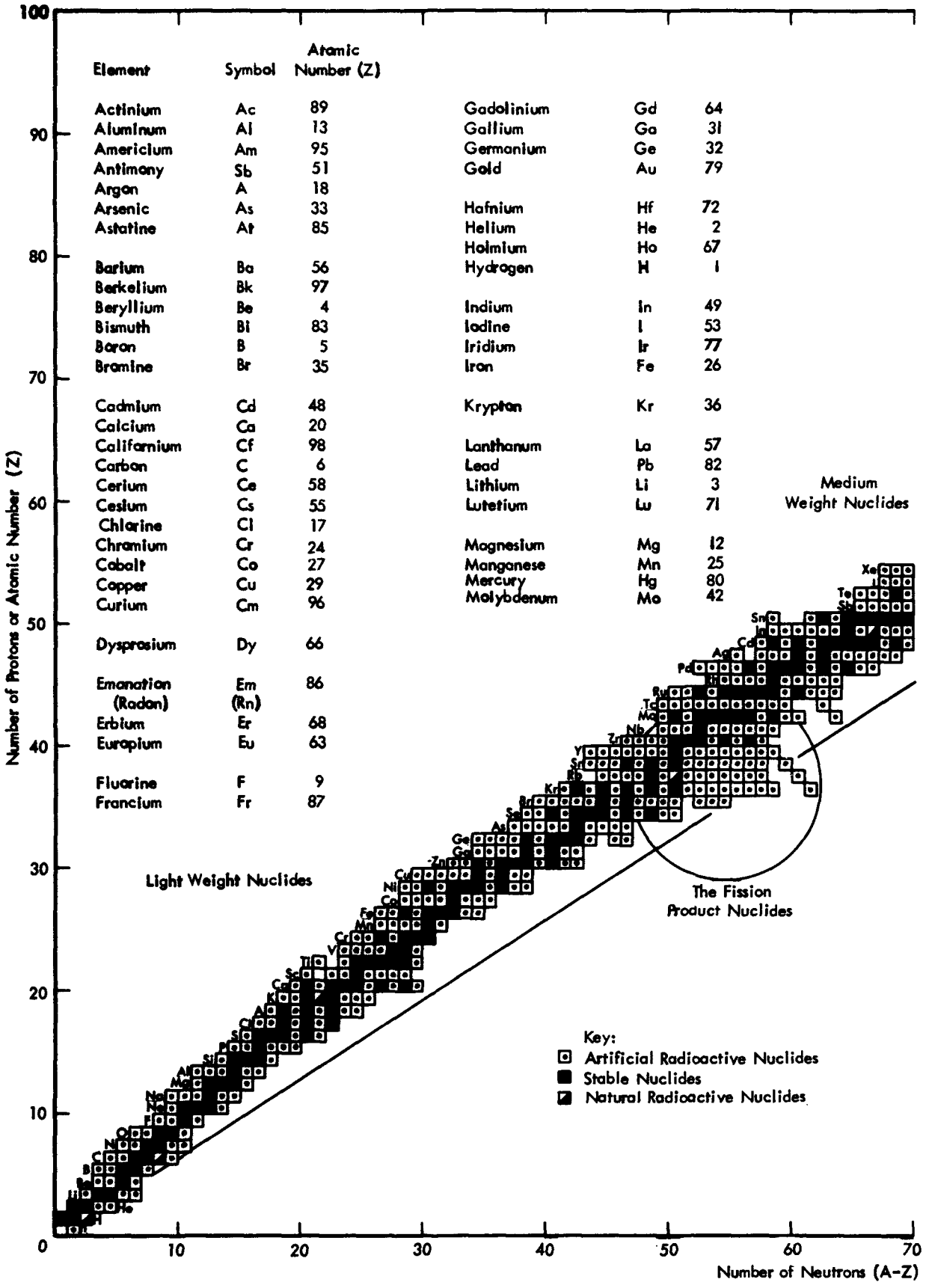
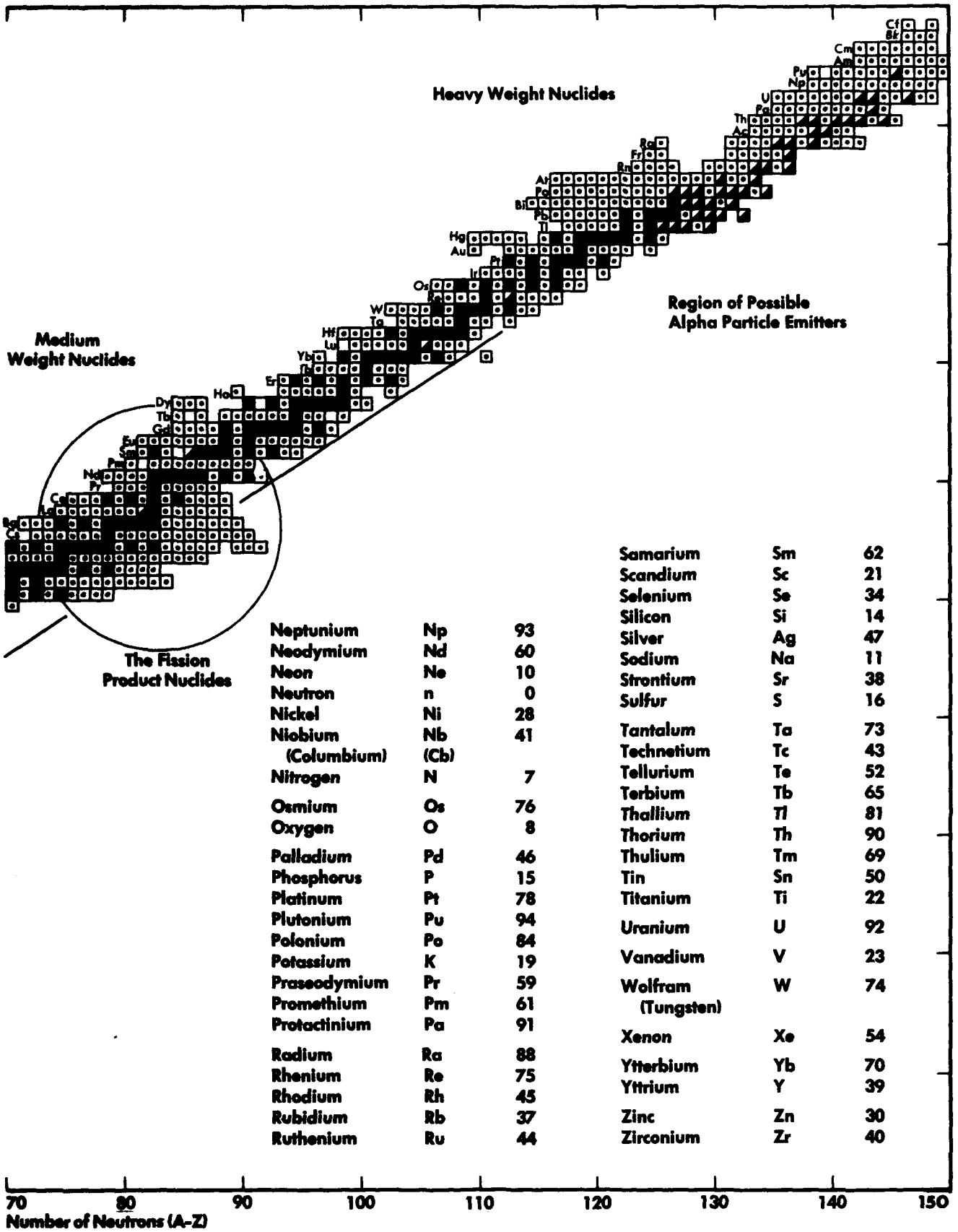


Fig. 2.14—The region of possible alpha-particle emitters which includes nuclides with atomic number of 84 and greater.



Neptunium	Np	93
Neodymium	Nd	60
Neon	Ne	10
Neutron	n	0
Nickel	Ni	28
Niobium (Columbium)	Nb (Cb)	41
Nitrogen	N	7
Osmium	Os	76
Oxygen	O	8
Palladium	Pd	46
Phosphorus	P	15
Platinum	Pt	78
Plutonium	Pu	94
Polonium	Po	84
Potassium	K	19
Praseodymium	Pr	59
Promethium	Pm	61
Protactinium	Pa	91
Radium	Ra	88
Rhenium	Re	75
Rhodium	Rh	45
Rubidium	Rb	37
Ruthenium	Ru	44
Samarium	Sm	62
Scandium	Sc	21
Selenium	Se	34
Silicon	Si	14
Silver	Ag	47
Sodium	Na	11
Strontium	Sr	38
Sulfur	S	16
Tantalum	Ta	73
Technetium	Tc	43
Tellurium	Te	52
Terbium	Tb	65
Thallium	Tl	81
Thorium	Th	90
Thulium	Tm	69
Tin	Sn	50
Titanium	Ti	22
Uranium	U	92
Vanadium	V	23
Wolfram (Tungsten)	W	74
Xenon	Xe	54
Ytterbium	Yb	70
Yttrium	Y	39
Zinc	Zn	30
Zirconium	Zr	40

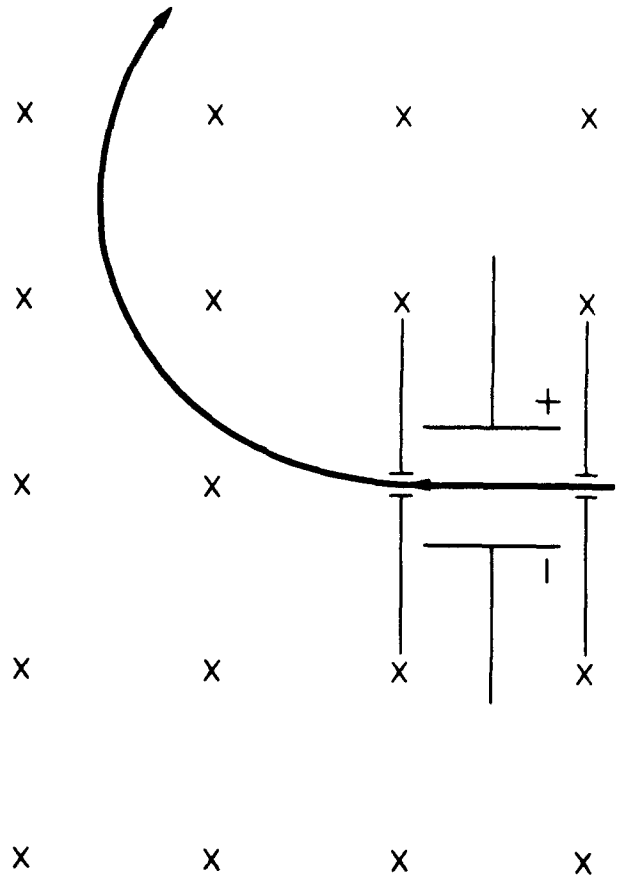


Fig. 2.15 — Method of determining particle velocity by sending a beam through a collimator which is in both an electric and magnetic field.

Table 2.9 — RANGES AND ENERGIES OF ALPHA PARTICLES EMITTED FROM SOME NATURAL RADIOISOTOPES[4]

Radionuclide	Mean range in air, cm	Energy, Mev
$_{86}Em^{219}$(An)	5.240 ± 0.015	6.559 ± 0.014
	5.692 ± 0.015	6.824 ± 0.014
$_{84}Po^{215}$(AcA)	6.457 ± 0.008	7.365
$_{83}Bi^{211}$(AcC)	4.984 ± 0.015	6.273 ± 0.012
	5.429 ± 0.015	6.619 ± 0.013
$_{84}Po^{211}$(AcC′)	6.555 ± 0.015	7.434
$_{86}Em^{220}$(Tn)	5.004 ± 0.018	6.2818 ± 0.0008
$_{84}Po^{216}$(ThA)	5.638 ± 0.008	6.7744 ± 0.0008
$_{83}Bi^{212}$(ThC)	4.730 ± 0.008	6.0537 ± 0.0007
$_{84}Po^{212}$(ThC′)	8.570 ± 0.007	8.7759 ± 0.0009
	9.724 ± 0.008	9.488 ± 0.004
	11.580 ± 0.008	10.538 ± 0.004
$_{86}Em^{222}$(Rn)	4.051 ± 0.008	5.4860 ± 0.0005
$_{84}Po^{218}$(RaA)	4.657 ± 0.008	5.9981 ± 0.0005
$_{84}Po^{214}$(RaC′)	6.907 ± 0.006	7.680 ± 0.0006
	7.793 ± 0.015	8.277 ± 0.003
	9.04 ± 0.02	9.066 ± 0.004
	11.51 ± 0.02	10.505 ± 0.004
$_{84}Po^{210}$(RaF)	3.842 ± 0.006	5.2984 ± 0.0021

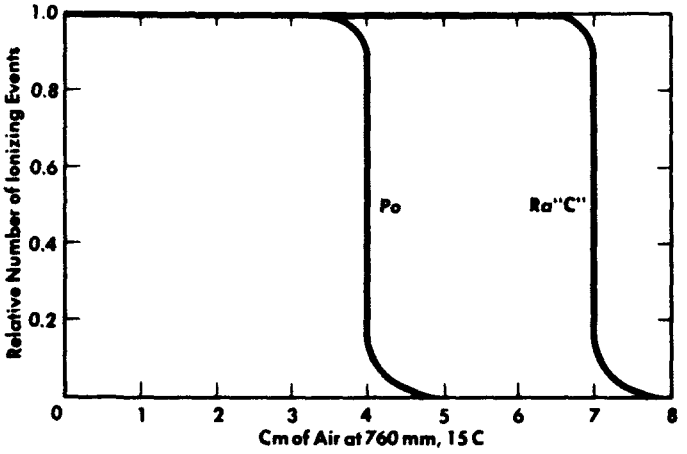


Fig. 2.16—Diagram showing decrease of alpha particle count as thickness of absorbing material increases.

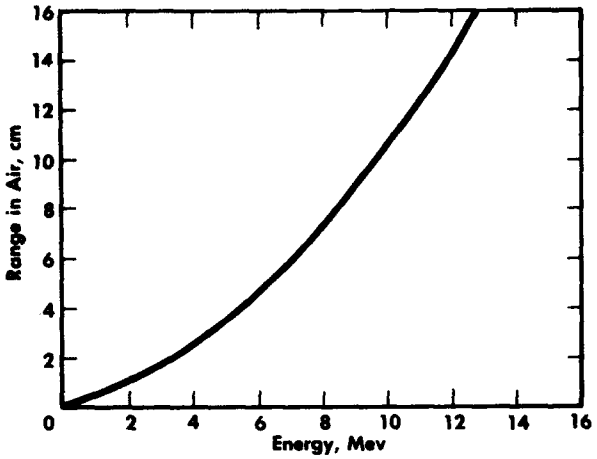


Fig. 2.17—A plot of alpha-particle energy vs. range in air at 760 mm, 15°C.

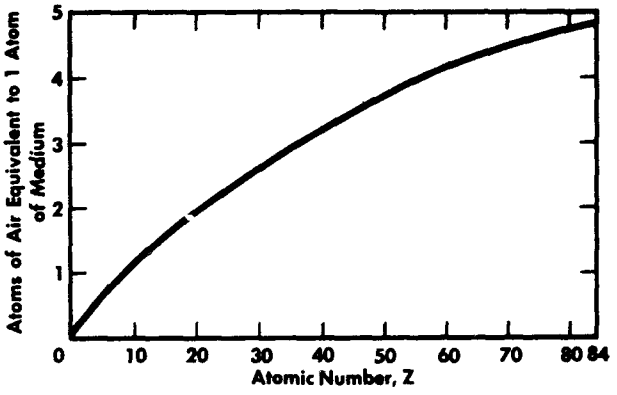


Fig. 2.18—Relation of the increase of atomic stopping power to the atomic number of the absorber.

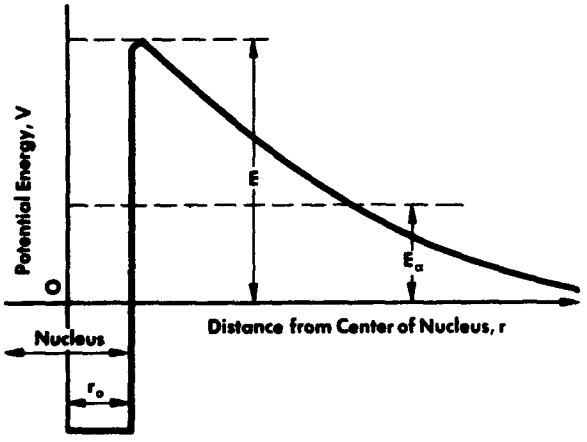


Fig. 2.19—Variation of particle energy with distance from center of nucleus.

As positively charged particles are known to exist within the nucleus, powerful short range attraction forces must exist within the nucleus. The resulting potential must vary with distance somewhat as shown in Fig. 2.19.

If the particles are thought of in classical terms as billiard balls and this potential replaced for the moment by gravitational potential, we can see that, to roll an incident particle into the nucleus, it must have an energy which will take it over the hump. Thus the maximum in the potential curve, E, can in principle be determined by finding the energy required for an incident particle to penetrate into the nucleus. However, classical physics has no explanation of the reason why an alpha particle of energy E, less than this maximum, can escape from the nucleus as it does in alpha emission.

The advent of wave mechanical methods offered the first solution of the paradox. Solution of the Schroedinger equation for an alpha particle within the nucleus yields a finite probability that the particle may be found at a given instant *outside* the potential barrier.[53] This has been referred to as the "tunnel effect."

It follows also, from these considerations, that the higher the energy of the nuclear alpha particle the greater the probability that it will be found outside the nucleus. This is another way of stating the Geiger-Nuttall rule.

Additional information regarding alpha particles and their decay is given in Refs. 4, 49, and 54 to 81.

BETA-PARTICLE EMISSION

Beta-particle radiation was first discovered by Becquerel[1] and was later studied by the Curies[2] about 1900. These early investigators identified this type of radiation as consisting of high-energy electrons. In 1932 Anderson[82] discovered the positron, which is an electron with a positive charge. The positron was first identified in cloud chamber pair production by cosmic radiation but was later observed in radioactive emissions.

To investigate the nature of beta-particle radiation, the charge of the particles and the charge-to-mass ratio can be measured in exactly the same manner previously outlined for alpha particles. The results of such measurements indicate complete physical identity between beta particles and high-speed electrons.[49]

In contrast to alpha radiation, beta particles are emitted in a continuous energy spectrum. A typical beta-particle energy distribution is illustrated in Fig. 2.20 for Bi^{210}.

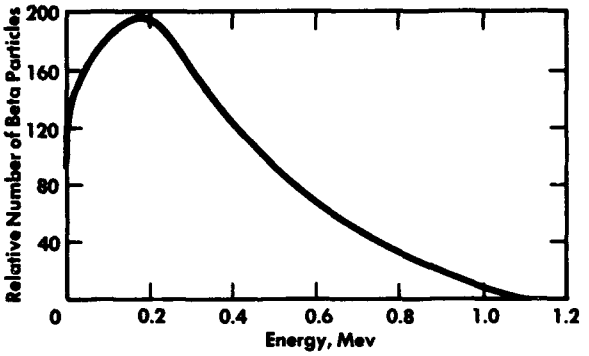


Fig. 2.20—A typical beta-particle energy distribution.

Such an energy distribution is typical of many beta emitters, and every beta emitter, regardless of the exact form of its energy spectrum, is characterized by having a definite maximum energy (E_{max}). It is this maximum energy, which is usually listed in tables of beta-particle energy. This continuous energy distribution of emitted beta particles cannot be explained by transitions between discrete nuclear energy levels as can alpha-particle emission.

Even if the beta particles were monoenergetic at emission and interacted subsequently with extranuclear electrons, only a finite number of energies would be possible instead of the observed continuous spectrum.

The theories of modern physics indicate that the electron does not exist within the nucleus to be emitted when sufficient energy is acquired. This may be deduced by application of the Heisenberg Uncertainty Principle,[83] which, in simple form, states that the simultaneous determination of the position and momentum of a subatomic particle is impossible. That is, if the momentum of a particle is measured precisely, its position must be uncertain, and, if its position is determined accurately, the momentum will be in doubt. This is not an experimental limitation but is a fundamental characteristic of subatomic particles. When compared with any detector the particles are so small that the detector must influence either their location or their momentum, permitting the accurate determination of only one of these properties at one time. For example, if the location of an electron is to be detected by means of a photon, the interaction between the photon and the electron will determine its position at the time of interaction but will change the momentum of the electron by an unknown small amount. Analytically the principle can be stated as

$$(\Delta p)\,(\Delta x) \geq h$$

or

$$(\Delta E)\,(\Delta t) \geq h \tag{2.33}$$

where Δp = uncertainty in momentum
Δx = uncertainty in position
ΔE = uncertainty in energy
Δt = uncertainty in time
h = Planck's constant

By taking Δx, the uncertainty in position, to be of the order of a nuclear diameter, the Uncertainty Principle may be used to show that the range ΔE of energy the electron could then have is very much greater than the nuclear binding energy. The electron, therefore, cannot be considered to be located within the nucleus.

An energy balance between parent and product nuclei leads to the conclusion that the energy represented by the maximum beta-particle energy, E_{max}, must be given up in beta-particle emission. For example, C^{14} emits a beta particle to become N^{14} which can be obtained independently from another reaction[4]

$$C^{14} + p \rightarrow N^{14} + n - Q \tag{2.34}$$

where Q has been determined experimentally to be 0.628 Mev. The mass difference between C^{14} and N^{14} is then, transforming energy to mass, where amu stands for atomic mass unit

$$\begin{aligned} m(C^{14} - N^{14}) &= m(\text{neutron}) - m(\text{proton}) - \frac{0.628\ \text{Mev}}{93616\ \text{Mev/amu}} \\ &= 1.008982 - 1.008142 - 0.000674\ \text{amu} \\ &= 0.000166\ \text{amu} \end{aligned} \tag{2.35}$$

But, this is exactly equal to the maximum beta-particle energy

$$\frac{0.155\ \text{Mev}}{93616\ \text{Mev/amu}} = 0.000166\ \text{amu}$$

As E_{max} is lost in every emission and only a fraction of this energy appears as the kinetic energy of the beta particle, one must either deny conservation of energy or postulate the existence of an undetected particle, the neutrino, which could share the emitted energy with the electron. Only thus can energy, momentum, and spin be conserved. The neutrino, with very

small mass relative to the electron, no charge, and spin $\frac{1}{2}$, fulfills the balance requirements of beta-particle emission. A particle with such characteristics as the neutrino would be extremely difficult to detect.

Careful experiments[84] in which a beta-particle emitter is placed inside a calorimeter designed to absorb all known disintegration products always yield the average energy (E_{avg}). That is, none of the energy carried away by the postulated neutrino is detected by calorimetry.

The Fermi theory of beta decay[85] can satisfactorily explain, for both positive and negative electrons, the theoretical problems discussed above in beta-particle emission. In this theory the electron and the neutrino are considered not to exist within the nucleus. They appear in the course of transitions between nuclear energy levels in a manner analogous to the appearance of X-ray photons during the transition from one electronic level to another.

When the beta particle emitted is negative, a neutron is transformed into a proton within the nucleus, increasing the net positive charge of the nucleus by one unit, one step up the periodic table. The reverse is true for positron (positive beta particle) emission. These emissions may be represented by these processes, respectively

$$ {}_0n^1 \rightarrow {}_1H^1 + -{}_1e^0 + {}_0\nu^0 \qquad (2.36) $$

$$ {}_1H^1 \rightarrow {}_0n^1 + {}_1e^0 + {}_0\nu^0 \qquad (2.37) $$

where ${}_0\nu^0$ represents a neutrino.

ABSORPTION OF BETA-PARTICLE RADIATION

As in the case of alpha particles, the range of beta particles in matter can be related to maximum particle energy. Once such a relationship as the plot shown in Fig. 2.21 is established, maximum energy can be determined by absorption measurements.[86,87]

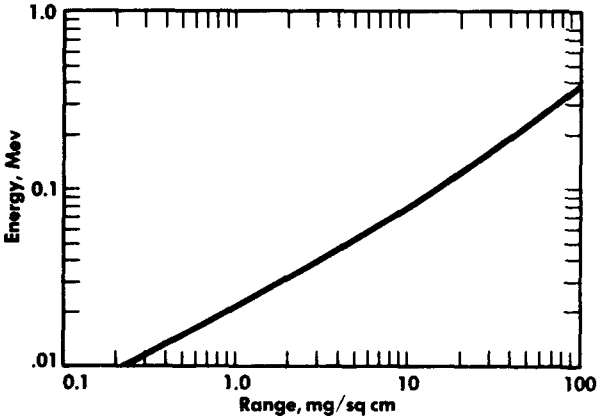


Fig. 2.21—Gross absorption properties, including scattering and radioactive losses as well as ionization losses, of a complex continuous energy spectrum.

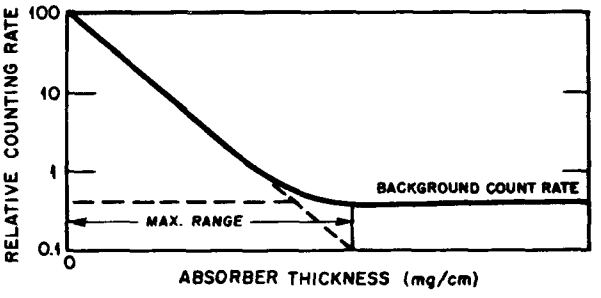


Fig. 2.22—Beta-particle counting rate vs. absorber thickness.[152]

Beta particles are much more penetrating than alpha particles. As an absorbing medium is interposed between the radioactive source and the detector, the counting rate initially decreases nearly logarithmically with absorber thickness, as shown in Fig. 2.22.

The maximum beta-particle range, and therefore energy, is determined by the point at which the beta-particle counting rate curve intersects the very nearly horizontal background counting rate due to gamma radiation. As visual determination of this intercept entails considerable uncertainty, another method of data analysis, due to Feather, is used where more precision is demanded.

The data plotted in Fig. 2.22 represent the gross absorption properties, including scattering and radioactive losses as well as ionization losses, of a complex continuous energy spectrum. The occurrence of a linear function would seem to be entirely fortuitous. The correlation between the slope, N, of this linear portion of the beta-particle absorption curve and energy, however, is significant, and the relation given by Eq. 2.38 is frequently used to determine the maximum energy.

$$\alpha = \frac{22}{E_{max}} 1.33 \tag{2.38}$$

where $\alpha = N/\rho$

ρ = density

Absorber thickness is commonly measured in milligrams per square centimeter, which is derived as the product of thickness and density. The important advantage gained by expressing absorber thickness in this way is that the range then becomes essentially independent of the nature of the absorbing medium. This occurs because the loss of energy by beta particles passing through matter depends on the total number of electrons with which they may interact. This electron density depends on the ratio of atomic number to mass number, Z/A, which decreases slightly with increasing Z. This variation is not large and for most work it is sufficiently precise to say that the range in milligrams per square centimeter is independent of absorber.

At higher energies, from 10 to 100 Mev and above, depending on the atomic number of the absorber, beta particles lose energy primarily by two radiative processes.[90]

The first process results from the deceleration of the charged beta particles in the electric field of force close to a nucleus and yields a continuous X-ray spectrum called bremsstrahlung. The magnitude of this phenomenon is difficult to calculate; however, a simple approximation is frequently used

$$\frac{\text{radiative energy loss}}{\text{ionization energy loss}} = \frac{EZ}{800} \tag{2.39}$$

where E = particle energy

Obviously the production of X radiation within a medium designed completely to absorb beta radiation is very important in shield design problems.

The second process is known as Čerenkov radiation[91] and is the result of high-speed electrons traversing a dielectric medium, like lucite, which has a low value for the phase velocity of light. At high energies, the electron may be traveling faster than the phase velocity of light in the medium and is then subject to a continuous radiative loss of energy. This radiant energy usually lies in or near the visible range and this process need not therefore be considered in the shielding calculations.

The half-lives of beta-particle emitting radioisotopes range from fractions of a second to millions of years. Table 2.10 lists some pure beta-particle emitting radioisotopes, their beta-particle energy (maximum) and maximum range.

Additional information on beta-particle emitters and beta decay is given in Refs. 4, 49, 54 to 57, and 92 to 117.

GAMMA-PHOTON EMISSION

Gamma radiation is electromagnetic radiation, as are radio waves, light, and X radiation, but of shorter wavelength and higher frequency than the other forms. Thus, except for its origin, gamma radiation is identical with "hard" or penetrating X radiation. The energy of the gamma radiation emitted by different radioisotopes varies considerably. A typical gamma energy of about 1 Mev would correspond to the maximum energy of X radiation from a million-volt X-ray machine.

Table 2.10—RANGES AND ENERGIES OF SELECTED BETA-PARTICLE-EMITTING RADIOISOTOPES[4]

Nuclide	End point energy, Mev	Range, mg/cm^2
Ra^{228}	0.053	6
Rb^{87}	0.13	20
Nb^{95}	0.146	30
Lu^{176}	0.22	48
Co^{60}	0.31	81
Zr^{95}	0.400	122
Be^{10}	0.555	181
I^{131}	0.600	213
Sb^{124}	0.65	254
Mn^{56}	0.73	277
Au^{98}	0.97	399
C^{11}	0.98	447
Ba^{140}	1.022	426
Mn^{56}	1.05	462
Cd^{115}	1.13	527
Bi^{210}(RaE)	1.17	508
N^{13}	1.24	557
Na^{24}	1.39	601
Sr^{89}	1.50	741
P^{32}	1.71	810
Te^{129}	1.80	812
Mg^{27}	1.80	821
Mg^{27}	1.80	885
Y^{90}	2.18	1065
Bi^{212}(ThC)	2.25	1023
Rh^{106}	2.30	1080
Pa^{234}	2.32	1105
Sb^{124}	2.37	1220
As^{76}	2.56	1384
Rh^{104}	2.6	1198
Mn^{56}	2.86	1440
Cu^{62}	2.92	1440
Pr^{144}	3.07	1575
As^{76}	3.12	1454
Rh^{106}	3.55	1770

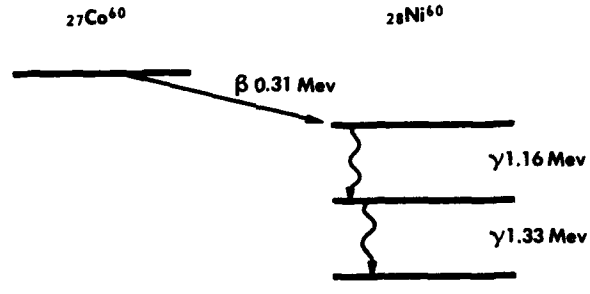


Fig. 2.23—A diagram showing the decay of radioactive Co^{60} to stable Ni^{60}.

Wavelengths of various types of electromagnetic radiation range from a millionth of an Angstrom up to 10^{17} A (1 A equals 10^{-8} cm). Frequencies range from 60 cycles/sec up to 10^{23} cycles/sec. There is considerable overlap in some regions (particularly gamma radiation and X radiation), and the range of wavelengths is not always well defined.[118]

The energy of gamma radiation is inversely proportional to its wavelength and directly proportional to its frequency, as expressed by the basic equation for all types of electromagnetic radiation.

$$E = h\nu = \frac{hc}{\lambda} = \frac{0.012354}{\lambda^*} \tag{2.40}$$

where ν = frequency
λ = wavelength
λ^* = wavelength in Angstroms
h = Planck's constant
c = velocity of light, centimeter per second

An excited nucleus, which neither fissions nor emits particles, must lose energy and so gain a more stable state by the emission of one or more gamma photons.

Excitation of the nucleus may be produced in a number of ways of particle bombardment, fission of a parent nucleus, or neutron capture. The stable isotope Co^{59} captures a neutron to produce Co^{60}, a well-known gamma emitter.

Regardless of how a nucleus reaches a given excited state, the gamma radiation emitted upon the return to a lower or ground state will be characteristic of the upper and lower states and independent of the manner of excitation. In other words, gamma radiation from a given radioisotope will always have a characteristic energy or energies. Figure 2.23 is a diagram[119] showing the decay of radioactive Co^{60} first by emission of a beta particle to Ni^{60} in an excited state. This then deexcites by emitting two gamma rays to produce stable Ni^{60}.

Radioisotopes that emit alpha and beta particles are often gamma emitters as well because the emission of alpha or beta particles leaves the daughter nucleus in an excited state. The daughter nucleus then achieves a more stable state by emission of one or more gamma photons. This emission follows almost instantly after the emission of the alpha or beta particle usually within 10^{-12} sec. Gamma emitters having longer half-lives usually decay by isomeric transition.

In addition to beta-particle emissions which may accompany gamma radiation, some X radiation and some photoelectrons may be observed as a result of the phenomenon known as internal conversion.[120] Not all gamma photons emitted from nuclei escape the electron cloud surrounding each nucleus. A gamma photon may interact with an inner-shell (K shell) electron with sufficient energy to eject the electron from its orbit. This electron will have an energy equal to the energy of the gamma photon less the binding energy of the electron in the atom. An electron from an outer shell (L shell) will then take the place of the ejected electron and the energy it loses in doing this appears as an X-ray photon.

The ratio of the number of conversion electrons emitted to the number of primary gamma rays emitted is known as the internal conversion coefficient. The internal conversion coefficient is inversely proportional to the primary gamma ray energy; therefore, the number of conversion electrons is small for energetic gamma photons.

Table 2.11 lists some commonly used gamma-emitting radioisotopes, their half-lives, and the gamma energy.[121]

INTERACTION OF GAMMA RADIATION AND MATTER

The absorption of gamma radiation in any medium occurs almost entirely by interaction with free or bound electrons. There are three distinct processes of interaction between the gamma photon and the electron: the photoelectric effect, the Compton effect, and pair production. The importance of each process depends on the energy of the gamma radiation and the atomic number of the absorber.

Table 2.11—CHARACTERISTICS OF SELECTED GAMMA-EMITTING RADIOISOTOPES[121]

Nuclide	Energy, Mev	Half-life	$\frac{mr}{(hr)\ (mc)}$ at 1 meter
Na^{22}	1.277; 1.30	2.6y	1.29
Na^{24}	(1.3679; 2.7537)	15.0h	1.92
Mn^{52}	(0.73; 0.94; 1.46)	6.0d	1.93
Mn^{54}	(0.84)	310d	0.49
Fe^{59}	(1.295; 1.097)	45d	0.65
Co^{58}	(0.81; 0.6)	72d	0.56
Co^{60}	(1.3316; 1.1715)	5.3y	1.31
Cu^{64}	1.35	12.8h	0.12
Zn^{65}	1.125	250d	0.30
As^{76}	0.57; 1.25; 1.84; 2.15	26.8h	0.40
Br^{82}	0.547; 0.608; 0.692; 0.766; 0.823; 1.031; 1.312	35.9h	1.5
I^{128}	(0.428)	25m	0.018
I^{130}	(0.744; 0.417; 0.667)	12.6h	1.25
I^{131}	(0.080; 0.28413)	8.1d	0.24
Cs^{137}	(0.6616)	33y	0.32
Ta^{182}	1.121; 1.223; 1.188	111d	0.6
Au^{198}	(0.41177; 0.676)	2.7d	0.25
Ra^{226}	(0.188)	1,620y	0.84

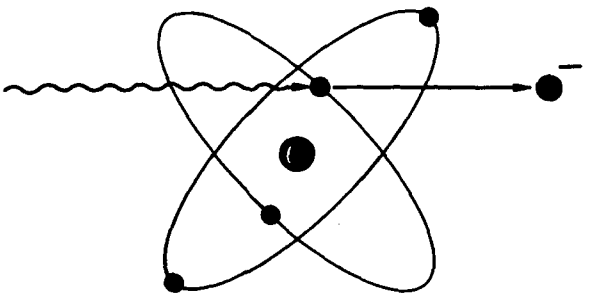


Fig. 2.24—Photon interacting with orbital electron ejects electron from one of the shells.

PHOTOELECTRIC EFFECT

A gamma photon may interact with an orbital electron in the electron cloud surrounding the nucleus and, in so doing, may transfer its energy to the electron ejecting it from one of the shells (Fig. 2.24) or raising it to an unoccupied position in another shell.

In the photoelectric effect the quantum of gamma radiation, now called a photon, disappears. Its energy is totally expended in overcoming the forces binding an orbital electron to an atom of the absorber and in imparting kinetic energy to this electron (see Fig. 2.24). This kinetic energy of the emitted electrons, the primary evidence of the particulate nature of gamma radiation, is found to depend only on the energy of the incident gamma radiation and the shell from which the electron is ejected. The energy of the incident gamma radiation as determined from such an energy balance is constant for a given wavelength.[122] The production of a photo-

electron, therefore, can be considered to be a single elastic collision between two "particles," one of which has no mass.

The relationship in this interaction is given by the Einstein Photoelectric Equation[123]

$$h\nu = w + \tfrac{1}{2}mv^2 \qquad (2.41)$$

where $h\nu$ = energy of photon
w = work function of the material
$\tfrac{1}{2}mv^2$ = kinetic energy of photoelectron

Any orbital electron can be ejected or shifted by this process as long as the energy relationship is satisfied. This interaction occurs mostly with electrons close to the nucleus (K shell). These electrons are bound with the greatest energy. As the energy of the gamma photon is increased above the absorption limit of these (K shell) electrons, the absorption by this process decreases. Heavier elements are effective at higher energy levels than the lighter elements because of the greater energy required to eject electrons (i.e., w is larger for heavier nuclei). Thus, while the photoelectric effect is important only up to about 0.10 Mev for aluminum, it is important up to about 1.0 Mev for lead.

The ejection of an electron by a gamma photon is followed by the emission of soft X radiation as the position of the ejected electron is refilled. Interaction with other orbital electrons not necessarily in the same atom, rapidly absorbs this soft X radiation. The photoelectric effect can be considered to be "true" absorption resulting in the complete removal of a gamma photon.

COMPTON SCATTERING

If the energy of the incident gamma radiation is increased until the energy required to remove an electron from its shell is negligible by comparison, the Compton effect becomes important.

At higher gamma energies there is a decreased probability of all of the energy and momentum of the gamma photons being absorbed in the photoelectric process. High-energy gamma photons are not completely removed by interaction with orbital electrons but behave like particles or billiard balls. Figure 2.25 is a diagram indicating the process of Compton scattering.[124]

In this type of interaction, kinetic energy is imparted to an electron, but all the energy of the incident photon is transferred to the emitted electron and a photon of somewhat lower energy than the incident photon results (see Fig. 2.25). This interaction can be explained in terms of a collision between two particles, where the incident particle loses part of its energy to the other particle. Energy and momentum are conserved.

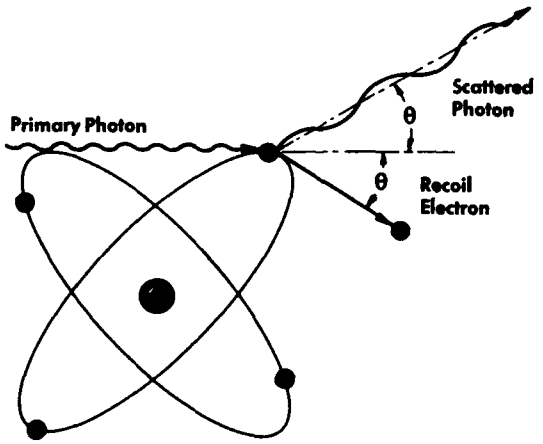


Fig. 2.25—Diagram indicating the process of Compton scattering.

The primary gamma photon can be treated as a particle whose equivalent mass is

$$m = \frac{h\nu}{c^2} \tag{2.42}$$

where m = equivalent mass of gamma photon
$h\nu$ = energy of gamma photon
c = velocity of light

The mass of an electron has an energy equivalence of 0.51 Mev. The relationship between the energy of the incident gamma photon and the scattered photon is

$$E = \frac{E_0}{1 + (E_0/0.51)(1 - \cos\theta)} \tag{2.43}$$

where E_0 = energy of incident gamma photon, Mev
E = energy of scattered photon, Mev
θ = scattering angle (see Fig. 2.24)

Substitution into Eq. 2.43 shows that where θ is small there is little degradation of energy and that where θ is large ($\pi/2$ or greater) the energy of the scattered photon will be less than 0.51 Mev, regardless of the energy of the incident gamma photon. Also, the greater the energy of the incident photon, the smaller will be the fraction (E/E_0) of the energy carried away by the scattered photon. By energy balance this indicates that ejection of electrons with much more energy will occur as the energy of the incident photon is increased.

Contrary to the photoelectric process, Compton scattering cannot be considered to be a true absorption process as the scattered photons may not be appreciably degraded in energy or deflected. As this process does not completely absorb the photons, radiation in this range creates some shielding problems.

PAIR PRODUCTION

As the energy of the incident radiation is increased to exceed twice the mass of the electron (1.02 Mev), another type of interaction becomes important. This is known as pair production, and a diagram of the process is shown in Fig. 2.26.

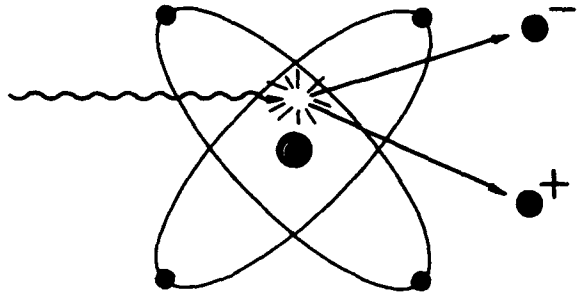


Fig. 2.26—Diagram of pair production.

An incident photon in the vicinity of an atomic nucleus disappears entirely and a pair of particles is produced, each with the mass of an electron but with equal and opposite charges. One particle is an ordinary electron, and the other is termed a positron. The process only occurs in the coulomb field of the nucleus. The atom does not directly participate in the process, but its presence permits the law of conservation of momentum to be satisfied.[125]

Figure 2.27 is a diagram of pair production. An incident gamma photon produces an electron and a positron, which travel from the point of interaction in curved paths because of the magnetic field in the cloud chamber. The paths of the two particles have opposing curvatures

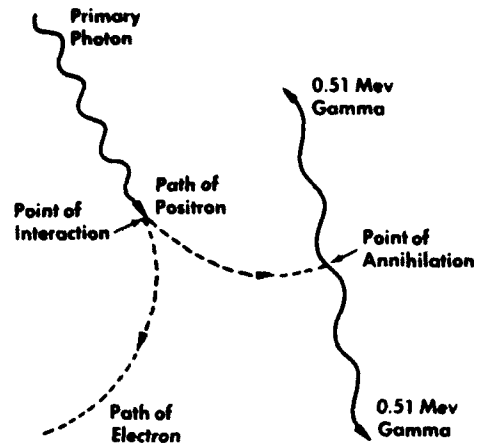


Fig. 2.27—Diagram of pair production.

because the particles have charges of different sign. Each point in the cloud chamber track represents an ionization, that is, an interaction between one of the pair of particles and an orbital electron of the atoms in the cloud chamber.

It is rather remarkable that a positron can interact with an orbital electron without annihilation, but in fact this is only possible when the positron has a high velocity. Repeated interaction degrades the energy of the positron until it finally combines with an electron in a process of annihilation, the energy appearing as two gamma photons, emitted in opposite directions, each of energy 0.51 Mev.

As the soft gammas (0.51 Mev) from the annihilation of positrons are easily stopped, and since both particles of the pair travel only a short distance in an absorber, the process of pair production can be considered to be true absorption with the complete elimination of the incident gamma photon.

Figure 2.27 illustrates the apparent exchange in nature of the energy, which first appears as radiation and then becomes matter, only to be changed back into radiation again by the positron annihilation with an electron. A discussion on the nature of waves and particles follows, as modern physics indicates that some wavelike properties must be ascribed to particles and some particulate properties to radiation.

NUCLEAR RADIATIONS AS WAVES AND AS PARTICLES

From the previous discussion of alpha, beta, and gamma radiation, it might appear that these radiations can be classified simply as particles or as waves. This classical outlook would lead to a predictable interaction with matter: the electromagnetic wave would add energy continually to the orbital electron with which it is interacting. A particle, on the other hand, would interact with other particles by "collision" with the resulting exchange of energy and momentum governed by the laws of classical "billiard ball" mechanics. These concepts nevertheless fail to explain a large number of observations.

All nuclear radiations appear to possess a dual character. In one experiment a certain type of radiation may exhibit the characteristics of particulate emission. De Broglie[155] hypothesized that all so-called particulate radiations, such as alpha and beta particles, protons and neutrons have characteristic wave properties in which the momentum of the particle is related to the wavelength by

$$mv = \frac{h}{\lambda} \tag{2.44}$$

The significance of the wavelength of a particle may be demonstrated by means of a simple experiment with monochromatic light. In this experiment light of wavelength λ passes

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through a narrow slit to a screen. If the slit width is of the order of the wavelength of the light, a characteristic diffraction pattern with maxima and minima appears on the screen, as shown in Fig. 2.28.

The maxima and minima in light intensity indicated in Fig. 2.28 may be explained in terms of interference of the diffracted monochromatic light. Measurement of the angular spacing of the minima is a sensitive method of wavelength measurement. If the light can be considered as a beam of particulate photons, the intensity curve becomes a measure of the probability of a photon following that path. Similarly, if the electrons, protons, or neutrons pass through a slit of the appropriate dimension, they produce diffraction patterns in exactly the same way as monochromatic light. The probability of a particle striking a particular spot on the screen will follow a curve with maxima and minima, as shown in Fig. 2.28. Davisson and Germer[154] studied the diffraction of electrons from a nickel crystal, calculating the wavelength from the resulting pattern and showed that this wavelength was equal to h/mv.

Neutrons are uncharged particles of essentially the same mass as the proton and, since there are no charges to complicate the interaction with matter, clearly exhibit wavelike properties. This wave nature of neutrons is shown by their diffraction from regular crystalline lattices. The resulting diffraction patterns are characteristic of radiation, having a wavelength comparable with the dimensions of the structure giving rise to the diffraction. Neutron diffraction was first reported by Mitchell and Powers[156] in 1936. Laue type diffraction patterns have been produced of the same pattern as produced by X radiation. This is a striking example of the dual character of subatomic particles.

Thus, the neutron can be assigned a wavelength according to the de Broglie relation

$$\lambda = \frac{h}{mv}$$

Substituting 6.6×10^{-27} erg-sec for Planck's constant, and substituting $\sqrt{2E/m}$ for v from $E = \frac{1}{2}mv^2$

$$\lambda = \frac{h}{\sqrt{2mE}} \tag{2.45}$$

If h is measured in erg-second, m in grams and E in ergs, the wavelength will have dimensions of centimeters. Also, as 1 amu of mass equals 1.67×10^{-24} g and 1 ev is equal to 1.6×10^{-12} erg, we have

$$\lambda = \frac{4.05 \times 10^{-9}\ \text{cm}}{\sqrt{2mE}}$$

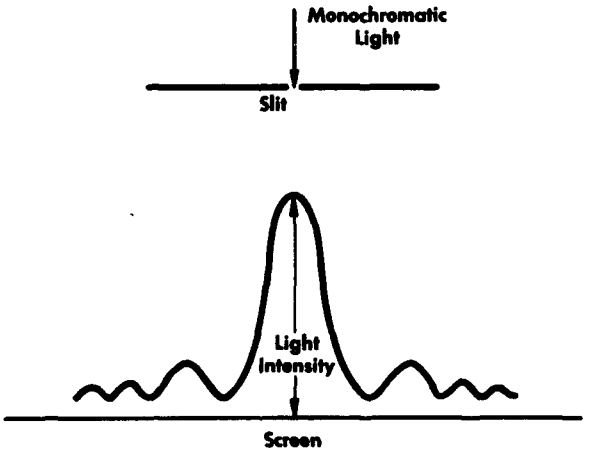


Fig. 2.28—Characteristic diffraction pattern with maxima and minima appearing on the screen.

As the mass of a neutron is approximately equal to 1 amu, this equation becomes

$$\lambda = \frac{2.86 \times 10^{-9} \text{ cm}}{\sqrt{E}}$$

$$\lambda = \frac{0.286}{\sqrt{E}} \text{ A} \qquad (2.46)$$

where E = energy of neutrons in electron volts

Since the wavelength varies with the energy of the neutrons, fast neutrons having wavelengths about 10^{-13} cm exhibit diffraction by nuclei and slow neutrons having about 10^{-8} cm wavelength exhibit diffraction due to atomic obstacles.

NATURE OF A PARTICLE

A particle may be defined as a concentration of matter, of small dimensions, with the characteristic physical property of mass. Associated with a moving mass, according to classical mechanics, are the quantities: momentum = mass × velocity, and kinetic energy = $\frac{1}{2}$ mass × (velocity)^2. Both of these quantities must be preserved in any classical interaction. It should be possible also to state exactly the position and velocity of a classical particle.

As the properties of alpha and beta particles are studied further, some ambiguities appear. It is not possible to describe these emissions entirely in terms of a stream of particles.

Careful experiments designed to measure the dimensions of these particles have been found to be lacking the two primary attributes conceptually assigned to a particle: a definite size and location. Only the probability of a particle being found at a specific location may be determined. The volume where a maximum occurs on this plot is the nearest approach to determination of the "position" of the particle. The extent in space of this maximum is an approach to a measure of the "size." Thus, the naive concept of these entities as simple classical particles is clearly not satisfactory.

Further, under certain conditions they exhibit characteristics of wave motion. A collimated stream of classical particles, moving in a straight line and passing through an aperture would form a pattern exactly equivalent to a geometrical shadow of the aperture. However, as previously discussed, the patterns determined experimentally for alpha and beta particles are identical in nature to the diffraction patterns characteristic of wave motion! The "wavelength" of the particles, as determined from diffraction patterns, is found to be

$$\lambda = \frac{h}{mv}$$

where h is Planck's constant, equal to 6.6×10^{-27} erg-sec. Alpha and beta particles emitted by radioactive materials have been shown to consist of positively charged helium nuclei and negatively charged electrons, respectively. Both have been demonstrated to be discrete entities, yet neither possesses the definite characteristics of a particle and both exhibit some characteristics of a wave.

NATURE OF A WAVE

The third type of radioactive emission, gamma radiation, was found to have wave characteristics and was considered to be continuous electromagnetic radiation with a definite wavelength associated with each emission.

In its interaction with matter, however, gamma radiation is found to behave in some respects as a stream of particles! Three types of interactions are observed as discussed in the section on absorption of gamma radiation: photoelectric effect, Compton scatter, and pair formation. In each of these interactions the energy exchanges are those characteristic of particulate collisions rather than the continuous transfer of energy characteristic of classical wave motion, and the formulas describing these events, developed using the concept of the gamma photon as a particle, are found to be obeyed experimentally.

Thus, the simple concepts of particles and waves are clearly insufficient to explain the interactions of emanations from radioactive materials with matter. The attempts to classify these radiations as either waves or particles appear to have been terminated and the modern scientist has come to accept the idea that there exists here a physical entity that will not fit solely into either of two irreconcilable concepts but partly into both.

The radiation emitted from a particular transition, whether visible light, X ray or gamma ray, is never of a single pure (monochromatic) wavelength. A range of wavelengths is always present. There is a peaking at one value and a symmetrical distribution on both sides of the maximum. When a complete Fourier analysis of such a wavelength distribution is made and the resulting wave form is calculated, the concept is obtained of a wave packet of finite length. Such a packet possesses both particle and wave properties.

ABSORPTION OF GAMMA RADIATION

The following paragraphs deal with the factors used to calculate gamma radiation absorption.

ABSORPTION COEFFICIENTS FOR NARROW BEAMS

Figure 2.29 shows a plot of the linear absorption coefficient for "narrow-beam" (collimated) gamma radiation in lead.[89] The absorption coefficient may be divided into three com-

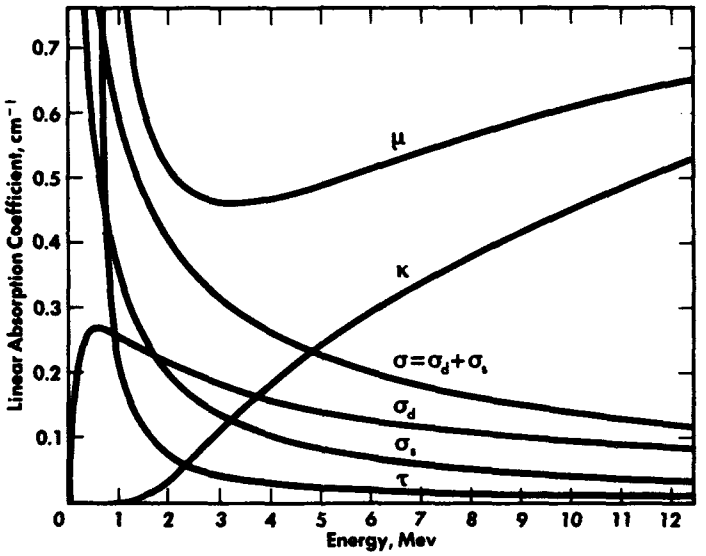


Fig. 2.29—A plot of the linear absorption coefficient for "narrow-beam" (collimated) gamma radiation in lead.

ponents, τ, σ, and κ, corresponding to the three processes of photoelectric effect, Compton effect, and pair production, respectively. These components are also shown in Fig. 2.29 and the dependence of each on the energy of the radiation is apparent. Absorption by photoelectric effect, τ, is shown to be most important in the low-energy region, particularly for the heavy elements. Figure 2.29 shows that below 0.5 Mev the primary contribution to μ is from τ. However, the curve of τ vs. gamma energy drops very rapidly in this range with the result that above 1.0 Mev there is very little absorption of gamma radiation by photoelectric effect. At medium energies of 0.5 to 5 Mev, Compton scattering, σ, is the most important absorption process. For the heavy elements, Compton scattering is less important below 0.5 Mev than the photoelectric effect, but for the lighter elements the importance of Compton scattering continues down to low energies.

The process of absorption by pair production has a definite threshold of 1.02 Mev corresponding to the energy equivalent of two electron masses. Above this threshold, absorption by this process increases rapidly, whereas absorption by other processes continues to decrease. If the gamma radiation is well collimated, that is, the rays are parallel, there will be no geometrical reduction in intensity, and the reduction will be solely due to absorption. Photons scattered by the Compton effect can be considered as lost, as they no longer contribute significantly to the collimated beam. Thus, there will be no degrading of the energy of the collimated photons as is the case in the absorption of charged particles. Each absorption event will result in the complete removal of a photon. At any absorber depth x, the rate at which the number of photons decreases will be directly proportional to the absorption coefficient and to the number of photons present, or

$$\frac{dI}{dx} = -\mu I \tag{2.47}$$

Integrating (for a homogeneous absorber)

$$I = I_0 e^{-\mu x} \tag{2.48}$$

where I_0 = intensity of gamma radiation at surface, roentgens per hour

I = intensity of gamma radiation at distance x, roentgens per hour

x = absorber thickness, centimeter

μ = linear absorption coefficient, reciprocal centimeters

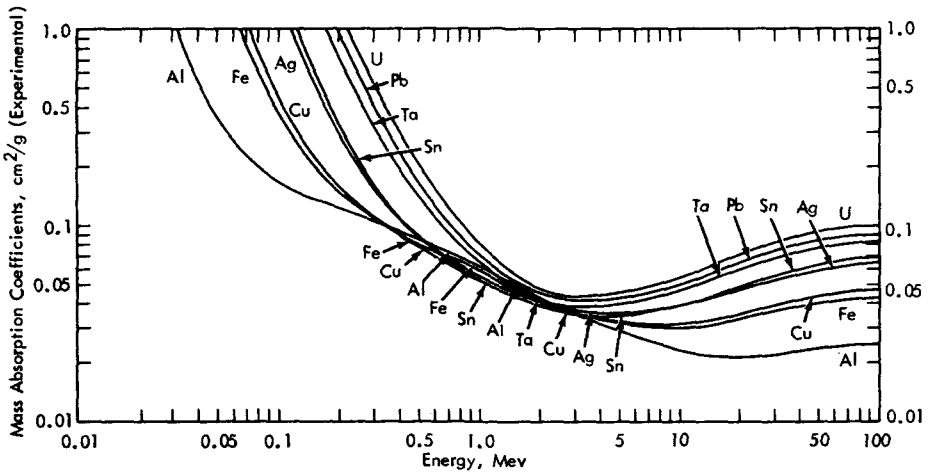


Fig. 2.30—Mass absorption coefficients for the photoelectric, Compton scatter, and pair production regions for some of the heavier elements as a function of energy.

In using Eq. 2.48 it is not necessary to express the absorber thickness length. Sometimes it is more convenient to use mass per unit area, especially when considering a nonhomogeneous absorber. In such a case x may be expressed in grams per square centimeter or in pounds per square foot. The absorption coefficient is then termed the mass absorption coefficient, μ_m, and has the units of square centimeter per gram or square foot per pound, respectively. The simple relationship $\mu_m = \mu\rho$, where ρ is the density of the absorber, is convenient in relating these coefficients. Figure 2.30 gives mass absorption coefficients for the whole range, the photoelectric, Compton scatter, and the pair production regions for some of the common elements as a function of energy.[153]

HALF-VALUE AND TENTH-VALUE THICKNESSES

Instead of using absorption coefficients, it is often more convenient to express the absorption in terms of half-value layers or tenth-value layers. A half-value layer is the thickness of an absorber that will transmit one-half of the beam, and, similarly, a tenth-value layer is the thickness of an absorber that will transmit one-tenth of the beam. The following relationships exist

$$\text{half-value thickness, } t_{1/2} = 0.693/\mu \tag{2.49}$$

$$I = \frac{I_0}{2^n} \tag{2.50}$$

where n = number of half-value thicknesses.

Figure 2.31 shows some tenth-value thicknesses (narrow beam) for some common materials as a function of energy.[126] Some half-value thicknesses (broad beam) of different foods have been determined in the Fission Products Laboratory of The University of Michigan.[127]

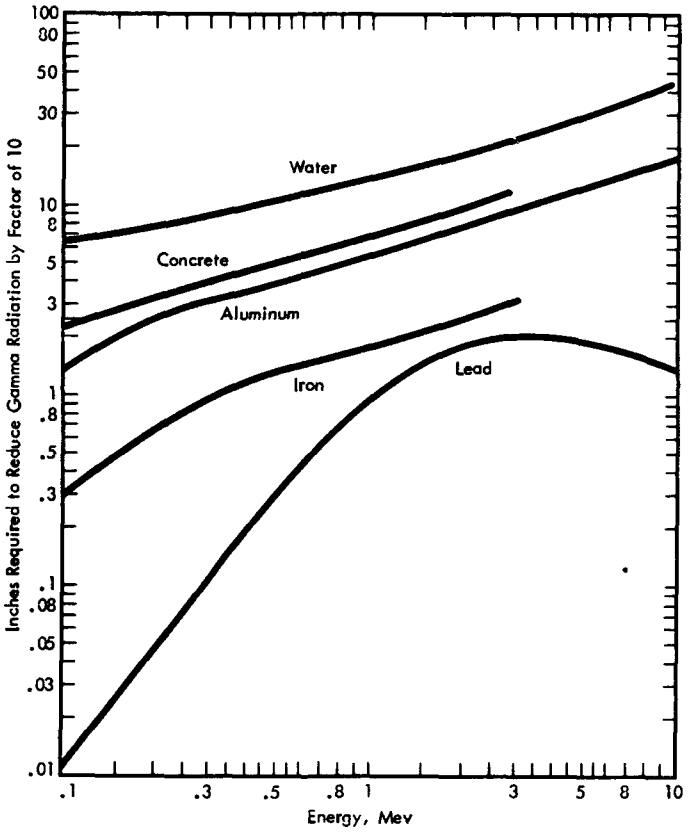


Fig. 2.31—Some tenth-value thicknesses for some common materials as a function of energy.

DIFFERENT ENERGIES

In the case of mixture of gamma-emitting radioisotopes such as the fission products, gamma radiation given off has a mixture of different energies. However, the absorption coefficients and half- and tenth-value thicknesses given are constant only for a given absorber and energy. When a mixture of energies is involved, the decrease in the intensity of a collimated beam is given by[90]

$$I = I_1 e^{-\mu x} + I_2 e^{-\mu_2 x} + \ldots I_n e^{-\mu_n x} \tag{2.51}$$

where $I_1, I_2, \ldots I_n$ = the intensities at the surface of the absorber of each of n-radiation energies

$\mu_1, \mu_2, \ldots \mu_n$ = absorption coefficients for the corresponding radiation energies

If a large number of different energies are involved or if the energy is continuous in distribution, approximate methods or laboratory measurements are required.

BUILD-UP FACTORS

In the ideal case of absorption of a perfectly collimated beam, the degradation of energy, scattering, and the production of secondary radiation can be omitted from the considerations; but in most real applications the build-up of secondary radiation is an important consideration. The radiation intensity at any point in an absorber consists of: the primary gamma photons, the photons resulting from Compton scatter and the secondary radiation composed of photoelectrons and electrons from Compton scatter and pair production, and the X radiation produced by the absorption of these electrons. The secondary electrons are absorbed much more readily than the primary gamma radiation.

For primary gamma radiation of a given energy passing through a given absorber, an equilibrium level of the production of secondary radiation is reached that is characteristic of the energy and the absorber. If the primary radiation passes from a poor absorber, such as air, to a denser medium, the increase in absorption of the primary radiation will result in a corresponding increase in the formation of secondary radiation. This produces a build-up of secondary electrons and X radiation from the absorption of these electrons. Figure 2.32 is a diagram (not to scale) showing such a build-up of secondaries as a primary photon passes through a denser medium[90]

The ratio of the intensities of the secondary and primary radiations can be expressed as follows

$$\frac{I}{I_0} = \frac{\mu_0}{\mu - \mu_0} (e^{-\mu_0 x} - e^{-\mu x}) \qquad (2.52)$$

where I = intensity of secondary radiation

I_0 = intensity of primary radiation

μ = absorption coefficient for secondary radiations

μ_0 = absorption coefficient for primary radiations

x = absorber thickness

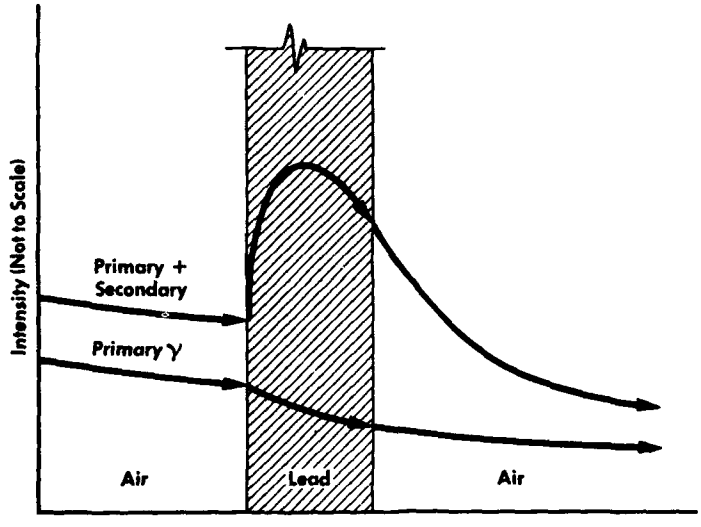


Fig. 2.32—Diagram showing a build-up of secondaries as a primary photon passes through a denser medium.

Table 2.12—DOSE BUILD-UP FACTOR B IN WATER FOR POINT ISOTROPIC SOURCE[128]

(Relaxation length, $\mu_0 x$)

E_0, Mev	1	2	4	7	10	15	20
0.256	3.09	7.14	23.0	72.9	166	456	932
0.5	2.52	5.14	14.3	38.8	77.6	178	334
1	2.13	3.71	7.68	16.2	27.1	50.4	82.2
2	1.83	2.77	4.88	8.46	12.4	19.5	27.7
3	1.69	2.42	3.91	6.23	8.63	12.8	17.0
4	1.58	2.17	3.34	5.13	6.94	9.97	12.9
6	1.46	1.91	2.76	3.99	5.18	7.09	8.85
8	1.38	1.74	2.40	3.34	4.25	5.66	6.95
10	1.33	1.63	2.19	2.97	3.72	4.90	5.98

Table 2.13—DOSE BUILD-UP FACTOR B IN ALUMINUM FOR POINT ISOTROPIC SOURCE[128]

(Relaxation length, $\mu_0 x$)

E_0, Mev	1	2	4	7	10	15	20
0.5	2.37	4.24	9.47	21.5	38.9	80.8	141
1	2.02	3.41	6.57	13.1	21.2	37.9	58.5
2	1.75	2.61	4.62	8.05	11.9	18.7	26.3
3	1.64	2.32	3.78	6.14	8.65	13.0	17.7
4	1.53	2.08	3.22	5.01	6.88	10.1	13.4
6	1.42	1.85	2.70	4.06	5.49	7.97	10.4
8	1.34	1.68	2.37	3.45	4.58	6.56	8.52
10	1.28	1.55	2.12	3.01	3.96	5.63	7.32

Table 2.14—DOSE BUILD-UP FACTOR B IN IRON FOR POINT ISOTROPIC SOURCE[128]

(Relaxation length, $\mu_0 x$)

E_0, Mev	1	2	4	7	10	15	20
0.5	1.98	3.09	5.98	11.7	19.2	35.4	55.6
1	1.87	2.89	5.39	10.2	16.2	28.3	42.7
2	1.76	2.43	4.13	7.25	10.9	17.6	25.1
3	1.55	2.15	3.51	5.85	8.51	13.5	19.1
4	1.45	1.94	3.03	4.91	7.11	11.2	16.0
6	1.34	1.72	2.58	4.14	6.02	9.89	14.7
8	1.27	1.56	2.23	3.49	5.07	8.50	13.0
10	1.20	1.42	1.95	2.99	4.35	7.54	12.4

Table 2.15—DOSE BUILD-UP FACTOR B IN LEAD FOR POINT ISOTROPIC SOURCE[128]

(Relaxation length, $\mu_0 x$)

E_0, Mev	1	2	4	7	10	15	20
0.5	1.24	1.42	1.69	2.00	2.27	2.65	2.73
1	1.37	1.69	2.26	3.02	3.74	4.81	5.86
2	1.39	1.76	2.51	3.66	4.84	6.87	9.00
3	1.34	1.68	2.43	3.75	5.30	8.44	12.3
4	1.27	1.56	2.25	3.61	5.44	9.80	16.3
5.1097	1.21	1.46	2.08	3.44	5.55	11.7	23.6
6	1.18	1.40	1.97	3.34	5.69	13.8	32.7
8	1.14	1.30	1.74	2.89	5.07	14.1	44.6
10	1.11	1.23	1.58	2.52	4.34	12.5	39.2

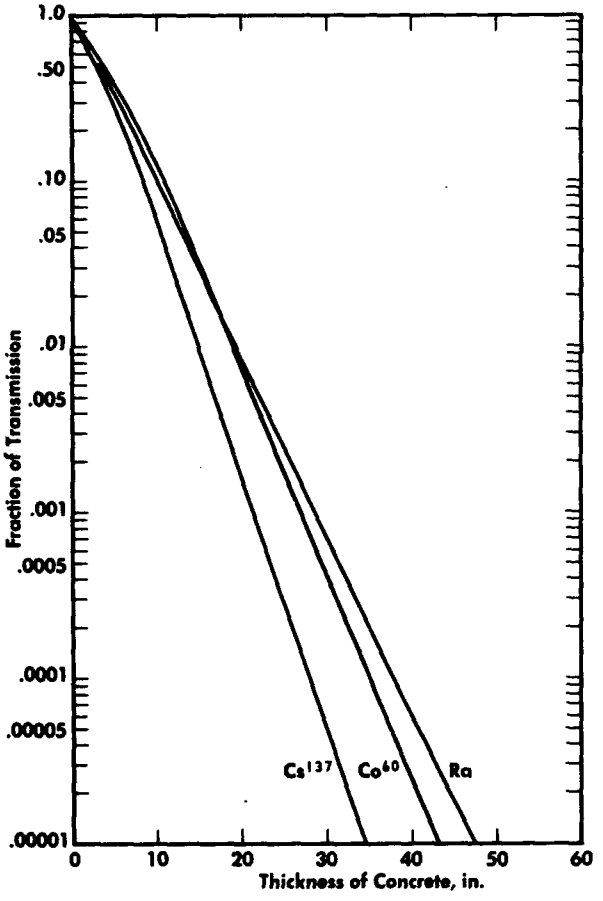


Fig. 2.33—Transmission in concrete of primary radiation from radium, Co^{60}, and Cs^{137} with corrections for secondary radiation.

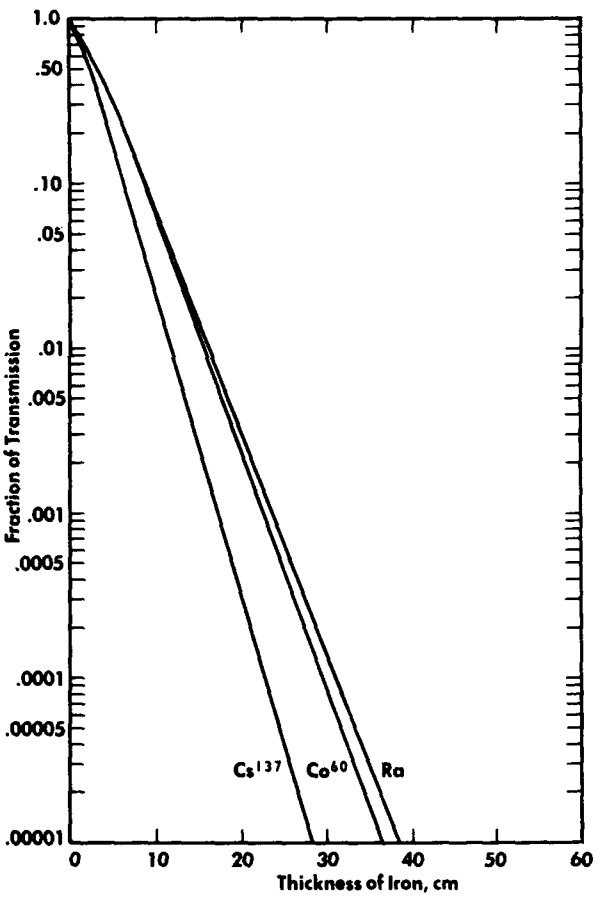


Fig. 2.34—Transmission in iron of primary radiation from radium, Co^{60}, and Cs^{137} with corrections for secondary radiation.

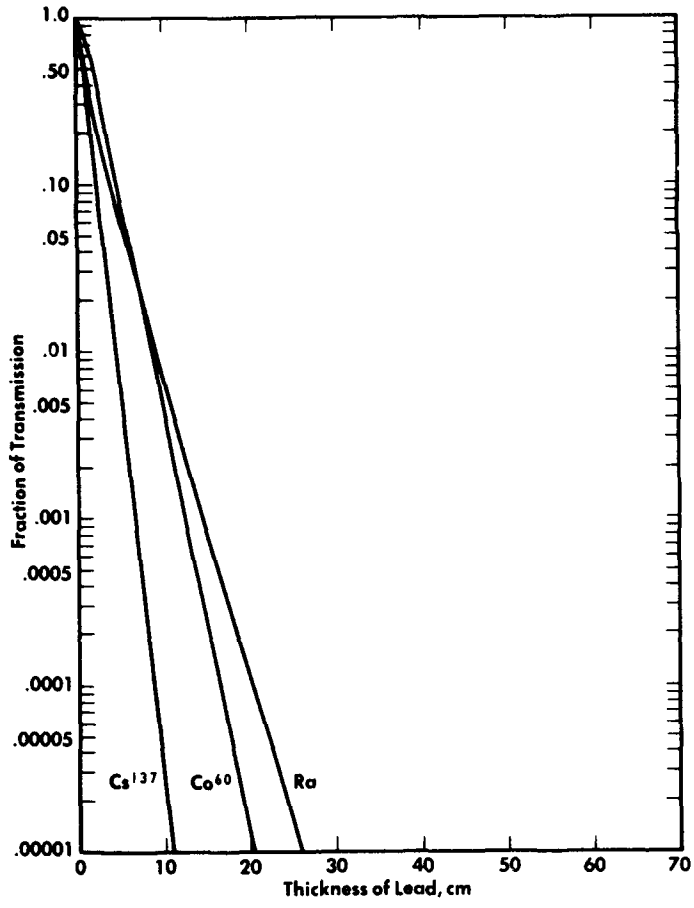


Fig. 2.35 — Transmission in lead of primary radiation from radium, Co^{60}, and Cs^{137} with corrections for secondary radiation.

At equilibrium and for a point value of x the relationship may be expressed as

$$\frac{I}{I_0} = \frac{\mu_0}{\mu - \mu_0} \tag{2.53}$$

Tables 2.12 to 2.15 give dose build-up factors B from a point source for water, aluminum, iron, and lead, respectively.[128] In using these factors the calculated intensity, I, based on absorption of a collimated beam is multiplied by factor B or

$$I = B\, I_0\, e^{-\mu x} \tag{2.54}$$

BROAD-BEAM COEFFICIENTS

Another method of calculation is to use coefficients determined for a broad beam rather than a narrow (collimated) beam. Figures 2.33 to 2.35 give transmission in concrete, iron, and lead, respectively, of primary radiation from radium, Co^{60}, and Cs^{137} with corrections for secondary radiation. These figures are based on broad-beam coefficients.[129]

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Chapter 3

CONTROLLED FISSION AND NUCLEAR REACTORS

The amounts of radiation from natural radioisotopes, such as radium, are minute compared with the tremendous yield from nuclear fission. Thus, nuclear reactors are the principal source of nuclear radiations and radioisotopes.

The neutron is the nuclear particle of greatest importance in the fission process. This chapter begins with a discussion of the neutron, its discovery, charge, mass, methods of detection, and capture cross section. Then follows a discussion of the discovery of nuclear fission, the fission process, the fission chain reaction, and the energy of fission.

The different methods of classifying nuclear reactors are considered, and the effect of neutron energy on choice of moderator and choice of coolant in the design of a reactor are discussed.

THE NEUTRON

The discovery of nuclear fission was preceded by the identification of the neutron in 1932 by Chadwick.[1] The existence of such a nuclear particle had been predicted some 12 years earlier by Harkins in the United States, Masson in Australia, and Rutherford in England. In 1920 Rutherford commented[2] on the possibility of the formation of a neutron from a proton and an electron, stating that, "Such an atom would have novel properties. Its external field would be practically zero ... and consequently it should be able to move freely through matter. Its presence would probably be difficult to detect." These predictions proved to be correct.

In 1930 Bothe and Becker, in Germany, reported[3] that the bombardment of beryllium by alpha particles from polonium produced a highly penetrating radiation. This radiation was thought at the time to be a high-energy form of gamma radiation, though it did not cause ionization. In 1932 the Joliots[4] repeated this experiment and observed that, when paraffin was placed in the beam of the new type of radiation, fast protons were ejected from the paraffin. The Joliots also considered the new radiation to be some form of electromagnetic radiation, but calculations from the measured energies of the "knock-out" protons lead to unreasonably high values for the energies of the supposed gamma photons. In 1932 Chadwick[1] solved the problem by suggesting that the new radiation was composed of particles with mass of unity on the atomic weight scale and without electric charge. He then showed how such a fast-moving particle could impart kinetic energy to hydrogen or other light nuclei, thus explaining the emission of energetic protons from paraffin bombarded by neutrons.

METHODS OF DETECTION

As neutrons are uncharged particles, they produce very little ionization in passing through matter. Therefore, instruments that measure radiation by the amount of ionization it produces cannot be used to measure neutron intensity. Neutrons are most easily detected by secondary

effects that result from the interaction between neutrons and various nuclei. Some of these interactions are listed below:

1. The capture of a neutron and the subsequent emission of a photon by the radioisotope thus formed.

2. Neutron capture followed by the prompt emission of a charged particle.

3. Neutron capture followed by fission of the compound nucleus.

4. Scattering of neutrons by a nucleus with conservation of kinetic energy and rest mass for the particles involved (elastic scattering).

5. Scattering of neutrons by a nucleus without conservation of kinetic energy and rest mass for the particles involved (inelastic scattering).

CHARGE

According to early experiments by Dee,[5] the charge of the neutron must be small because the neutron produces as a maximum only about one ion pair per meter in air. Dee stated that the maximum possible charge of the neutron is less than $^1/_{100}$ the charge of the proton. Today, however, neutrons are recognized to be uncharged particles. The lack of charge allows neutrons to induce nuclear reactions more readily than other particles since they can easily penetrate the nuclear potential barrier.

MASS

The mass of the neutron was determined by Chadwick and Goldhaber[6] by photodisintegration of deuterons (heavy hydrogen) in deuterium (heavy water) by gamma quanta of known kinetic energy from thorium to produce a neutron and a proton as given by Eq. 3.1. The kinetic energy of the deuteron may be taken as approximately zero, and the kinetic energy of the proton is measured with an ionization chamber.

$$ {}_1H^2 + \gamma \rightarrow {}_0n^1 + {}_1H^1 \qquad (3.1) $$

The mass of the neutron can then be determined since in a nuclear reaction, as indicated by Eq. 3.1, the total energy is conserved. The mass of the neutron was found to be 1.008937 atomic mass units.[6] Mass (except for small amounts converted into energy) and electric charge are conserved in any nuclear reaction. The mass number of a nucleus is the sum total of the number of protons and the number of neutrons in that nucleus. The charge of the nucleus is of course equal to the number of protons it contains. Because of the conservation mentioned above, the sum of the mass numbers (superscripts) must be the same on both sides of a nuclear reaction equation. Similarly the sum of the charge numbers (subscripts) must balance.

In the preceding equation, a neutron indicated by ${}_0n^1$ has zero charge and a mass number of 1. The deuteron indicated by ${}_1H^2$ contains 1 proton in the nucleus and has a mass number of 2, the sum of the mass of proton plus a neutron.

NEUTRON CAPTURE CROSS SECTIONS

The probability can be calculated that a neutron will interact with a nucleus in an interaction such as one of the following: fission, inelastic scattering or elastic scattering. This probability depends on the product of the neutron flux, ϕ, expressed as incident neutrons per square centimeter per second; N, the number of nuclei of the type considered per cubic centimeter; and a proportionality constant that is the measure of the events per neutron per nucleus. This constant has units of square centimeters and is termed the cross section, σ. For any particular reaction we may write

$$ \left(\text{rate of reaction } \frac{\text{events}}{\text{cm}^3 \cdot \text{sec}}\right) = \left(\phi, \frac{\text{neutrons}}{\text{cm}^2 \cdot \text{sec}}\right) \left(N, \frac{\text{nuclei}}{\text{cm}^3}\right) (\sigma, \text{cm}^2) \qquad (3.2) $$

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A cross section exists for each possible event, namely, σ_f, for fission σ_{sc} for elastic scattering of neutrons, σ_c for the n,γ reaction, etc. The sum of these cross sections gives a total cross section σ_t, or

$$\sigma_t = (\sigma_f + \sigma_{sc} + \sigma_c) \tag{3.3}$$

If the material interacting with neutrons contains nuclei of many species (N_1, N_2, ... N_n), the rate of neutron removal is given by

$$-\frac{dN}{dt} = \phi(N_1\sigma_1 + N_2\sigma_2 + N_3\sigma_3 \ldots N_n\sigma_n) \tag{3.4}$$

The derivative dN with respect to time, t, gives the rate of change of the neutron population and is negative because neutrons are being removed by capture. In this case, N_1, N_2,... N_n represents the number of nuclei of species 1, 2... n, and σ_1, σ_2, ... σ_n the respective total cross section for each species.[7] In a reactor operating with natural uranium, both U^{235} and U^{238} interact with neutrons and, because U^{235} fissions and U^{238} does not, the rate of neutron removal can be expressed as

$$-\frac{dN}{dt} = \phi\,[N_{238}\sigma_c{}^{238} + N_{235}\,(\sigma_c{}^{235} + \sigma_f{}^{235})] \tag{3.5}$$

Giving the cross section σ in square centimeters is not convenient since the areas involved are of the order of 1×10^{-24} cm^2. For convenience, the unit barn is used, where 1 barn is equal to 1×10^{-24} cm^2. Since some materials have very small cross sections, the thousandth part of the barn, the millibarn, equal to 1×10^{-27} cm^2, is also used. Table 3.1 gives absorption cross sections of various isotopes for thermal neutrons.[8]

THE DISCOVERY OF NUCLEAR FISSION

In 1933, one year after the identification of neutrons by Chadwick,[1] artificial radioactivity was discovered by the Joliots[9] when they bombarded aluminum, magnesium, and boron with alpha particles. In the following year, Fermi[10] suggested the possibility of producing elements with an atomic number greater than 92 by bombarding uranium with neutrons. The results of later experiments performed to produce these transuranic elements (elements having an atomic number greater than 92) eventually led to the discovery of nuclear fission.

Fermi and his coworkers discovered in their initial experiments that neutron bombardment of uranium led to a product with four beta-particle activities, each having a different half life. Since naturally radioactive uranium disintegrates by emitting alpha particles, the existence of these beta-particle activities gave evidence of a new process that was believed at the time to be responsible for the formation of one or several transuranic elements. To support this hypothesis, several chemical tests were performed. A solution of uranium salt was bombarded with neutrons and a manganese salt added. Manganese dioxide was precipitated along with a product containing two of the beta-particle activities. Further chemical tests showed that the emitters of these beta particles could not be any element within the atomic number range of 86 to 92. Thus, it was concluded that element 93 had been produced, a chemical homologue of manganese, i.e., an isotope, occurring in the same column of the periodic table.

Noddack[11] criticized this conclusion, suggesting that since many elements were precipitated with manganese dioxide, it was possible that the bombarded nuclei had split into nuclei of elements of lower atomic number. But the criticism was ignored and many workers (especially I. Curie, Hahn, Strassmann and Meitner) began to search for transuranic elements, bombarding uranium, thorium, and protactinium with neutrons. A number of beta particle activities were discovered and checked by chemical and physical methods to discover the nature of the emitters. Until 1939 the emitters of the beta particles were considered to be radioactive substances having an atomic number greater than 92, and several new radioactive series were

Table 3.1—THERMAL-NEUTRON (2200 m/sec) ABSORPTION CROSS SECTIONS[8]*

Element	Isotope	Isotopic abundance, %	Cross section, barns
H (natural)			0.33
	H^1	100	0.33
	H^2	0.015	0.45 mb
He (natural)			Variable
	He^3	0.00013	np 5200
	He^4	1.00	0
Li (natural)			67
	Li^6	7.5	nα910
	Li^7	92.5	33 mb
Be	Be^9	100	9.0 mb
B (natural)			759
	B^{10}	18.8	nα3990
	B^{11}	81.2	50 mb
C (natural)			4.5 mb
	C^{12}	98.9	
	C^{13}	1.1	1.0 mb
N (natural)			1.78
	N^{14}	99.6	np 1.70, nγ0.10
	N^{15}	0.37	0.024 mb
O (natural)			0.2 mb
	O^{16}	99.76	very small
	O^{17}	0.037	nα0.5
	O^{18}	0.20	0.21 mb
F	F^{19}	100	10 mb
Ne (natural)			2.8
Na	Na^{23}	100	0.49
Mg (natural)			59 mb
Al	Al^{27}	100	0.22
Si (natural)			0.13
P	P^{31}	100	0.19
S (natural)			0.49
C_l (natural)			31.6
A (natural)			0.62
K (natural)			1.97
Ca (natural)			0.43
Ti (natural)			5.6
V (natural)			4.7
Cr (natural)			2.9
Mn	Mn^{55}	100	12.6
Fe (natural)			2.43
Co	Co^{59}	100	34
Ni (natural)			4.5
Cu (natural)			3.59
Zn (natural)			1.06
Zr (natural)			0.18
Mo (natural)			2.4
Cd (natural)			2400
In (natural)			190
Sn (natural)			0.65
Xe (natural)			35
	Xe^{135}	0	3.5×10^6
Sm (natural)			6500
	Sm^{149}	13.8	50,000
Eu (natural)			4500
Gd (natural)			44,000
Hf (natural)			115
Ta (natural)			21.3
Au	Au^{197}	100	94
Hg (natural)			380
Pb (natural)			0.17
Bi	Bi^{209}	100	32 mb
Th	Th^{232}	100	7.0
	Th^{233}	0	1400
Pa	Pa^{233}	0	37
U (natural)			nγ3.50, nf 3.92
	U^{235}	0.714	nγ101, nf 549
	U^{238}	99.3	2.80
	U^{239}		22
Pu	Pu^{239}		nγ361, nf 664

*NOTE:
Cross sections are in barns except where otherwise stated. (1 barn = 10^{-24} cm^2)
mb = millibarn = 10^{-3} barns = 10^{-27} cm^2
np = n,p reaction, (absorption of neutron followed by proton emission)
nf = absorption of neutron followed by nuclear fission
nγ = n,γ reaction, (absorption of neutron followed by emission of gamma radiation)
nα = n,α reaction, (absorption of neutron followed by emission of alpha particle)

suggested to account for these findings. All the series suggested began with uranium and went through several disintegrations in which beta particles were emitted. Transuranic elements with atomic numbers as high as 96 or 97 were incorrectly believed to be formed. (It should be mentioned that chemical analysis was particularly difficult as only minute samples of the new radioactive material were available for analysis.) Finally, in 1939, a set of experiments was performed by Hahn and Strassmann[12] that completely proved that the radium series of isotopes were isotopes of barium and the actinium series were isotopes of lanthanum.

These findings required the concept of a completely different type of nuclear reaction resulting from the bombardment of uranium with neutrons. The production of La^{140} (Z = 57) and Ba^{139} (Z = 56) indicated a new process in which the uranium nucleus had split into fragments that were nuclei having intermediate atomic weights. Hahn and Strassmann reasoned that if nuclei with masses of about 140 and Z of about 57 were produced as fragments of uranium that has a mass of 238 and Z of 92, other fragments with masses of about 100 and Z of about 35 should be found. They were able to find Sr^{90} (Z = 38), Y^{90} (Z = 39), and Kr^{85} (Z = 36). Through further chemical experiments, other elements were found including xenon, cesium, bromine, tellurium, iodine, molybdenum, antimony, and rubidium. From this chemical evidence, it was concluded that bombarding uranium nuclei with neutrons caused the uranium nucleus to split into two nuclei of intermediate atomic weights. It was then predicted that the product nuclei would possess large kinetic energies and therefore produce many ion pairs in traveling through a gas. Energies of the fragments were observed[13,14] to reach 100 Mev, which is high compared to the energies of alpha particles. The splitting of the uranium nucleus was proven both chemically and physically and was called fission.[15,16]

THE FISSION PROCESS

The fission processes occurring in a nuclear reactor have been discussed in several texts.[17–22] Briefly stated, the fission process is as follows. A fissionable nucleus becomes unstable after the entry of a neutron. The nucleus may then split into small nuclei, a process in which more neutrons are emitted and a large amount of energy is released. The energy appears in the form of nuclear radiations and as kinetic energy of the fission fragments and neutrons.

If a second fissionable nucleus captures any of the neutrons emitted in this process, then, it too may split, emitting more neutrons which in their turn cause further fissions. Since neutrons may escape from the mass of fissionable material or may be captured by nonfissionable nuclei, more than 1 neutron must be emitted per fission for this series of processes to occur. This series of fissions is termed a chain reaction.

Figure 3.1 shows the fissioning of a U^{235} nucleus after it has become unstable by capture of a thermal neutron. This fission process produces two radioactive fragments and 2 neutrons.

The fission shown schematically in Fig. 3.1 is only one of the many possible ways the excited ($_{92}U^{236}$) nucleus may decay.[23] This neutron capture and decay may also be expressed by the equation

$$_{92}U^{235} + {}_{0}n^{1} \rightarrow (_{92}U^{236}) \rightarrow {}_{42}Mo^{103} + {}_{50}Sn^{131} + 2{}_{0}n^{1} + \text{Energy} \qquad (3.6)$$

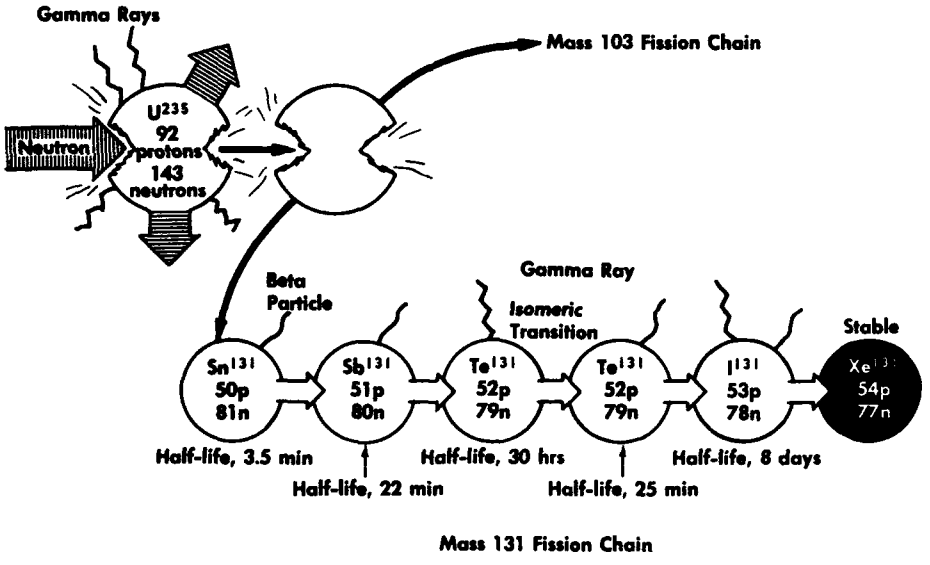


Fig. 3.1—The fissioning of a U^{236} nucleus after it has become unstable by capture of a thermal neutron.

The interaction between the neutron and the uranium nucleus forms an unstable nucleus with a mass number increased by 1 but with no increase in the number of protons, that is, the charge number (atomic number) remains the same.

The molybdenum isotope $_{42}Mo^{103}$, Eq. 3.6, has 42 protons in its nucleus and the tin isotope $_{50}Sn^{131}$ has 50 protons, giving a total of 92—the original number of protons in the uranium nucleus. However, the mass number of the molybdenum isotope is 103, which, added to the tin mass number of 131, gives a total of 234, 2 less than 236, the sum of 235 for the uranium and 1 for the neutron. The equation is balanced by the 2 neutrons released in the fission.

Fission can occur in various nuclides under different conditions. Slow neutrons cause U^{235} to fission, and the fast neutrons, with energies of 1 Mev or greater, produce fission in thorium, U^{235}, and U^{238}. Fission in uranium and thorium may also be caused by bombardment with high energy alpha particles, protons, deuterons, or gamma rays; Pu^{238} and U^{233} are formed by neutron capture by U^{238} and Th^{232} respectively; Pu^{239} and U^{233} will split when bombarded by either slow or fast neutrons.

In the fission process, neutrons and gamma radiation are emitted as well as the two fission fragments. Ternary fission, the division into three fragments, has been observed but occurs only about once per 2×10^5 binary fissions. Only about once in every 400 fissions is an alpha particle emitted. The emission of light nuclei with masses between 4 and 12 is more common, occurring about once in every 80 fissions.

The values for the ratio of neutrons to protons in the stable isotopes of such fission products as krypton, iodine, xenon, and cesium vary from 1.17 to 1.52. Uranium-236, however, has 144 neutrons to 92 protons or a considerably higher ratio of 1.57. Thus, when excited nucleus of U^{236} splits into two smaller nuclei, at least one nucleus may have a greater number of neutrons than is compatible with stability. It has been found experimentally that in order for the unstable nucleus to gain stability, 1 or more electrons, or in a higher energy state, 1 or more neutrons, must be emitted. An example of a beta-particle decay chain, one method of stabilizing an isotope, is given in Eq. 3.7 for the decay of Ba^{141}. Barium-141 is one of the fission products of uranium and has an unstable nucleus since it has 3 extra neutrons. Praseodymium-141 is the final product of this beta-particle decay chain

$$_{56}Ba^{141} \xrightarrow{\beta} {}_{57}La^{141} \xrightarrow{\beta} {}_{58}Ce^{141} \xrightarrow{\beta} {}_{59}Pr^{141} \tag{3.7}$$

A small amount of activation energy is sufficient to fission U^{235} whereas a much greater amount of energy is required to fission U^{238}. The reason for this is related to the number of protons and neutrons in the two different nuclei. Atoms with an even number of protons and an odd number of neutrons are less stable than atoms with an even number of protons and an even number of neutrons. The uranium isotopes $_{92}U^{235}$ and $_{92}U^{233}$ and the plutonium isotope $_{94}Pu^{239}$ having odd numbers of neutrons may be fissioned with neutrons of almost zero energy (thermal neutrons), whereas the isotope of uranium having an even number of neutrons in the nucleus, $_{92}U^{238}$, is much more stable and requires an activation energy of about 1 Mev to cause fission.

Fission probabilities are of theoretical and practical importance. A thermal neutron may be either scattered or captured by the U^{235} nucleus to form the compound nucleus, U^{236}. The compound nucleus may fission or emit gamma radiation. If it emits gamma radiation, the nucleus decays to the ground state of U^{236} and the nucleus in turn emits a 4.5 Mev alpha particle with a half-life of 2.4×10^7 years. Table 3.2 lists thermal cross sections for various reactions in natural uranium, U^{235}, U^{238}, and Pu^{239}.

As mentioned previously the fission reaction shown in Fig. 3.1 and Eq. 3.6 is only one of many ways in which the $_{92}U^{236}$ nucleus can split; studies of yields show that a wide variety of fission products is obtained, ranging from zinc (mass number of 72) to samarium (mass number 158). Figure 3.2 shows a plot of the percentage yield vs. mass number for the fission[24,25] of U^{235} and Pu^{239}

Ninety-seven per cent of the fission products can be classed in either the light group (mass numbers of 84 to 104) or the heavy group (mass numbers of 130 to 149). Products from the most probable kind of fission have mass numbers of from 95 to 134, and these contribute about 6 per cent of the total mass. The yield curve seen in Fig. 3.2 passes through 117 as its minimum, which corresponds to fragments of equal mass. It then drops off rapidly at 72 and 162.

Table 3.2—PROBABILITY OF VARIOUS REACTIONS FOR THERMAL NEUTRONS EXPRESSED IN BARNS[26]

Cross section	Natural U	U^{235}	U^{238}	Pu^{239}
Fission, σ_f	4.18	580	0	750
Radiative capture, σ_c	3.5	107	2.75	3.5
Scattering, σ_s	8.3	9.0	8.3	

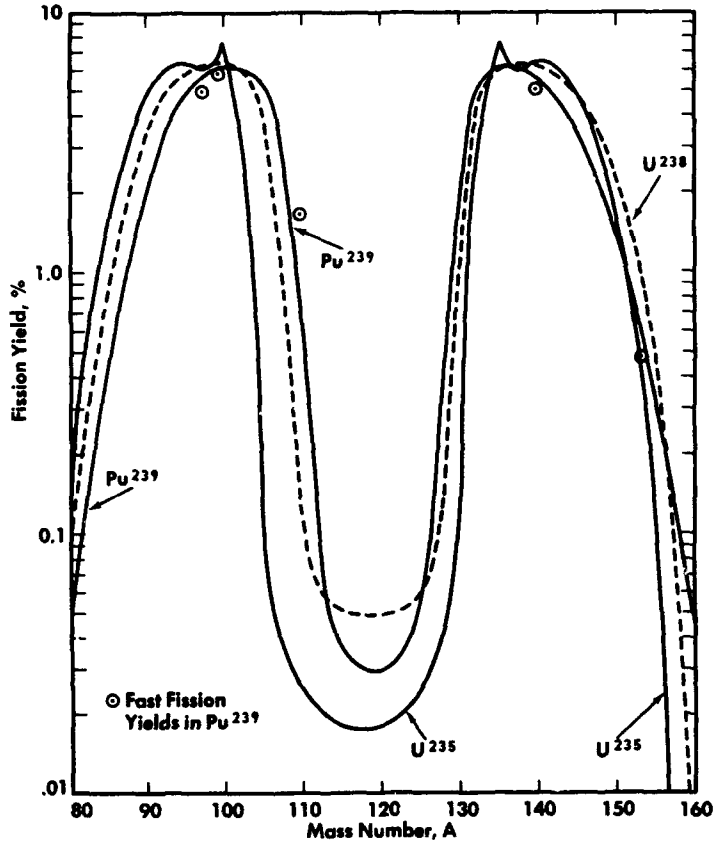


Fig. 3.2—A plot of the percentage yield vs. mass number for the fission of U^{235} and Pu^{239}.

The possible steps in the nuclear fission process were discussed by Bohr and Wheeler.[23] Using the liquid-drop model (Fig. 3.3), they could account for numerous fission properties and successfully predicted the possibility of spontaneous fission. The model also allowed them to predict the ability of a heavy nucleus to undergo fission with slow or fast neutrons.

The shape of the liquid drop nucleus depends on the balance between surface tension forces and Coulomb repulsive forces. The drop may gain excitation energy by capturing a slow neutron. In this instance, oscillations begin within the drop, distorting the spherical shape into an ellipsoidal form. The two forces work against each other as the excitation energy attempts to continue the distortion while the surface tension tends to force the drop back to its original shape. When the excitation energy is large enough, a dumbbell shape is acquired by the nucleus. Then the two end bells may be further pushed apart by the Coulomb repulsive forces, splitting the dumbbell into two spherical fragments. But when the excitation energy is not sufficiently large, the surface tension forces return the ellipsoid to a sphere, and the energy is liberated by emission of gamma photons. This last process is neutron capture rather than fission.

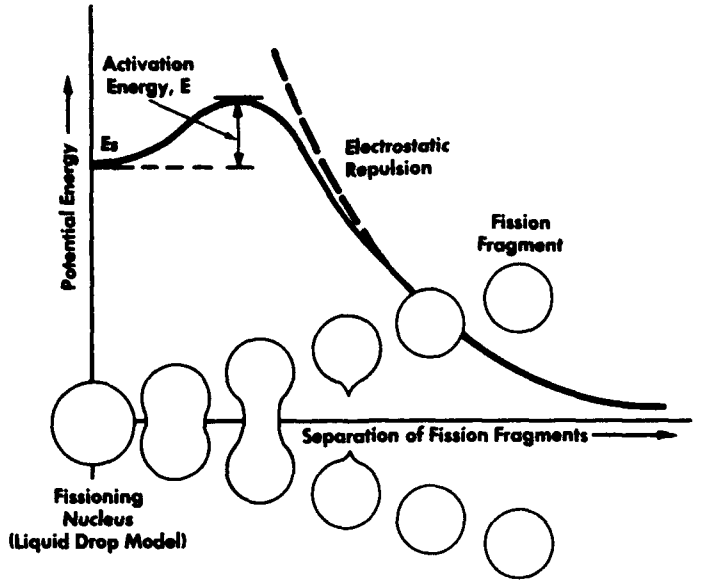


Fig. 3.3—The liquid-drop model.

THE FISSION CHAIN REACTION

In the fission process of the isotopes Th^{232}, U^{233}, U^{235}, U^{238}, or Pu^{239}, more than one neutron is generally emitted per fission. The U^{235} nucleus yields an average of 2.46 neutrons per thermal neutron reacting while Pu^{239} yields about 2.88 and U^{233} about 2.54. The fact that these yields are greater than unity leads to the possibility of a chain reaction in a mass of fissionable material. A nuclear reactor is a system where the arrangement of fissionable and nonfissionable material allows for a controlled fission chain reaction. Contrastingly, the arrangement within an atomic bomb creates a chain reaction that increases at an explosive rate.

The stability, or instability, of a chain reaction depends on the number of neutrons produced in the fission process as opposed to the number of neutrons lost. Fast or high energy neutrons are more difficult to capture and are more easily lost. When a high energy neutron, a product of a previous fission process, strikes a uranium nucleus, the results are often inelastic scattering or a nonfission capture, primarily n,γ reactions. Furthermore, since a fast neutron travels a distance of the order of inches to contact a nucleus, the neutron may escape or leak through the system before it interacts. There is a critical amount of a fissionable isotope required to sustain any chain reaction. For example, no amount of natural uranium by itself will create a chain reaction, because of the predominance of the nonfission capture processes. However, by enriching natural uranium with a little U^{235}, a critical concentration can be acquired for which an infinite mass (practically, this means there is a high ratio of volume to surface) can sustain a chain reaction. For every degree of greater enrichment, there is a corresponding smaller critical mass. Those neutrons that exceed the limit needed to sustain a reaction must be allowed to escape the system or a runaway reaction will result.

If the neutrons from fission are slowed down, they are more easily captured and are more effective in producing a chain reaction. Therefore, one approach to producing a fission chain reaction is to slow down the fast neutrons produced in fission. The probability of interaction increases when the time allowed for the operation of the mutual attraction force increases. Neutron reactions have been found to occur at rates inversely proportional to the neutron velocity.

Hydrogen nuclei have the most efficient mass for slowing neutrons because interaction of equal masses results in transferring about half the energy, the maximum possible, of the impinging neutron. Thus, water is a good moderator and may be used to surround the neutron source and uranium to slow down the high speed neutrons. The process is one of statistical neutron diffusion. Through a series of collisions with hydrogen and oxygen nuclei, the fast neutrons will lose sufficient energy to reach a thermal equilibrium with the moderator molecules. The

neutrons will then have ordinary molecular kinetic energies of only a few thousand feet per second, which will correspond to the velocity of the water molecules at the existing temperature instead of the original velocities of tens of thousands of miles per second.

However, no amount of ordinary water and natural uranium in combination produces a critical chain-reaction condition. The reason is that many neutrons that could produce fission are lost in the following ways: through capture in ordinary hydrogen by the reaction

$$ {}_1H^1 + {}_0n^1 \rightarrow {}_1H^2 + \gamma(2.2\ \text{Mev}) \tag{3.8} $$

and through nonfission resonance capture in uranium. Also, in any finite size system, there is a loss of neutrons through leakage.

In an attempt to minimize nonfission capture, Fermi, Szilard, and Pegram working at Columbia University from 1939 to 1941 used carbon as a moderator.[17] This method of assembling a critical mass requires a large amount of carbon and natural uranium and results in a larger reactor core than the other methods. Moderating by carbon rather than water requires more material because the carbon is only about one-third as effective as the hydrogen (in the water moderator) in slowing down neutrons. Yet, the lower capture cross section of carbon makes a nuclear chain reaction feasible using carbon as a neutron moderator. It was, however, difficult to produce pure graphite and uranium free from the impurities that parasitically capture neutrons. The basic physical data were first obtained from the 8- to 10-ft cubes of graphite-uranium lattice in the test piles set up at Columbia University. With the establishment of the Metallurgical Laboratory at the University of Chicago under the direction of Dr. Arthur H. Compton, Fermi and his associates[17] achieved the first sustained nuclear chain reaction at West Stands, Stagg Field on Dec. 2, 1942.

The ratio of the number of neutrons in one generation to those in the previous generation is called the multiplication factor, k, or

$$ k = \frac{\text{number of neutrons in one generation}}{\text{number of neutrons in the previous generation}} $$

However, because some of the neutrons are lost during slowing down and diffusion, the effective multiplication factor, k_{eff}, is equal to k times p, where p is the probability that the neutrons will not be lost. If k_{eff} is greater than unity, the reaction will be supercritical and will progress at an increasing rate, as in the case of the A-bomb or the case of reactor start-up. On the other hand, if k_{eff} is equal to unity, the reaction is critical and fission occurs at a constant rate, as in a reactor operating at constant power. If k_{eff} is less than unity, the reaction cannot continue, and the condition is termed subcritical. The value of k depends upon the fissionable material used, whereas the value of p depends chiefly upon the size and shape of the fuel assembly.

It is also possible to define the effective multiplication factor, k_{eff}, as the ratio of the production rate of neutrons, P, to the combined rate of absorption, A, and the leakage rate, L

$$ k_{eff} = \frac{P}{A+L} \tag{3.9} $$

where A represents any type of absorption, resulting in fission or just parasitic capture with emission of gamma rays.

If the average number of neutrons emitted per fission is n while F is the fission process rate, then

$$ P = nF \tag{3.10} $$

and Eq. 3.9 can be rewritten

$$ k_r = \frac{nF}{A+L} $$

from which we get

$$k_{eff} = n\left[\frac{F}{A}\frac{1}{1+(L/A)}\right] \tag{3.11}$$

The amount of fissionable and nonfissionable material and the cross sections for fission and neutron capture determine the ratio F/A, while the ability of the fissionable mass to contain and absorb neutrons determines the ratio L/A. The quantity L/A increases without limit when the size of the fissionable mass decreases, thus increasing neutron leakage and decreasing neutron absorption. The limit of k_{eff} therefore decreases to zero. The quantity L/A approaches zero as the fissionable mass increases and k_{eff} increases towards the limiting value nF/A. Thus, when the composition of the fissionable material gives $nF/A > 1$, there exists some size for which $k_{eff} = 1$. The fissionable material is critical at this size so that this size is known as the critical size and the mass at this point is the critical mass.

If a fission chain reaction is not controlled, it progresses at an increasing rate with an almost instantaneous release of vast quantities of radiation and energy. When the reaction is controlled and fission is made to occur at a slower and constant rate, the slower rate of liberation of energy and nuclear radiation allows these to be harnessed for industrial use.

CONTROLLED FISSION

Nuclear fission may be controlled in a nuclear reactor by limiting the number of neutrons available for capture with neutron-absorbing control rods. A reactor consists of the fuel, a coolant, structural frame, control mechanism and, except in the case of fast neutron reactors, a moderator to slow the neutrons to thermal energies.

Since the building of the first nuclear reactor in the form of an uranium-graphite pile in Chicago, many other reactors have been designed and operated throughout the world. The Chicago reactor was followed by a large air-cooled graphite-uranium lattice pile in Oak Ridge, Tenn. This was the first reactor involving major engineering problems of fuel element fabrication and handling, instrumentation and control, and heat transfer and coolant flow. Next, large graphite-uranium piles, cooled with water from the Columbia River, were completed at Hanford Works in the State of Washington. These reactors were not designed to produce high temperatures for the efficient practical use of heat energy, even though the energy liberated was in the hundreds of thousands of kilowatts, but to use the by-product neutrons from the chain reaction to produce plutonium by the reaction

$$_{92}U^{238} + {}_{0}n^{1} \rightarrow {}_{92}U^{239} \xrightarrow{\beta} {}_{93}Np^{239} \xrightarrow{\beta} {}_{94}Pu^{239} \tag{3.12}$$

This use of neutrons for a by-product process is possible because only one of the secondary neutrons emitted per fission is necessary to maintain a chain reaction. The abundant U^{238} isotope captures a neutron to form U^{239} (23-min half-life), which disintegrates in two steps, emitting beta particles to become element 93, Np^{239} (2.3-day half-life) and then element 94, Pu^{239}, an alpha-particle emitter with a half-life of 25,000 years. Plutonium-239 is fissionable with slow neutrons, and this man-made isotope is thus a substitute for U^{235} for power and weapon purposes.

In a nuclear reactor, the core contains the fissionable material. If a suitable type of nonfissionable material surrounds the reactor core, neutrons can be reflected back into the core, reducing both the critical size and mass. However, if the reactor core does not have enough fissionable material or has too much absorbing material, so that $nF/A < 1$, then a reflector is of no use since there will not exist a size for which a chain reaction can be sustained. Also, in a small reactor core, the ratio of surface to volume will be greater than for a larger reactor core, and more neutrons per unit volume can escape without causing fission.

Natural uranium contains only 0.7 per cent fissionable U^{235}, the remainder being U^{238}. Consequently, the capture of neutrons by U^{238} increases the critical mass in a natural uranium reactor, as compared to the critical mass of pure U^{235}. The critical mass and size for natural uranium reactors may be decreased by increasing the percentage of U^{235} in the fuel so as to

provide an enriched fuel.[26] Table 3.3 illustrates the effect of enrichment and choice of moderator on minimum reactor size.[27]

The process of enrichment is expensive because the two isotopes of uranium have identical chemical properties and therefore cannot be separated chemically. Large and elaborate gaseous diffusion plants are used for such enrichment. The operation of these plants depends on the fact that there is a minute difference in the diffusion constants of the gaseous hexafluorides of U^{235} and U^{238} because of the slight weight difference. Uranium-238 after neutron capture, may react according to Eq. 3.12 to produce Pu^{239}, which is fissionable, but this conversion consumes neutrons. In addition to the neutrons lost by the processes already mentioned, the coolant, structural members, and the control system will also capture neutrons. Indeed, conserving neutrons is one of the principal problems in a reactor design.

Table 3.3—REACTOR CORE SIZES AND MATERIALS FOR 100 Mw (HEAT) REACTORS[27]

Fuel	Approx. smallest dimension, ft*	Moderator	Coolant	Structural materials
Natural uranium	25	Graphite	Ordinary water	Aluminum, zirconium
	10	Heavy water	Ordinary or heavy water	Aluminum, zirconium
Slightly enriched uranium	8	Graphite	Sodium	Zirconium, stainless steel
	8	Graphite	Ordinary water	Aluminum, zirconium
Highly enriched uranium	4	Ordinary water	Ordinary water	Aluminum, zirconium
	4	Beryllium	Ordinary water	Stainless steel
	4	None	Sodium	Stainless steel

*Reactor cores have various shapes: natural uranium ones are usually in the form of a regular cube; other cores are arranged as a rectangular cube or a right cylinder.

By controlling the number of neutrons present in the reactor core, it is possible to control the power level of a reactor since the number of neutrons in the core is proportional to the number of fissions per unit time, and the fission rate is proportional to the power level. Boron, enclosed in a steel rod, may be introduced into the reactor as a neutron absorbing material. By adjusting the position of the rod within the reactor core, the value of k_{eff} can be changed in the desired direction. During reactor operation, these control rods must be moved out of the reactor at a very slow rate to keep the power level constant since the fissionable material is consumed while the reactor operates, decreasing the F/A ratio. When all control rods are out and the compensation limit reached, new fissionable material is required, or the reactor will become subcritical and stop.

In a nuclear reactor the neutron decay process claims only a negligible fraction of the neutrons produced. Although the neutron is unstable, decaying with a half-life of about 12 min, the time interval, or time of a neutron generation between the emission of a neutron through the fission process and its subsequent absorption or leakage, is usually smaller than milliseconds. Thus, the neutrons usually either escape or are captured before they have time to decay.

Both neutron-fission cross sections and absorption cross sections are complex functions of the neutron energy. Neutron energy is governed by the neutron fission spectrum, by the elastic and inelastic scattering cross sections of neutrons, and particularly by the size of the reactor. It is therefore difficult to obtain the effective multiplication constant, k_{eff}, for a given distribution of fissionable material within the reactor.

In a natural uranium graphite-moderated reactor operated at a constant power level, the processes that occur in one generation of neutrons can be considered. If the natural uranium captures 100 neutrons, the U^{235} will capture thermal neutrons primarily, and neutrons of other

energies will be captured both by U^{235} and U^{238}. The capture of thermal neutrons in U^{235} will result in fission and the production of about 256 new neutrons. If the reactor is critical, this process continues as a chain reaction with a distribution of neutron interactions in a typical reactor (basis 100 neutrons) such as listed in Table 3.4.

Table 3.4—DISTRIBUTION OF NEUTRON INTERACTIONS IN A THERMAL NEUTRON NATURAL URANIUM REACTOR

Type of interaction	Number of neutrons
New fissions (primarily U^{235})	39
Radiative capture by U^{238}	35
Radiative capture by U^{235}	8
Absorbed by moderator	12
Absorbed by structural material	2
Escaped from core	3
In excess (either absorbed by control rods or available for increasing power level)	1
Total	100

THE ENERGY OF FISSION

The fission process creates fragments emitted at all angles with tremendous kinetic energies. The fission of U^{235} may be considered as an example of the large amounts of energy released. Uranium-235 has an atomic number, Z, equal to 92 and a mass, A, equal to 235. A typical pair of fission products are Ba^{141}, with Z = 56, and Kr^{92}, with Z = 36. During this fission process there is an immediate release of 3 neutrons, so that the reaction can be written in the equation

$$_{92}U^{235} + {}_{0}n^{1} \rightarrow {}_{92}U^{236} \rightarrow {}_{36}Kr^{92} + {}_{56}Ba^{141} + 3{}_{0}n^{1} + E \qquad (3.13)$$

where E is the energy released. The energy, E, is obtained by subtracting the mass of the final products from the mass of the initial reactants. The difference in mass in this reaction is 0.2154 amu, so that E = ~200 Mev (1 amu = 931 Mev). By way of comparison, the energy released per atom in chemical reactions, such as combustion, is only a few electron volts, and the energy from alpha-particle disintegration averages from 5 to 10 Mev. Thus, atom for atom the fission energy is about a hundred million times larger than the energy of a chemical reaction and 20 to 40 times larger than the released energy in alpha-particle disintegration.

Usually only two fragments are produced in the fission of a nucleus. These two particles have equal but opposite momentum and kinetic energies, which are in the inverse ratio to their masses. Ionization methods, along with some infrequently used calorimetric methods, have been used to measure kinetic energy of fission fragments and the distribution of energy among fragments. The energy distribution for both slow and fast neutrons for fission fragments of U^{235} was obtained by Fowler and Rosen.[28] Using a specially designed ionization chamber, they obtained the graph shown in Fig. 3.4 for energy distribution in fast neutron fission. Studies with slow neutrons showed two distinct peaks at 61.4 and 93.1 Mev. Fast neutrons having energies of 0.001 to 1 Mev produced fission neutrons having a similar curve of energy distribution but with peaks having slightly different energies. It was assumed, in determining the kinetic energy from the ionization current, that a fission product transfers the same amount of energy as an alpha particle in producing an ion pair in a gas. This assumption, however, introduces some error.

In another study of energy distribution of fission fragments, Leachman used the time-of-flight method.[29] In this procedure thermal neutrons a[illegible]ed to irradiate a thin foil having a film of either U^{233}, U^{235}, or Pu^{239}. Scintillation detec[illegible]ntify the pairs of fragments. One

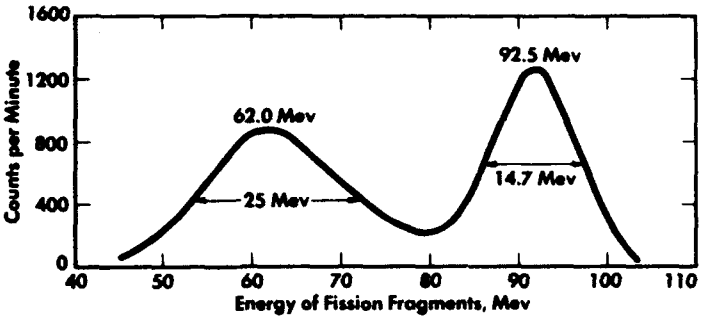


Fig. 3.4— Energy distribution in fast neutron fission.

fragment passes along several hundred centimeters of an evacuated tube before it strikes the detector, while the other fragment travels only 1 cm and strikes the detector. The screen of a cathode-ray tube picks up the pulses from both detectors, and these pulses are photographed. Then the distance between the two peaks gives the fragment velocity, and the kinetic energy is calculated from the velocity. Energies calculated using time-of-flight have larger values than those determined by the ionization method. The ionization method gives an average total kinetic energy of about 155 Mev; the time-of-flight method gives about 167 Mev.

Table 3.5 shows the estimated distribution of the energy of a typical fission reaction in which 162 Mev is taken as the kinetic energy of the fission fragments.

Converting fission energy to electrical energy is another major problem. There is no method known by which the tremendous kinetic energy of the fission fragments may be transformed directly into useful work or electrical energy. Likewise, the nuclear radiations released at the time of fission cannot be converted directly into electrical energy in the reactor. While nuclear radiations from radioactive material can be converted directly into electrical energy, the conversion efficiencies are low. (See discussion of electric storage batteries in Chap. 5). The kinetic energy of a fission fragment can be dissipated into heat, the lowest form

Table 3.5— ENERGY DISTRIBUTION OF A TYPICAL FISSION REACTION[8]

Energy distribution	Mev per fission	Kilowatt hr per fission
Energy available* immediately after the fission process		
Kinetic energy of fission fragments	162	
Kinetic energy of prompt neutrons produced in the fission process	6	
Energy of instantaneous gamma photons	6	
Energy from absorption of excess neutrons captured in nonfission processes by reactor materials	8	
Total	182	8.08×10^{-18}
Energy available from decay of the fission products		
Energy from fission product gamma radiation	5	
Energy from fission product beta particles	5	
Total	192	8.52×10^{-18}
Unavailable energy		
Energy carried away by neutrinos accompanying the fission product beta decays	11	0.488×10^{-18}

*Or potentially available if use is made of the heat generated in the shield by absorption of gamma radiation.

of energy, if the fragments are decelerated through interaction with other atoms in the reactor core. On conversion of heat to higher forms such as electrical or mechanical energy, an appreciable portion will be lost. Beta-particle nuclear radiations also are degraded in energy by deceleration and absorbed in the core of the reactor, and the energy of these radiations is converted to heat. Gamma radiation is more penetrating and is absorbed to a lesser extent by interaction of gamma photons with atoms in the reactor core. Much of the gamma radiation is absorbed in the heavy shield surrounding the reactor.

REACTOR TYPES

The general types and characteristics of reactors have been discussed in various texts and articles.[17–22,30–35] For example, power reactors are designed for operation at high temperature and for utilization of the heat they produce whereas research reactors operate at low temperature, provide for utilization of their neutrons, and waste their heat.

Reactors may also be classified by the type of fuel. Liquid fuel is homogeneous, and a reactor using liquid fuel is termed a "homogeneous" reactor. When the fuel is solid, it is usually contained in a structure called a fuel element. This type of reactor is termed "heterogeneous" because the core contains not only the fuel element but also structural materials and coolant.

Most power reactors use fuel elements and therefore are in the heterogeneous class. Examples are the Shippingport Pressurized Water Reactor, the first full scale nuclear power plant in the United States, experimental boiling water reactors at the Argonne National Laboratory and at Vallecitos, Calif., a sodium graphite reactor near Los Angeles, and the British Calder Hall Reactor. The graphite reactors and the widely used pool type research reactor are also heterogeneous.

There are a number of homogeneous research reactors—the first being built at the Los Alamos Scientific Laboratory in 1944. A small experimental homogeneous power reactor was operated at the Oak Ridge National Laboratory in 1952, and a larger one is being built. The fuel of these reactors is uranium sulfate dissolved in water.

Another method of classifying reactors is according to the energy range of most of the neutrons in the core. Reactors then fit in three categories: thermal, intermediate, and fast.

Reactors are also classified according to their production of new fissionable material from the fertile material they may contain. If a reactor is operated to convert either U^{238} to Pu^{239} or Th^{232} to U^{233}, it may be termed a "converter." If more fissionable material is produced than is consumed, it is also a breeder.

Conversion, in the case of uranium and plutonium, makes possible the chemical isolation of the fissionable material, which can then be used for weapons or to enrich reactor fuel. The plutonium production reactors at Hanford are prominent examples of reactors built and operated for conversion.

It is important that breeder reactors, or converters with a high conversion ratio, come into use before many years so that the extensive supply of fertile materials will constitute an energy source as well as the much smaller amounts of fissionable materials. Use of fertile material as a source of fuel will require repeated recycling through the chemical processing of spent fuel elements, reconstitution into metal, and fabrication into new elements. There are losses of the valuable fissionable materials in each operation. It is important to recognize that the gain per cycle is small, and the time required to double the amount of initial fissionable material is long.

Breeding was demonstrated on a small scale in 1951 by the experimental breeder reactor at the National Reactor Testing Station. This reactor also produced the first electricity from nuclear fuel when in December 1951 it operated with an output of 100 electrical kilowatts. Larger experimental breeder plants are being built in England and the United States.

NEUTRON ENERGY

The production of neutrons is the key to the fission reaction, and neutron energy is an important consideration in reactor design. Two types of neutrons can be emitted in the fission

process: prompt neutrons, which constitute 99 percent, and delayed neutrons. The emission time for the prompt neutrons is possibly as small as 10^{-14} sec and the energies vary from less than 0.05 Mev to more than 17 Mev. Delayed neutrons may require several minutes for emission. Neutrons from thermal fission of U^{235} have been studied to determine the spectrum of neutron energy. In some of these investigations, a cloud chamber filled with hydrogen gas and water vapor was used to determine the energy[36] of neutrons in the low energy range of 0.05 to 0.7 Mev.

In order to study neutron energies in the energy range between 0.4 and 17 Mev, Watt[37] and Hill[38] used coincidence counters to measure the range distribution of recoil protons from hydrogenous materials. A broad maximum for neutron intensity is found near 0.75 Mev. The relative number of neutrons then decreases exponentially as the energy declines to 2 Mev. The energy distribution between 0.075 and 17 Mev is described in the empirical formula

$$N(E) = C \sin h\,(2E)^{1/2}\, e^{-E} \tag{3.14}$$

where N(E) is the relative number of neutrons per unit energy range as a function of the neutron energy. Figure 3.5 gives the experimental results, which show good agreement with Eq. 3.14. The average energy value of the neutron is 2.0 ± 0.1 Mev.

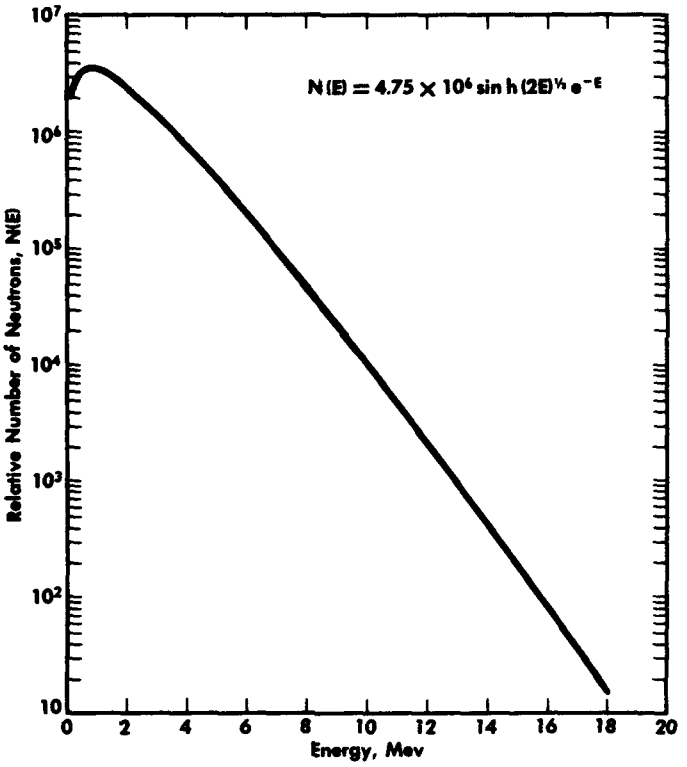


Fig. 3.5—Experimental results in energy distribution.

Table 3.6—REACTOR CLASSIFICATION BY NEUTRON ENERGY

Energy region	Approximate range of energy, ev	Approximate average energy, ev	Velocity of neutron of average energy, ft/sec
Thermal	10^{-3} to 10^{-1}	0.025	7,200
Intermediate	10^{-3} to 10^{-5}	10,000.00	1,400,000
Fast	(1 to 15) × 10^{6}	1,000,000.00	45,000,000

Nereson[39,40] studied a similar energy range of Pu^{239} and U^{235}, using emulsion plates. He was interested in the energy spectra of 0.5 to 8 Mev. His findings agree with the above results and with Eq. 3.14. He further showed that neutrons from Pu^{239} had a maximum energy in the 0.6 to 0.8 Mev region.

Table 3.6 gives the typical neutron energies and velocities for the three energy regions common to reactors.

Thermal neutrons, while diffusing through a weakly absorbing medium, obey very closely the Maxwellian velocity distribution indicated in Fig. 3.6.

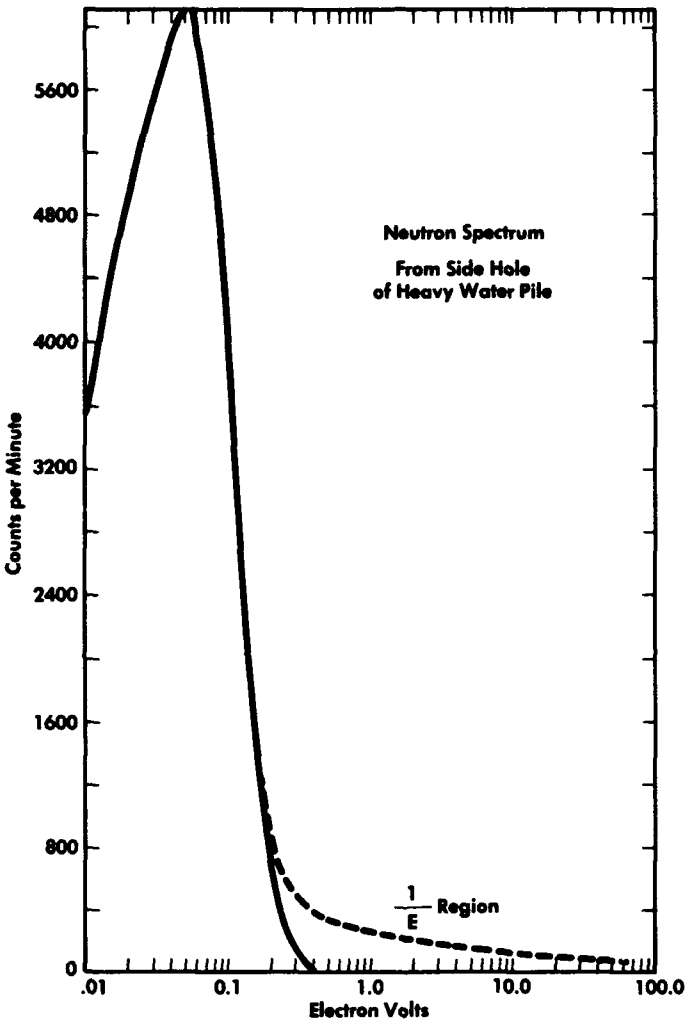


Fig. 3.6— Maxwellian velocity distribution.

If the Maxwellian distribution equation is differentiated with respect to velocity v and equated to zero, the maximum of the curve shown in Fig. 3.6 can be obtained. This maximum gives the most probable neutron velocity, v_m, and this is equal to

$$v_m = \frac{2kT^{1/2}}{M} \tag{3.15}$$

where T = absolute temperature of the material with which the neutrons are in thermal equilibrium, °K

k = Boltzmann's constant
= 1.38×10^{-16} erg/deg
M = neutron mass

The kinetic energy corresponding to the most probable velocity at a particular temperature T is kT, but the average kinetic energy of the neutrons is equal to $\frac{3}{2}$ kT. The velocity corresponding to energy E is obtained from the kinetic energy relationship $E = \frac{1}{2} mv^2$, which gives

$$v = (1.4 \times 10^6 \sqrt{E}) \text{ cm/sec} \tag{3.16}$$

where E is in electron volts.

Most of the reactors to date have operated with thermal neutrons. These reactors have the advantage of a small critical mass with a correspondingly small inventory cost. Nevertheless, thermal reactors have a number of disadvantages. The principal disadvantage is the amount of moderator required, which, in the case of heavy water, may be quite expensive. Slow neutrons are more easily captured by structural materials and by coolants, and the temperatures may be low for maximum thermal efficiency if water is the coolant. Steel and other structural materials capture thermal neutrons readily. Therefore, to maintain good neutron economy, other metals such as aluminum or zirconium, which have a low capture cross section for thermal neutrons, must be used in the core of the reactor and for cladding fuel elements. For this purpose, aluminum has the disadvantage of a limited service temperature, and zirconium is very expensive. Another disadvantage of the thermal-neutron reactor is that it cannot breed on the U^{238}-Pu^{239} cycle. However, a thermal-neutron reactor can be operated as a converter to produce Pu^{239} from natural uranium by the use of an efficient moderator.

Many future reactors are expected to operate with fast neutrons. Since most materials have smaller capture cross sections for fast than for thermal neutrons, a much greater selection of coolants and structural materials is permitted for fast reactors. Liquid metals may be used as coolants, and materials that will withstand high operating temperatures, such as stainless steel, may be used for structures and cladding. These higher operating temperatures permit higher thermal efficiencies in the production of power. A fast reactor has the further advantage that it can operate as a breeder on the U^{238}-Pu^{239} cycle. Some of the disadvantages are the requirement for a relatively large critical mass of fuel, the difficulty of working with liquid metals, and greater difficulty of control of fast-neutron as compared to thermal-neutron reactors.

The Enrico Fermi atomic power plant, being constructed at Monroe, Mich., by the Power Reactor Development Co. is to be a fast-neutron breeder-type power reactor.[31,42] The plant is to develop about 300 Mw of heat and produce about 100 Mw of electricity.

The intermediate reactor operates, as the name implies, with neutrons of energy between the thermal and the fast ranges. The critical mass for an intermediate reactor is larger than that for the thermal reactor but less than that required for the fast reactor. A wider selection of materials for construction and as coolants is permissible than with thermal neutron reactors. Liquid metals can be used as coolants, permitting higher thermal efficiency.

The submarine intermediate reactor, the initial power plant for the *Seawolf*, operated primarily on neutrons in the intermediate neutron energy range.

MODERATORS

A moderator is used in thermal reactors to decrease the energy of the neutrons born at fission at energies of about 1 or 2 Mev down to about 0.025 Mev. The ideal moderator should perform this function without capturing the neutron. If the moderator has a large mass number, it will not be very efficient in absorbing energy from the neutrons. This may be illustrated by considering a rubber ball thrown against a solid wall. In such a case the ball rebounds with nearly all the energy it had in striking the wall. However, if a ball strikes another ball of comparable size and elasticity, it may transfer a large portion of its energy to the second ball as kinetic energy. Thus, the ideal moderator should have a mass that is not very much larger than that of a neutron.

Atoms of low mass include hydrogen, helium, lithium, beryllium, boron, carbon, nitrogen, and oxygen. These are listed in the order of their efficiency in slowing neutrons. Four are gases at standard conditions and do not have sufficient density to provide adequate moderation

of neutron energy in the available core volumes of a reactor. However, helium under high pressure might be a satisfactory moderator. Both lithium and boron capture neutrons readily and so they are unsatisfactory as moderators. Consequently, in the elemental state, only carbon and beryllium may be used as moderators. Beryllium has the disadvantage of being rather expensive and of having undesirable physical properties. The diatomic gases can be used in the combined state, when they may exist as liquids or solids. Hydrogen and oxygen in the form of water may thus be used as a moderator. Normal hydrogen captures neutrons, becoming heavy hydrogen, so heavy water will obviously be a more efficient moderator than ordinary water. If enriched fuel is used, neutron absorption by plain water may be tolerated; but if natural uranium is used, heavy water must be used. The principal disadvantage of heavy water is its high cost.

The Materials Testing Reactor (MTR) has highly enriched uranium in the fuel elements permitting use of ordinary water as the moderator and coolant.[43,44] However, the reflector surrounding the core of the MTR contains beryllium, which has a much lower capture cross section for thermal neutrons than water. The requirements for a good reflector are essentially the same as for a moderator: the reflector should have a high cross section for neutron scatter so as to provide good reflection of escaping neutrons back into the core without neutron capture.

Fast neutrons suffer elastic collisions with the atoms of the moderator and are reduced to thermal energies in about 10^{-4} sec, assuming the inelastic scattering contribution to be very small. After this slowing down, the thermal neutrons diffuse in the medium for about 10^{-3} sec before being captured or escaping out of the assembly.

The neutron can lose most of its energy in a head-on collision with light nuclei. The average number of collisions required to reduce neutrons of energy E to thermal energy is

$$n = \ln\left(\frac{\frac{E}{0.025}}{\xi}\right) \tag{3.17}$$

where ξ = average logarithmic decrement per collision.

It is interesting to note that the average logarithmic decrement in energy is independent of the initial energy. For values of $M > 10$

$$\xi \doteq \frac{2}{M + 2/3} \tag{3.18}$$

The average number of collisions required to reduce neutrons having an energy of 2 Mev to thermal energy (0.025) is obtained from Eq. 3.17 and is

$$n = \ln\left(\frac{\frac{2 \times 10^6}{0.025}}{\xi}\right) = \frac{18.2}{\xi} \tag{3.19}$$

In Table 3.7 this average number of collisions is given.[21]

Inspection of Table 3.7 shows that light hydrogen should be a good moderator but in fact only those substances that have a large moderating ratio $\xi(\sigma_s/\sigma_a)$ are good moderators, for example, light hydrogen has a large scattering cross section and a large value of ξ but also has a large value of absorption cross section (σ_a), making it a poor moderator in comparison with deuterium. Table 3.8 gives the moderating ratio and the scattering and absorption cross sections for some of the important moderators.[21]

COOLANTS

Most reactors in operation today use thermal neutrons. This leaves the coolant as the primary variable, and classification of reactors by the nature of the coolant is common. Most of the cooling systems are described in this section. Succeeding chapters will suggest modifications of some of these types for various industrial applications.

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Table 3.7—COLLISIONS REQUIRED TO THERMALIZE NEUTRONS WITH VARIOUS MATERIALS[21]

Material	Mass No.	ξ	Collisions to thermalize 2-Mev neutrons
Hydrogen	1	1.000	18
Deuterium	2	0.725	25
Helium	4	0.425	43
Lithium	7	0.268	67
Beryllium	9	0.209	87
Carbon	12	0.158	114
Oxygen	16	0.120	150
Uranium	238	0.00833	2150

Table 3.8—MODERATING RATIO AND CROSS SECTIONS FOR SOME MODERATORS[21]

Moderator	Moderating ratio, $\xi \frac{\sigma_s}{\sigma_a}$	σ_a, barns	σ_s, barns
Water	72	0.32	20–80
Heavy water	12,000	9.2×10^{-4}	15.3
Helium	83	8.0×10^{-3}	1.55
Beryllium	159	9.0×10^{-3}	6.9
Carbon	170	4.5×10^{-3}	4.8

WATER-COOLED REACTORS

The simplest heterogeneous water-cooled reactor is the pool type research reactor. It consists essentially of a core of aluminum-clad plate type fuel elements at a depth of about 20 ft in a pool of water. At low power, heat is removed by natural convection. In pool reactors of moderate power, the water is circulated by a pump below the core; and for installations that operate at higher power, forced circulation through the core and through an external heat exchanger is necessary. For still higher powers, the sides of the pool are in effect moved in toward the core until the pool becomes a compact vessel. The vessel is sealed to contain the low pressure set up during forced circulation of the water through a heat exchanger. The sealing also confines short-lived radioactive nitrogen produced by the high flux. The Engineering Test Reactor at the National Reactor Testing Station in Idaho is the largest reactor of this type. It operates at a power level of 175,000 kw of heat and produces a maximum thermal flux of 6×10^{14} neutrons/cm^2/sec.

Most water-cooled research reactors use ordinary water and hence require enriched uranium. It is possible to use natural uranium only if heavy water is both moderator and coolant. The Canadian NRX and a number of European research reactors employ these materials.

In research reactors the temperature of the water is kept well below the boiling point. For generating electric power, the system is pressurized to about 2000 psi and operated at a temperature of 500°F. The first plant of this kind was the prototype for the submarine *Nautilus;* the largest is the Shippingport central station.

The aqueous homogeneous research reactor is also water cooled; the fuel, uranyl sulfate, is in a water solution. Sometimes the heat in the core is removed by water of a separate system that circulates through a tube in the core. In other research reactor installations, the fuel solution is pumped from the core through an external heat exchanger. An external exchanger is also used for the aqueous homogeneous power reactor which, like the Shippingport reactor, operates at high pressure so that temperature sufficiently high for power generation can be reached

ORGANIC LIQUID-COOLED REACTORS

An organic-moderated reactor experiment is being operated at the Testing Station in Idaho to test the value of diphenyl as a reactor coolant. Diphenyl is attractive because it has a high boiling point, which means that high pressurization is not necessary to reach efficient power plant temperatures. Disadvantages include decomposition of the organic compound by radiation, low heat transfer coefficient, and hazard of fire and explosion.[50,51]

BOILING WATER REACTORS

A boiling water reactor is cooled by steam produced in the pressure vessel containing the core. A high-pressure water cooling system and external boiler are not required, which greatly simplifies the design. Experimental plants are operating in Idaho, at Argonne National Laboratory, and at Vallecitos, Calif.; also, a large plant is under construction near Chicago.

LIQUID-METAL-COOLED REACTORS

Sodium is the most used of the liquid metal reactor coolants. Its boiling point of 1600°F makes possible high-temperature operation with only the pressurization necessary to circulate the coolant. Unfortunately radioactivity induced in sodium requires considerable shielding and makes maintenance difficult. Also, care must be taken to avoid any possibility of chemical reaction between sodium and water. Because of radioactivity, the first sodium reactors are designed with an intermediate sodium heat transfer loop so that nonradioactive sodium will carry heat into the boiler where steam is generated.

A small experimental sodium-cooled, graphite moderated thermal power reactor is in operation near Los Angeles; a large fast breeder plant, also cooled by sodium, is being built near Detroit.

A liquid-metal-cooled homogeneous reactor, with fuel dissolved in the coolant, is being developed by the Brookhaven National Laboratory and others. The fuel-carrying coolant is the metal, bismuth. High-temperature operation and breeding U^{233} from thorium are among the potential advantages.

GAS-COOLED REACTORS

Air-cooled open-cycle reactors, such as the Oak Ridge and Brookhaven graphite reactors, are used for research. Air from the atmosphere is drawn through these reactors and then discharged to the atmosphere. Reactors of this basic type could be useful in supplying process heat.[45,46]

Air-cooled reactors have a number of disadvantages, such as poor heat-transfer coefficients of air and the induced radioactivity in the air. Also, the high pressure drop necessary to force sufficient air through the reactor at high velocities requires a large amount of energy for pumping.

The Brookhaven reactor, the largest of this type, uses filtered air at the rate of 1,500,000 lb/hr. Graphite-moderated, air-cooled natural uranium research reactors were among the first built in the United States; but because of their large size and cost and because of the availability of enriched uranium, it seems improbable that more will be constructed here.[46]

Until 1958 the reactor was loaded with 50 tons of natural uranium and had a power level of 28 Mw of heat; now it is being gradually charged with enriched fuel elements.

In closed-cycle, gas-cooled reactors, the gas is constantly circulated from the reactor through a heat exchanger and back to the reactor. The British Calder Hall Reactor, using carbon dioxide, is of this type.[48,49] In this country helium is seriously considered as a coolant.

Induced radioactivity in helium as a result of neutron capture is a minor problem and, therefore, no secondary shielding would be required around a gas turbine driven with compressed helium from a helium-cooled reactor.[47] As this secondary shield may represent a sub-

stantial fraction of the total shielding required for a reactor, there is considerable advantage to a coolant that does not become radioactive. A helium-cooled reactor would require only about two-thirds the shielding of a comparable pressurized-water-cooled reactor and about half the shielding of a comparable liquid-sodium-cooled reactor.

A disadvantage of using helium is the requirement of a large number of turbine and compressor stages as compared to a system employing a heavier gas, such as carbon dioxide, nitrogen or air. Also, to obtain suitable heat transfer across the reactor, it may be necessary to resort to intercooling and regeneration, which complicate the design. The power needs for pumping large volumes of helium is another disadvantage.

The volume of helium circulated can be reduced by increasing the pressure, but a compromise must be reached among requirements for maximum gas density, minimum equipment volume, and permissible shell thickness for large-diameter vessels. The pressure would probably be in excess of 1000 psi; and such a high pressure raises some other problems, such as those of diffusion of helium through vessel walls and the difficulty of remotely controlled unloading of the fuel elements from the pressure vessel.

Carbon-dioxide-cooled closed cycle reactors have been shown considerable favor in England, which has an extensive program[49] that includes 17 large reactors scheduled to go into operation between 1956 and 1965.

The Calder Hall reactor, completed in 1956, is graphite moderated and produces 50 Mw of electricity. The core is contained in a vessel 40 ft in diameter and 60 ft high, fabricated of 2-in. thick welded boiler plate steel. More than 1000 tons of graphite are required for the moderator; 20 tons of carbon dioxide are circulated through the reactor and heat exchangers. Steam is generated by transfer of heat from the carbon dioxide in four boilers 18 ft in diameter and 70 ft high. Four 2000-hp blowers circulate the carbon dioxide.[48]

To assist the reader who may desire more detailed information on some reactor projects, additional references are given.[50–154]

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Chapter 4

REACTORS AS INDUSTRIAL SOURCES OF RADIATION

Nuclear reactors produce tremendous quantities of nuclear radiations. The gamma flux in a reactor core has a much higher intensity than that produced by any other controllable means. If powerful sources of ionizing radiation are needed for industrial uses, nuclear reactors warrant major consideration.

This chapter discusses proposed industrial applications of reactor irradiation, along with the chemical reactions that can proceed in a reactor core, and the possibility of direct conversion of fission energy to chemical energy. Also considered will be some of the problems of food irradiation; the possibility of using fission gases or liquid fuels as gamma sources; the use of reflector solutions in gamma radiation facilities; and the production of the gamma emitter in the core, in various types of blankets, and in a sodium coolant.

Perhaps the soundest economical application would be to provide combined nuclear power, process heat and/or gamma radiation. With such a multipurpose reactor, the cost of gamma irradiation should be based on the difference between power costs from the nuclear plant and those from a steam plant. Since the costs of power from these two sources are expected eventually to compete (without considering other factors), by-product gamma radiation may become very cheap for industrial processing.

Radioactive liquid fuel can be circulated to provide gamma sources in the same way as radioactive fission gases; this fact and attendant problems are discussed. Also discussed is alternative use of a solid fuel element reactor, with radioactive liquid metal coolant pumped into an adjacent facility for use as a gamma source. In addition, there are diagrams and discussion of a multipurpose aqueous homogeneous reactor and details of a multipurpose liquid-metal-fuel reactor (LMFR). The chapter considers problems of the fission gas radiation source, and discusses multipurpose use of an LMFR in a rubber tire plant.

RADIATIONS FROM THE REACTOR CORE

A reactor is an intense source of nuclear radiations—primarily beta particles, gamma rays, and neutrons. In power reactors, these radiations are ordinarily dissipated as heat which can be used directly in process industries or converted into electricity. The radiations might also be utilized directly, however, in promoting chemical reactions, sterilizing medical supplies, or irradiating food.

About half the gamma radiation produced in a reactor results from prompt fission gammas, or from gammas released as a result of neutron capture by different materials in the core.

Using the reactor core as a radiation source, neutron radiation takes place simultaneously with gamma radiation. This, however, may not be a disadvantage if when bombarded by neutrons the material to be irradiated does not become significantly radioactive. Hydrocarbons such as ethylene, or mixtures of two pure gases such as nitrogen and oxygen, may be exposed

to both gamma radiation and neutrons to promote a specific chemical reaction since neutron capture by such simple systems usually does not produce radioisotopes with long half-lives. While the neutron radiation may not contribute materially to the yield of the desired chemical product, it presents in such applications no serious problem of radioactive contamination.

Gamma radiation accompanied by a comparatively low neutron flux might be utilized successfully for sterilization.[1] Energy from gamma radiation, neutrons, and fission fragments, has been used in promoting chemical reactions.[2] The direct transformation in the reactor core of the kinetic energy of fission fragments and gamma and beta radiation into chemical energy might be a promising method of using this energy without complex apparatus for heat transfer at high temperatures.[3] This direct transformation, however, introduces many difficulties that must be overcome before reactors can be designed solely for direct conversion of fission energy to chemical energy.

REACTOR CORE PROMOTION OF CHEMICAL REACTIONS

ADVANTAGES OF CONVERTING FISSION ENERGY TO CHEMICAL ENERGY

Certain chemical reactions may be promoted by one or more of the nuclear emissions such as the neutrons and alpha, beta, and gamma radiations. Also, the tremendous kinetic energy of the fission fragments might be utilized to some extent to promote chemical reactions.[2] This use of the kinetic energy of fission fragments indicates new industrial possibilities for nuclear reactors.

The conventional steps in converting heat to electrical and then chemical energy through the promotion of a chemical reaction by an electrical potential, have a maximum efficiency of only about 10 per cent.[2] Direct conversion of the kinetic energy of fission to chemical energy can possibly bypass the costly intermediate stage of electrical energy production. Nuclear reactor radiation can ionize and excite molecules, promoting certain chemical reactions; but a relatively large dose of gamma radiation is equivalent to only a small amount of energy (1 r, or 2.22×10^{-6} cal). Therefore, gamma radiation alone has not usually been considered as an energy source for endothermic reactions but, rather, as a special catalyst to "trigger" or promote exothermic reactions. If, however, the reactions were conducted in a nuclear reactor, where large quantities of heat as well as high concentrations of ionizing radiation are available, both endothermic and exothermic reactions might be promoted.

Pure hydrocarbons are not made significantly radioactive by neutron capture and fewer restrictions occur in locating an irradiation chamber for such chemicals than would be the case with a food irradiation facility. Even if the chemicals irradiated are not pure hydrocarbons, they may contain other elements that become highly radioactive by neutron capture. Such radioactivity can be reduced to a tolerable level by storing the chemical products until the radioisotopes have decayed. This procedure is dependent on the elements involved and on the half-life of the radioisotopes formed. Another problem is the polymerization of hydrocarbon materials under the influence of radiation to form a cake or gum on surfaces.

SYSTEMS SUITABLE FOR CHEMICAL REACTION IN A REACTOR CORE

Unfortunately, because of the induced radioactivity, many chemical reactions are unsuitable to reactor cores. The elements to be considered for such chemical reactions are limited to either those with very low cross sections for thermal neutron capture, or those which, after neutron capture, produce radioisotopes that have quite short half-lives and would rapidly decay to stable isotopes. Carbon, hydrogen, oxygen, and nitrogen, and several less common elements, fall into this general class. The half-lives of the radioisotopes produced by neutron capture from oxygen, however, may require some hold-up of streams containing neutron-irradiated oxygen to allow time for the dissipation of radioactivity. Storage for a few minutes would permit safe handling, and with storage for a few hours the radioactivity originating from oxygen would be almost completely gone. The only radioisotope remaining after short storage of compounds produced from carbon, hydrogen, oxygen, or nitrogen would be a negligible quantity of C^{14} (half-life 5800 years). The minor residual radioactivity of irradiated carbon com-

pounds would probably be important only in foods. Other food elements, such as phosphorus, have much higher neutron capture cross sections, and would present much more of a problem than carbon.

So far as the chemicals that are not used in food are concerned, thermal neutron capture cross section is still the basic limitation in promoting chemical reactions in a reactor core. Compounds containing elements with high capture cross sections might, in addition to becoming radioactive, act as poisons in the reactor, decreasing its efficiency. If the kinetic energy of fission fragments is to be used, another limitation arises. The components of the system, particularly the chemical products, must be easily separated from the fission products. Liquefaction combined with distillation is the procedure suggested by Harteck and Dondes[3] for separating gaseous chemical products from gaseous fission products. Chemonuclear reactors were reviewed[72,97] in 1961 and the synthesis of ethylene glycol by fission fragments considered.[98]

OXIDATION OF NITROGEN IN A REACTOR CORE

Harteck and Dondes reported a number of studies indicating that it is feasible to oxidize nitrogen in a reactor core.[3] They first demonstrated that fission energy could be directly converted to chemical energy. This was accomplished by placing as little as 5 mg of uranium oxide powder containing about 90 per cent enriched U^{235} in a quartz vessel containing 20 cm^3 of mixed nitrogen and oxygen at a pressure of 1 atm. When this vessel and charge were placed in a reactor, the yield was increased 60 times over that observed in the experiment without the uranium oxide powder.

Studies on the oxidation of nitrogen in a reactor indicated that the conditions for the maximum yield of nitrogen dioxide would be (1) mixture composition, 80 per cent N_2, 20 per cent O_2; (2) pressure, 10 atm.; and (3) temperature, 200°C.

Under these conditions, a series of batch experiments showed that the nitrogen-oxygen mixture was essentially completely oxidized to a mixture of nitrogen dioxide and nitrous oxide.[3] The yields in G (the number of molecules produced per 100 ev consumed) were about 4 to 5 for nitrogen dioxide and 2 to 2.5 for nitrous dioxide. Greater refinement of the optimum conditions would probably increase these yields.

In another study made concerning direct use of fission-fragment energy, Diethorn et al. concluded that it is doubtful if a process would be economically feasible in which the fission energy in the core of a nuclear reactor is used solely for chemical synthesis.[4] For maximum efficiency in such a reactor, the fission fragments must be released from the fuel material, and must transfer their energy to the chemical reactant, and not to other fuel particles nor to a cooling stream of inert material. For a chemical synthesis in which the reactants are gases,[5] this requires not only fuel-particle sizes no larger than 5 μ, but also a minimum volume ratio of fuel to gaseous reactants. However, requirements for criticality demand a certain concentration of fuel in order that efficient use of fission-produced neutrons may be obtained. The compromise reached between these requirements will probably be one in which the fission-fragment energy absorbed by the reactants is significantly lower than desired.

The yield of chemical product per pass through the reactor core is expected to be low probably less than 0.05 per cent. For liquid-phase reactions the yield per pass will be less than 0.5 per cent and the G-value for these reactions are higher than those for gas-phase reactions by a factor of about 10.

One of the most difficult problems is that the chemical product obtained from direct-core processing may be contaminated with a very large amount of radioactivity. The largest portion of this radioactivity will be the result of the presence of fission product atoms intermingled with the chemical product. With many chemical systems a smaller but more troublesome fraction will be provided by the induced radioactivity in the chemical reactants and products themselves. Since this activity will come from nuclides which are isotopic with the chemical reactants and products, ordinary chemical and physical separation methods useful in the case of fission product removal cannot be used. Separation of these isotopic materials will be necessary to some degree since the activity contributed by them, although small, in most cases will be more than the permissible level for chemical materials. The techniques required for such purifications may well be as expensive as the synthesis of the material itself.

REACTOR CORE PROCESSING OF FOODS

TYPES OF APPLICATIONS

Many studies recently completed or in progress indicate that gamma irradiation can increase the utility or reduce costs in production of a great variety of commodities. As nuclear reactors increase in number, gamma irradiation will probably become an integral part of industry. On the basis of research, it would appear that this new process of gamma irradiation would assume greatest importance in the food industry. A short review of some of the research performed at the Fission Products Laboratory of the University of Michigan is presented to show the possible extent of such applications.

The process of subjecting food to doses of gamma radiation that do not completely sterilize, that is, do not destroy all the micro-organisms found in the food, is termed radiopasteurization.[6,7] A high radiopasteurization dose of about 1 Mrep* (one million roentgen equivalents physical) given to sea food, meats and vegetables extends their storage life at room temperature by 3 to 5 times the normal, and their unfrozen, refrigerated storage life from weeks to months.[8–10] Even medium radiopasteurization at about 100 krep reduces the population of micro-organisms in foods by over 90 per cent and extends refrigerator shelf life appreciably.[11,12] A process that is very promising is subradiopasteurization at 10 to 20 krep. A dose of this magnitude will not destroy enough of the micro-organisms in fresh foods to prolong storage life materially. Such a dose, however, will inhibit sprouting in potatoes and onions.[13–16] It will sterilize insects infesting grain and cereal products[17–19] and destroy the eggs and larvae of the Mexican fruit fly, stopping spread of infestation.[96] Such a dose can also be used to treat pork for the prevention of trichinosis. Similarly, control of tapeworm in beef, pork, and fish, and a number of other diseases caused by parasites[19–23] may be possible with gamma radiation. At present, several types of food, notably certain tropical fruits, are banned from import into the United States because of possible infestation. This ban could be lifted for foods given subradiopasteurizing doses. In addition, larger doses of about 4.8×10^6 rep are being studied by the U. S. Army Quartermaster Corps for radiosterilization.[1]

PROBLEMS OF USING THE REACTOR CORE

Since gamma radiation from the reactor core is accompanied by a high neutron flux, appreciable radioactivity would be induced by the capture of neutrons in food elements. This, of course, cannot be tolerated. Some consideration was given by the Internuclear Co., Inc.[1] to the possibility that a water shield might more rapidly attenuate neutrons than gamma photons.

Figure 4.1, based on an assumed maximum integrated tolerance[1] of 1.7×10^7 neutrons/cm^3 shows how with increased distance from the source, neutrons from the core are attenuated and the dose of gamma radiation that can be absorbed without exceeding a given neutron tolerance increases.

With a water-shield thickness of only 140 cm, a gamma radiation dose of only about 1 krep could be tolerated without exceeding the integrated mean neutron flux specified. If a water-shield thickness of 220 cm were used, the gamma radiation dose might be increased to about 17 krep. This is a maximum dose because of the production of neutrons by the high-energy (up to 8 Mev) gamma photons. This γ,n reaction for deuterium has a gamma threshold of 2.23 Mev. Since neutrons from this reaction are produced in the water, increasing the water-shield

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*For several years (1950-1956), it was common to report radiation doses in tissue and aqueous solutions in terms of rep (or megarep). This same unit was used for chemical systems in which the material had neither the unit density nor the same absorption characteristics as water or tissue. The rad (or megarad) is preferred today as a more accurate unit of dosage particularly when applied to materials differing appreciably from water either in density or absorptive properties. The conversion of units from earlier reports therefore, is not practical unless the absorptive properties of each material in question is known.

thickness does not materially further reduce neutron capture in the food. It appears, consequently, that reactor cores are unsuitable for the processing of foods, a conclusion reached by the Internuclear Co., Inc.[1] and also by Loftness.[24]

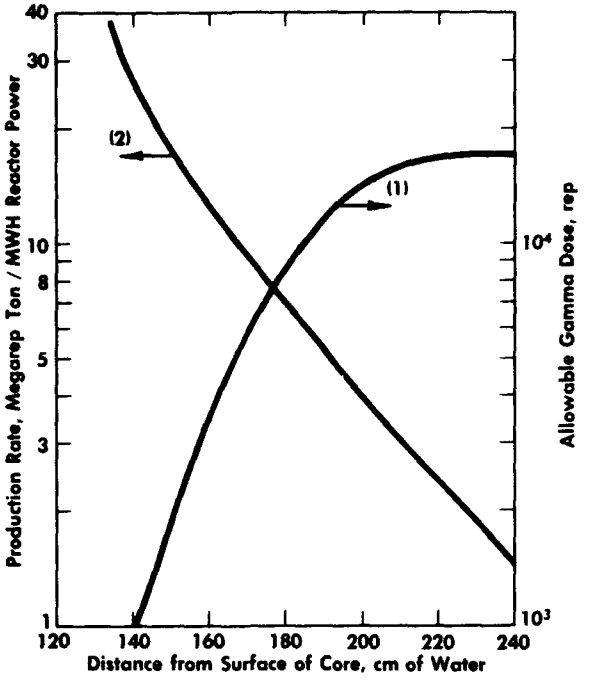


Fig. 4.1 — Allowable doses of gamma radiation for direct core gamma irradiation. (Courtesy of Internuclear Co.)[1]

RADIOACTIVITY OF GASEOUS FISSION PRODUCTS FROM FUEL SOLUTIONS

Two major groups of fission products come from U^{235}. One group with mass numbers between 80 and 120 contains a large percentage of krypton; the other group with mass numbers 120 to 160 has a large percentage of xenon. About 52 per cent of the total fission products have gas precursors. The total irradiation potential of these gaseous fission products is thus very high. Removing them will, in addition, appreciably ease the subsequent problem of fuel processing.

Table 4.1 — DISTRIBUTION OF THE FISSION PRODUCTS BY CHEMICAL FAMILY 10 SEC AFTER FISSION[25]

Fission products	Wt.% of total
Rb + Cs	20.0
Sr + Ba	2.6
Y + La + Ce	4.5
Sb	4.8
Mo + Te	5.2
Br + I + Ta	27.7
Xe + Kr	32.4
Total	97.2

Table 4.1 indicates the weight per cent distribution of about 97 per cent of the fission products (10 sec after fission), grouped by chemical families[25] and shows that xenon and krypton, both strong gamma emitters, comprise 32.4 per cent of the total listed. Of the 192 Mev per fission, about 10 Mev appear as energy of radioactivity in fission products. Thus, an ap-

preciable percentage of the energy of fission might be recovered as gamma radiation energy from the fission gases xenon and krypton. Manowitz[25] and Loftness[24] both estimate that about 1 per cent of the total fission energy can be so utilized if the fission gases are separated and used within about 1 min after fission.

THEORETICAL YIELD RELATIONSHIPS

A theoretical study of the gamma activity of fission product gases from a homogeneous reactor was reported by Donaldson.[26] Calculation of the build-up in curies of a fission product nuclide in a reactor requires knowledge of the power level of the reactor, the decay constant for the radioisotope, the fission yield for the radioisotope, the conversion factor of watts to fissions per second, and the number of disintegrations per second per curie (dis/sec-curie).

The rate of production, in atoms per second, of a radioisotope formed directly by fission is

$$\left(\frac{dN_1}{dt}\right)_f = y_1 Wf \tag{4.1}$$

where y_1 = fission yield of isotope 1 in atoms per atom of fissioned fuel
W = operating power level in watts
f = fission rate per watt
$= 3.1 \times 10^{10}$ assuming 192 Mev/fission

As the isotope is formed, however, it in turn decays at the rate

$$\left(\frac{dN_1}{dt}\right)_d = -\lambda_1 N_1 \tag{4.2}$$

where λ_1 is the decay constant for isotope 1.

The total time rate of change of the amount of isotope present during reactor operation then is

$$\frac{dN_1}{dt} = \left(\frac{dN_1}{dt}\right)_f + \left(\frac{dN_1}{dt}\right)_d = y_1 Wf - \lambda_1 N_1 \tag{4.3}$$

Integrating, the number of atoms present after reactor operating time, t, is

$$N = \frac{y_1 Wf}{\lambda_1}\left(1 - e^{-\lambda_1 t}\right) \tag{4.4}$$

The number of curies is related simply to the number of atoms present

$$A_1 = \frac{\lambda_1 N_1}{c} = \frac{y_1 Wf}{c}\left(1 - e^{-\lambda_1 t}\right) \tag{4.5}$$

where A_1 is the activity of isotope 1 in curies, and c is 3.7×10^{10} dis/sec-curie.

If isotope 1 decays with a decay constant of λ_1 to form radioisotope 2, which decays with a decay constant of λ_2, and so on, similar but more complicated integrations[26] yield the number of curies of the successive members of the decay chain present after t sec of reactor operation.

The results of these integrations are

$$A_2 = \lambda_2 \frac{Wf}{c}\left[\frac{y_1 + y_2}{\lambda_2}\left(1 - e^{-\lambda_2 t}\right) - \frac{y_1}{\lambda_2 - \lambda_1}\left(e^{-\lambda_1 t - \lambda_2 t}\right)\right] \tag{4.6}$$

$$A_3 = \lambda_3 \frac{Wf}{c}\left[K - Le^{-\lambda_1 t} + Me^{-\lambda_2 t} - (K - L + M)\, e^{-\lambda_3 t}\right] \tag{4.7}$$

where $K = \frac{y_1 + y_2 + y_3}{\lambda_3}$

$$L = \frac{y_1\lambda_2}{(\lambda_1 - \lambda_2)(\lambda_1 - \lambda_3)}$$

$$M = \frac{y_1\lambda_2}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_2)} - \frac{y_1 + y_2}{\lambda_3 - \lambda_2}$$

At the time of reactor shutdown, the activities (A_1, A_2, A_3, etc.) of the radioisotopes resulting from fission and decay during operation will be given by Eqs. 4.5 to 4.7. After reactor shutdown, production of fission products ceases and decay continues. The concentrations of the various radioisotopes during the decay period may be expressed by Eq. 2.1 (see Chap. 2).

$$A_1 = A_{0(1)}\, e^{-\lambda_1 t} \tag{4.8}$$

$$A_2 = \frac{\lambda_2}{\lambda_2 - \lambda_1} A_{0(1)}(e^{-\lambda t} - e^{-\lambda_2 t}) + A_{0(2)}\, e^{-\lambda_2 t} \tag{4.9}$$

$$A_3 = ke^{-\lambda_1 t}\, le^{-\lambda_2 t} + (A_{0(3)} - k - 1)\, e^{-\lambda_3 t} \tag{4.10}$$

where A . A_2, A_3 = activities of radioisotopes 1, 2, and 3 resulting from successive decay after reactor shutdown for time,

t = time after shutdown, sec

$$k = \frac{\lambda_2\lambda_3 A_{0(1)}}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)}$$

$$l = \frac{\lambda_3 A_{0(2)}}{\lambda_3 - \lambda_2} - \frac{\lambda_2\lambda_3 A_{0(1)}}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_2)}$$

DECAY SCHEME

Isotopes of bromine, krypton, iodine, and xenon may all exist as fission gases in an aqueous homogeneous reactor. Other isotopes that decay and produce these gases must also be considered. Isotopes with very short half-lives (about 1 sec or less) may be neglected since they will decay before they can be removed from the reactor. Also, isotopes with very long half-lives (of several years or more) may also be neglected because a typical reactor cycle would prevent sizeable accumulation of isotopes with long half-lives. With these considerations, Donaldson[26] assumed the decay scheme shown in Table 4.2.

CALCULATED FISSION GAS BUILD-UP

Using the previous relationships and the decay scheme listed in Table 4.2 and assuming that:

1. The gases do not escape the reactor until some time that is considerably greater than 1 sec,
2. Accumulated gas is flushed periodically; and
3. Conversion by neutron capture is not significant; Donaldson[26] calculated the build-up of gamma activity in gaseous products shown in Fig. 4.2.

FISSION GASES CONTINUOUSLY PRODUCED IN A MULTIPURPOSE REACTOR

The properties of the gaseous fission products of a 75-Mw multipurpose reactor[27] are summarized in Table 4.3. The yields reported are based upon 83.3 g of U^{235} fissioned; 10 min were allowed for removal of the fission products from the reactor core to the irradiation facility, where they would be held for 24 hr. It was assumed that the gaseous isotopes were removed from the fuel material as soon as they formed, decay taking place during the first 10 min in the pipes that conducted the core gases to the irradiation facility. Solid materials,

Table 4.2— DECAY SCHEME FOR GASEOUS FISSION PRODUCTS AND ISOTOPES PRODUCING GASEOUS PRODUCTS[26]

Br^{82}	→ stable			
Se^{83m}	90% → Br^{83}	→ Kr^{83m}	→ stable	
Se^{83m}	10% → Se^{83}	→ Br^{83}	→ Kr^{83m}	→ stable
Se^{84}	→ Br^{84}	→ stable		
Br^{85}	→ Kr^{85m}	→ stable		
Br^{87}	→ Kr^{87}			
Br^{88}	→ Kr^{88}			
Kr^{89}	→ *			
Kr^{90}	→ *			
Kr^{91}	→ *			
Kr^{92}	→ *			
Kr^{93}	→ *			
Kr^{94}	→ *			
Te^{131m}	→ Te^{131}	→ I^{131}	1% → Xe^{131m_2}	→ stable
Te^{131m}	→ Te^{131}	→ I^{131}	99% → stable	
Sb^{132}	→ Te^{132}	→ I^{132}	stable	
Sb^{133}	→ Te^{133}	→ I^{133}	Xe^{133}	→ *
Sb^{134}	→ Te^{134}	→ I^{134}	stable	
Te^{135}	→ I^{135}	30% → Xe^{135m}	Xe^{135}	→ *
Te^{135}	→ I^{135}	70% → Xe^{135}		
I^{135}	→ stable			
I^{137}	94% → Xe^{137}	→ *		
I^{137}	6% → stable	→ Xe^{136}		
I^{138}	→ Xe^{138}			
I^{139}	→ Xe^{139}			
Xe^{140}	→ *			
Xe^{141}	→ *			

* Decays to stable solid through a long chain.

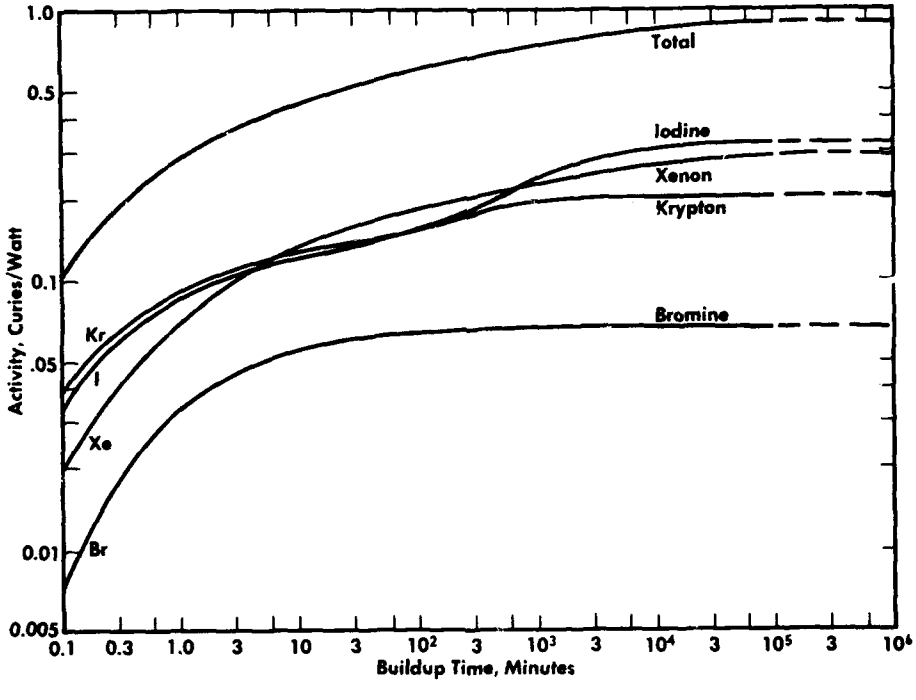


Fig. 4.2—Buildup of gamma activity in gaseous products per watt of reactor power (log-log scale).[26]

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formed as decay products, are deposited in both the pipeline and the irradiation facility. From left to right the columns in Table 4.3 list the elements and active isotopes, the half-lives of these elements, the beta and gamma emission energies of the isotopes in millions of electron volts, the fission yield in atom per cent, the mass of gas exposed to target in grams, and the beta and gamma power output which is the product of mega electron volts (Mev) and megacuries (Mc).

Table 4.3 — PROPERTIES OF FISSION GASES CONTINUOUSLY PRODUCED IN A MULTIPURPOSE HOMOGENEOUS REACTOR[27]

Nuclide	Half-life	Beta energy, Mev	Gamma energy, Mev	Fission yield, at.%	Mass of gas, M, exposed to target	Beta power output, P_β, Mev-Mc*	Gamma power output, P_γ, Mev-Mc	Gamma activity, Mc
Kr^{83}	108 m		0.033	0.59	0.0175		0.0125	0.406
Kr^{85}	4.36 h	0.82	0.149	1.0	0.077	0.179	0.129	0.288
			0.3					
Kr^{87}	78 m	3.2		2.0	0.043	1.30	0.169	0.12
Kr^{88}	2.77 h	2.4		3.0	0.158	4.93	0.895	0.20
Rb^{88}	18 m	5.0						
Kr^{89}	2.6 m			4.6	0.00025	0.303		
Rb^{89}	15 m	4.5						
Inactive Isotopes					1.01			
Xe^{133}	5.27 d	0.35	0.081	6.3	2.72	0.059	0.158	0.505
			0.232					
Xe^{135m}	15.3 m		0.52	1.8	0.54		0.392	0.751
Xe^{135}	9.2 h	0.93	0.25	5.9	1.29	1.02	0.808	3.24
Xe^{137}	3.9 m	4.0		6.0	0.0018	0.91		
Xe^{138}	17 m			6.2	0.0342	2.39	3.32	2.77
Cs^{138}	33 m	2.6	1.2					
Xe^{139}	41 s							
Cs^{139}	9.5 m	2.3	0.165	6.3	8.3×10^{-7}	1.67×10^{-3}	3.3×10^{-4}	
Ba^{139}								
Inactive Isotopes					9.8			
Total Power Output						11.31	5.88	8.28

* Mev-Mc is the product of energy, expressed in Mev, and activity expressed in Mc.

Fission product gas radiation sources must be contained in a leakproof system with walls thin enough to allow most of the radiation to pass. Beta particles are readily absorbed and would be attenuated in the container wall, so only the gamma energy output would be available for irradiation purposes. Thus, only about one-third the total emitted energy could be used as gamma radiation in most irradiation facilities. This corresponds to 0.72 per cent of the total energy from the reactor.

The basis of calculation used to determine various quantities in Table 4.3 are given below.

(1) Reactor power	75 Mw
(2) Fuel consumption, m	83.3 g of U^{235} per day
(3) Time taken to remove gas from reactor core to irradiation cell, T_1	10 min
(4) Time of hold-up in irradiation cell, T_2	24 hr
(5) Time of decay in fuel, T_0	0
(6) Total mass of gas at target (volume at STP, 3.21 liters)	15.7 g

(7) Total mass of gas in pipe line (volume at STP, approximately 30 ml)	0.167 g
(8) Mass of material deposited at target	7.16 g/day
(9) Mass of material deposited in pipe-lines	5.8 mg/day

FISSION GAS RADIATION SOURCE

With the anticipated future demands of the process industry for megacurie gamma ranges, the problem of providing satisfactory sources of radiation becomes important. The use of fuel elements, considered in earlier studies (Refs. 4, 5, 9-11, 28-30), has many limitations and could fulfill the needs of only a limited number of irradiation facilities.

Fission gases from a homogeneous reactor can serve as an efficient source of radiation. If the reactor and irradiation facility are coupled together and fission gases are transferred directly from the reactor to the irradiation facility, there is little decay of gamma activity. The gases can be pumped in and out of the radiation source without using the complicated devices required in handling radioactive solids. Fission gases can be stored in tubes of any reasonable shape and size. Any cooling required can be by a double-pipe heat exchanger connected to the gas line. At low pressures, the gases have low densities. They therefore absorb little radiation and increase in temperature only slightly. This considerably reduces the problems of cooling and thermal failure. On the other hand, if fuel elements are used as the radiation source, cooling to prevent failure as a result of thermal stress is a serious problem. The container material selected for the fission gases should be one that absorbs only negligible radiation and will not overheat. The annulus between two concentric container tubes could be evacuated to enable immediate detection of any leaks in the system. Escape of fission gases is the chief objection to the use of such facilities, since even small quantities, free in the air, are extremely dangerous. Sensitive pressure detectors could be used in the annulus to determine any pressure changes due to leakage of gases. The second pipe could serve as an emergency container, enabling the operator to remove fission gases from the source when a leak was detected. Another safety precaution which reduces the possibility of contaminating the personnel area would be to ventilate the chamber continuously and monitor the exhaust duct. A monitor installed on this line and shielded from the high-energy source should detect any radioactive gases. Such precautions reduce the probability of seriously contaminating the surroundings.

Table 4.4 lists the maximum permissible breathing concentrations for the gaseous fission products.[26]

Table 4.4 — MAXIMUM PERMISSIBLE CONCENTRATIONS FOR GASEOUS FISSION PRODUCTS[26]

Radioisotope	Maximum permissible concentration, $\mu c/cm^3$
Br^{82}	7×10^{-7}
Br^{83}	5×10^{-6}
Br^{84}	2×10^{-6}
Kr^{85}	2×10^{-6}
Kr^{85m}	2×10^{-6}
Kr^{88}	4×10^{-6}
I^{131}	3×10^{-9}
I^{132}	8×10^{-8}
I^{133}	9×10^{-9}
I^{134}	2×10^{-6}
I^{135}	3×10^{-8}
I^{136}	2×10^{-5}
Xe^{133}	4×10^{-6}
Xe^{135}	2×10^{-6}

The use of gases permits some flexibility in source configurations. The source used in the design shown in Fig. 4.3 consists of six passes of two concentric, thin-walled, stainless steel tubes.[31] The tube spacings (Fig. 4.3) were calculated to give a uniform field at approximately 3 ft from the source plane.

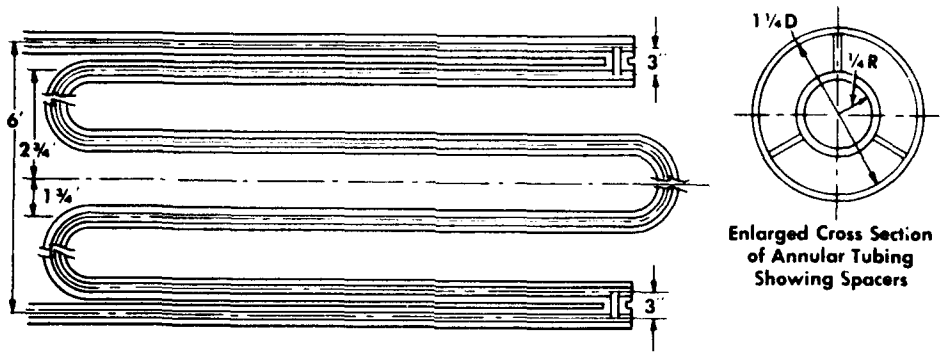


Fig. 4.3 — Diagram of fission-gas source.[31]

In the proposed design a pump is included to provide continuous circulation of the gases within the source, maintaining uniform activity throughout. The gases pumped would consist of fission gases dispersed in an inert carrier gas to facilitate pumping, and would pass from the reactor core through a cold trap to remove any vapors or solid particles that may have been carried from the system. The pressure of the system can be varied; increasing pressure increases source strength in the facility. The equilibrium volume of fission gases is about 25 per cent of the system volume. A water line is provided to flush out the source and remove any solid daughter products accumulating in the system. The flowsheet of the removal of fission gases is given in Fig. 4.4.

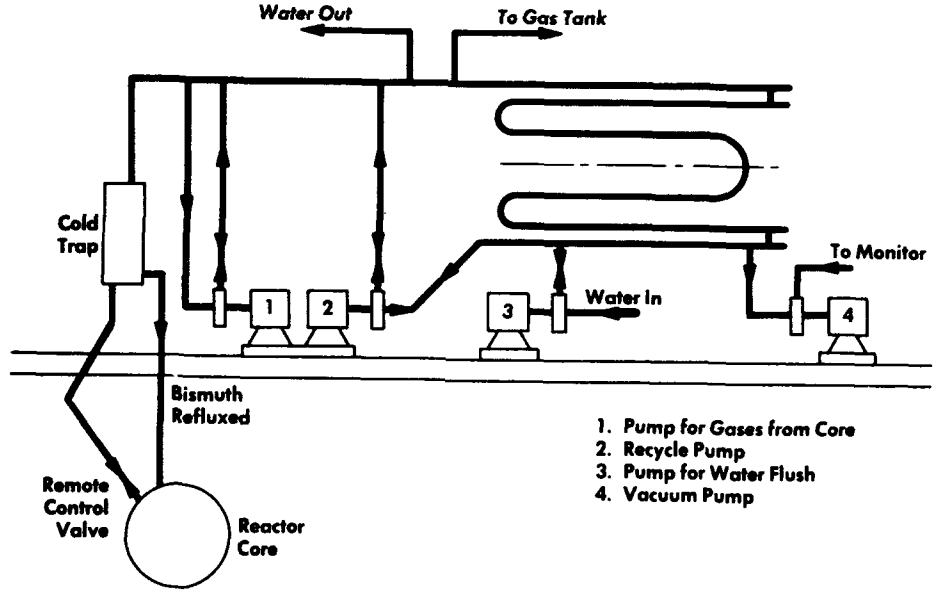


Fig. 4.4 — Flowsheet of fission-gas source.[31]

In calculating source strength it was assumed possible to concentrate in the source all the xenon and krypton theoretically present at a given time. The pump should be capable of pumping essentially all the fission gases to the source once they are released from the fuel solution. The diffusion time required for gaseous fission products to reach the surface following their production appears to be the controlling factor. It would be expected that homogeneous

aqueous-solution reactors would readily release the gases, whereas a liquid-metal homogeneous reactor (LMHR) might require a slightly greater period of time because of the difference in fuel viscosity. Experimental work performed at The University of Michigan with the bismuth-uranium fuel system indicates that it will "off gas" readily, thus minimizing the loss of gaseous nuclei by neutron capture.

The rapid removal of fission gases is desirable on the basis of neutron economy, because Xe^{135} has an exceptionally large neutron-capture cross section ($\sigma_a = 3.5 \times 10^6$ barns). The continual removal of Xe^{135} from the reactor will save the fraction of reactivity otherwise used in overcoming the xenon poison and removal of the Xe^{135} simplifies reactor control.

FISSION PRODUCT GAS USED OUTSIDE THE REACTOR CORE

Use of pure gamma radiation eliminates the problem of neutron-induced radioactivity. It offers unique advantages as an initiating agent for the synthesis and polymerization of plastics,[32] some of which follow:

1. With head-sensitive monomers, the reaction can start at lower temperatures.
2. Polymerizations can be completed in the solid state.
3. Resulting polymers will be free of contamination from any catalyst particles.

Alterations in physical properties of the polymers caused by irradiation may be of commercial importance. The General Electric Company is now marketing a polyethylene irradiated by high-speed electrons which has an appreciably higher melting point than nonirradiated polyethylene.[33] Rubber can be vulcanized by gamma irradiation without using heat or sulfur compounds.[34,35] By present estimates, however, the cost of rubber vulcanized in this manner would be high because of the large dose of radiation required. Halogenation reactions give appreciable yield of products using relatively low doses of radiation.[36,37]

Studies were reported on the radiation polymerization of monomers such as acrylonitrile[38,39] before 1950. Radiation polymerization rates for acrylonitrile[40] and for styrene, vinyl acetate and butadiene were reported more recently.[41–44] In 1953 gamma radiation promotion of copolymerization of styrene and methyl methacrylate[45] and additional data on the radiation polymerization of acrylonitrile[46,47] were reported. Studies have also been made on polymerization of a number of fluorinated monomers[32] and on polymerization of n-vinylpyrollidone in aqueous solutions (for use as a blood plasma substitute).[32] Because polyethylene has great commercial importance, gamma radiation-induced polymerization of ethylene has been of considerable interest.[48–53]

The list of reactions promoted by gamma radiation could be extended considerably; however, promotion of chemical reactions by radiation is discussed in fuller detail in Chap. 7. Additional information on this subject, particularly in the field of polymers, is given in Refs. 54 to 78 of this chapter.

FISSION GAS IRRADIATOR USING A MOBILE REACTOR

Manowitz discussed the use of ionizing radiation from fission gases of a mobile reactor for chemical processes.[25] He suggested an aqueous homogeneous reactor similar in some respects to the one described by Loftness about 1 year later which is described later in this chapter. A proposed flowsheet is shown in Fig. 4.5.

The optimum core radius was calculated to be about 7 in., with a $\frac{1}{8}$-in.-thick stainless steel shell between the core and the reflector. A 100 per cent enriched U^{235} solution is used in the core; this radius would require a critical mass of about 1200 g. The total fuel inventory was estimated at 30 liters, at 80 g of 100 per cent enriched U^{235} per liter, giving a total of 2400 g. Manowitz also proposed the use of a $2\frac{1}{2}$-ft-thick air-cooled graphite reflector rather than the aqueous reflector suggested by Loftness. The estimated residence time in the core was 1.5 sec with a circulation rate of 160 gal/min. The average neutron (n) flux in the core was estimated to be 2×10^{13} neutrons/cm^2/sec.

As shown in Fig. 4.5, some of the fuel is flashed into vapor in a spray chamber above the core. The vapor carries the minute quantities of highly radioactive fission gases and also the

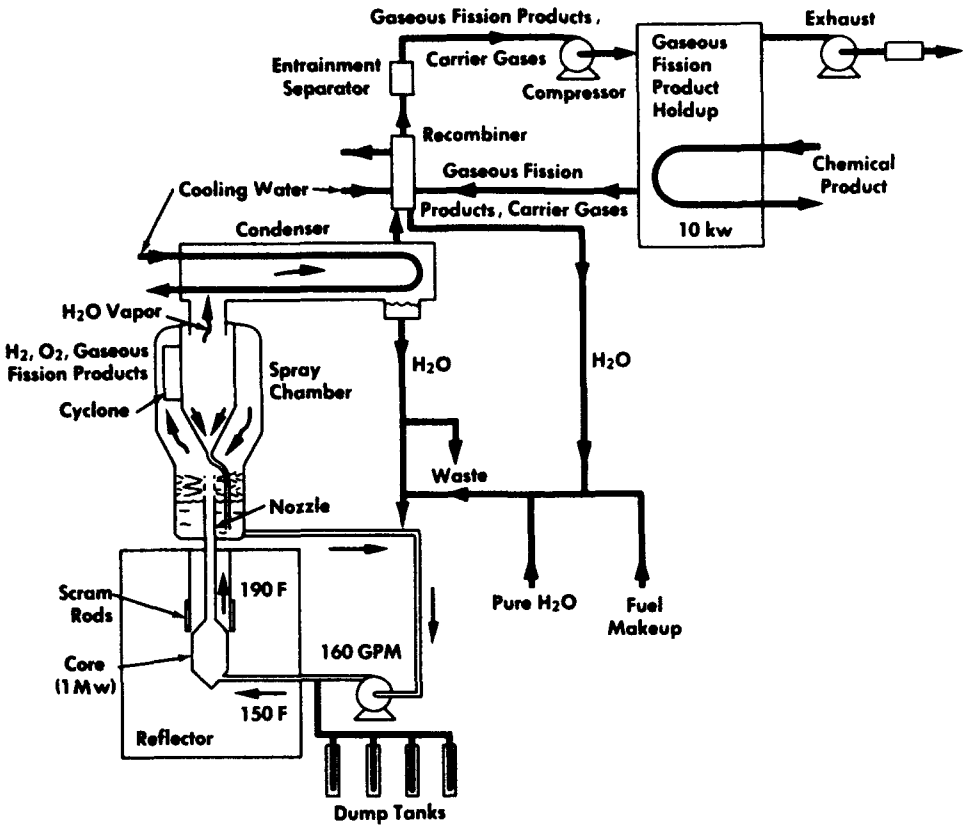


Fig. 4.5 — Gaseous fission-product irradiation facility proposed by Manowitz.[25]

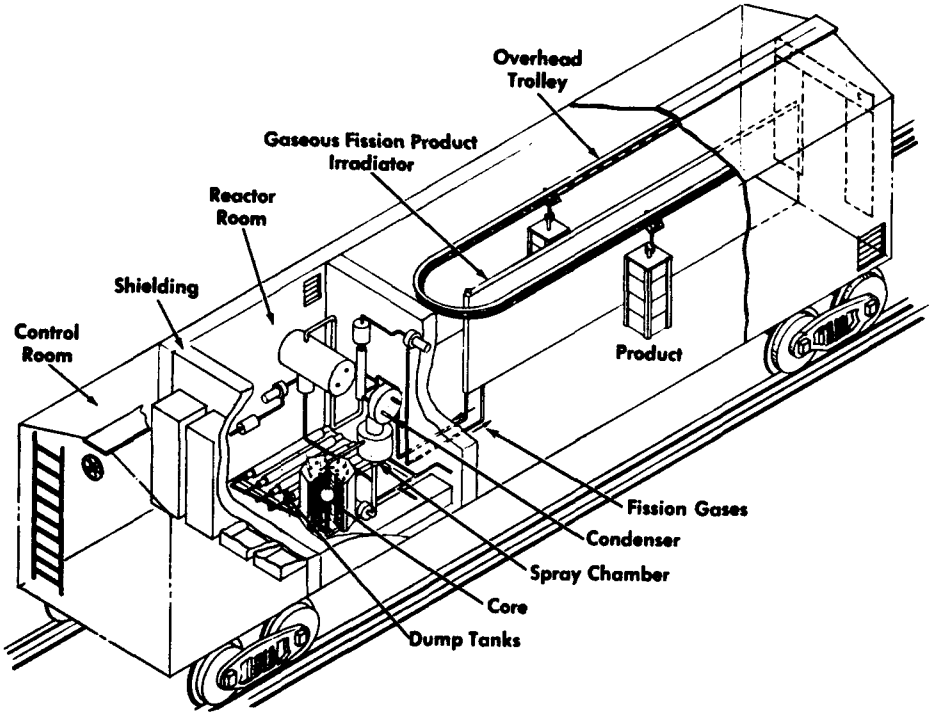


Fig. 4.6 — Proposed reactor boxcar.[25]

flashing of vapor serves to cool the fuel solution. The vapor containing the fission gases then passes through a cyclone separator and a condenser which returns the condensate and any entrained droplets of fuel to the liquid fuel system. Fission gases are picked up by a gas such as hydrogen, or by one of the inert gases, and circulated through a recombiner, entrainment separator and fission gas radiator. The recombiner allows the hydrogen and oxygen produced by water radiolysis to react. Water thus produced is removed in the entrainment separator.

Manowitz suggested that the entire facility could be placed in a railway car, (Fig. 4.6), with the obvious advantage of being able to move the gamma source near origin points of products such as seasonal crops or perishables.

The car would have enough shielding to protect railway personnel from the fission products accumulated in the fuel. Gaseous fission products would decay so rapidly that a brief shut-down before moving the car would lower the radiation field around the fission gas radiator to a tolerable level. Additional shielding would be required when the reactor is operating.

Figure 4.6 shows a monorail conveyer system which might be adapted to the irradiation of packaged food products; the conveyor would probably be replaced with piping for irradiating fluid chemicals.

FISSION GASES AND REFLECTOR SOLUTIONS AS FOOD IRRADIATION SOURCES

Most reactors employ a reflector around the reactor core to provide better neutron economy. A reflector consists of a suitable thickness of moderator material. Neutrons escaping from the core and entering the reflector may interact elastically with the atoms of the reflector medium and return to the core, with a conservation of neutrons. Some of the neutrons entering the reflector, however, will be captured and can produce radioisotopes there. If the reflector is of water, various salts may be dissolved in it and radioisotopes produced. Loftness[24] suggested an aqueous solution of $MnSO_4$ for the reflector. On neutron capture, radioactive Mn^{56}, a gamma emitter with a half-life of 2.6 hr, would be produced. Use of such a reflector solution as an additional gamma source would increase the radiation capacity by about 10 per cent above that possible by the use of the fission gases alone. No fissioning would occur in the reflector solution; it could also be circulated through a gamma radiator without danger of inducing radioactivity in the food being irradiated.

Loftness[24] considered the design of an irradiation facility for food using gamma radiation both from separated fission gases and from a circulating reflector solution. The design data for such a facility are given in Table 4.5 and the flowsheet in Fig. 4.7.

Table 4.5 — DESIGN DATA FOR GAMMA-IRRADIATION FACILITY USING FISSION GASES AND REFLECTOR SOLUTION AS RADIATION SOURCES[24]

Nominal power level	1 Mw
Fuel solution	U^{235} enriched UO_2SO_4, in H_2O
Core, ID	18 in.
Core material	Stainless steel
Core wall thickness	$\frac{1}{8}$ in. (minimum)
Core volume	50 l
Specific power	20 kw/l
Reflector	$MnSO_4$, in H_2O
Reflector thickness	10 in.
Reflector tank	Stainless steel
Core pressure	60 psi
Core temperature	133°C (275°F)
Fuel circulation	100 gpm

The liquid fuel is pumped (Fig. 4.7) through the core at a rate of 100 gpm at a pressure of 60 psi. The exit stream leaves at 275°F, is pumped through a heat exchanger where it is cooled to 200°F. The fuel then passes to a centrifugal separator where fission gases, gases formed by radiolysis of the water, and recycle gases are quickly separated from the liquid. The cooled, degassed liquid fuel then returns to the reactor core to repeat the cycle.

The gases stripped from the fuel are taken through a condenser and a recombiner where the hydrogen and oxygen from radiolysis are reacted. The gases then go to the irradiation chamber where they pass through a flat slab serving as a plaque source. The gases are removed by a jet pump operated by the circulating liquid fuel. The gases from the jet pump mix with the circulating liquid fuel as it enters the heat exchanger. This recombination of the streams serves two purposes: the recombined water and decay products are returned to the fuel system; and the circulated gases aid in stripping the minute amounts of very active fresh fission gases from the liquid fuel.

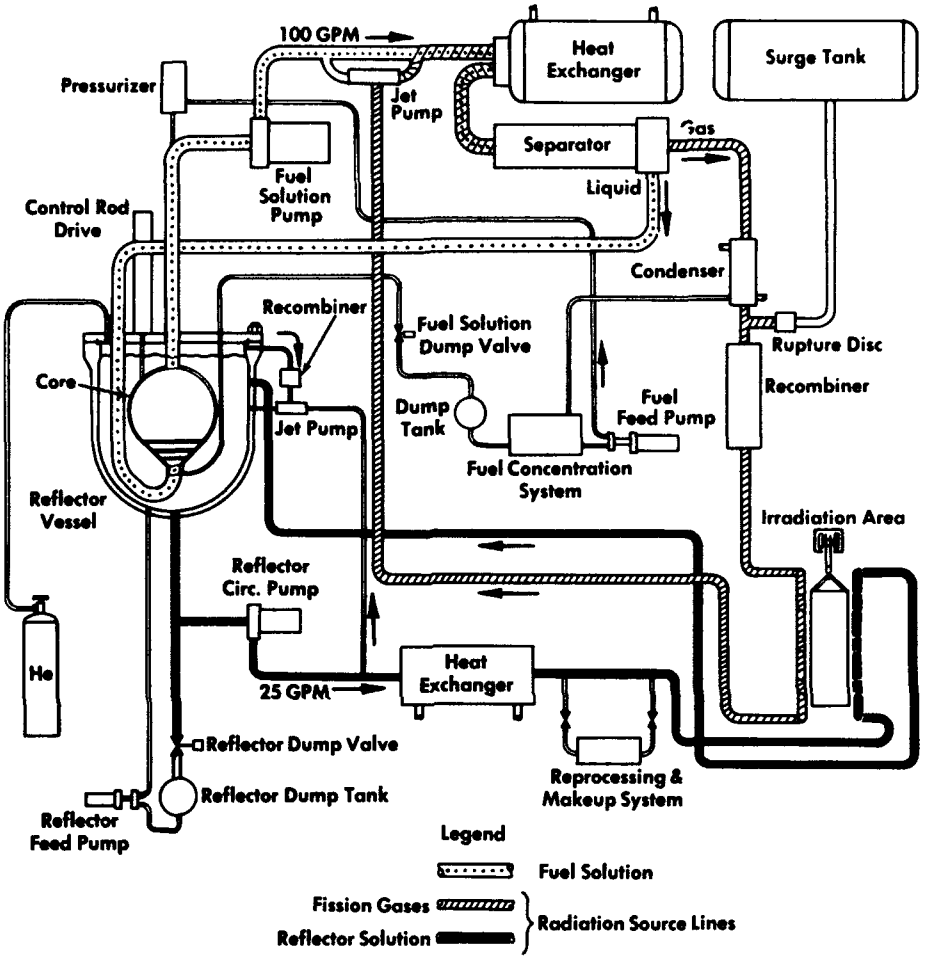


Fig. 4.7 — Flowsheet for gamma-irradiation facility using fission gases and reflector solution as radiation sources.[24]

A circulating reflector solution containing dissolved manganese sulfate and neutron-activated manganese sulfate is circulated through another radiator parallel to the fission gas radiator. The radiation chamber, 9 by 16 ft, is surrounded with concrete shielding about 7 ft thick. Loftness proposes a monorail system for conveying food in and out of the radiation chamber.[24]

In a review[1] of the types of gamma sources that might be produced by a nuclear reactor, the use of fission gases was considered very promising. Further development, however, was believed necessary. The main advantage of using fission gases as the radiation source may be realized if the reactor is also employed for other purposes, such as power production, as well as for food irradiation.

If fission gases are desired as radiation sources, a homogeneous-fuel reactor is required. Reactors with homogeneous fuel have many advantages: small core size, simple core design, and no costly fuel-element fabrication. The problem of corrosion, however, exists in both the

aqueous fuel and liquid-metal fuel homogeneous reactors. Although fission gases hold great promise as gamma sources, use of this design depends on satisfactory solution of the corrosion problems associated with homogeneous reactors.

RADIOACTIVITY FROM MIXED FISSION PRODUCTS IN FUEL SOLUTION

Utilizing fission products from homogeneous reactors by bleeding a stream of the highly radioactive fuel solution presents more problems than use of fission product gases. For instance, unless the bleed stream is from the same stream being removed for processing, the inventory of fissionable material increases. Such a system is considered feasible, however, because frequently the feed material must be "cooled" for some time before processing. Another problem is that the fuel solution emits delayed neutrons. Table 4.6 gives properties, yields, and energies of these delayed neutrons.[79]

Table 4.6 — PROPERTIES OF DELAYED NEUTRONS IN SLOW-FISSION OF U^{235} (79)

Half-life T_i, sec	Mean-life T_i, sec	Decay constant λ_i, sec^{-1}	Fraction, B_i	Energy, Mev
0.43	0.62	1.61	0.00084	0.42
1.52	2.19	0.456	0.0024	0.62
4.51	6.50	1.151	0.0021	0.43
22.0	31.7	0.0315	0.0017	0.56
55.6	80.2	0.0124	0.00026	0.25

Some materials, certain foods among them, are sensitive to neutron capture. Obviously they cannot be irradiated by fuel solutions that have not had time to cool enough. Such solutions should be cooled about 10 half-lives or about 600 sec before use, to irradiate these materials. Cooling time would not necessarily increase inventory costs significantly. If heavy water were used as the moderator solvent in the homogeneous reactor, some neutrons would be generated by γ,n reactions in deuterium nuclei. This fact must be considered if the material being irradiated is very sensitive to neutron capture. Molten-metal homogeneous fuels have the advantage as opposed to aqueous fuel that delayed neutrons will not be moderated and the capture cross sections for these fast neutrons will be small in consequence. The high temperatures required, however, may be a disadvantage. Many materials do not capture neutrons. Petroleum hydrocarbons, many plastics, and rubber could safely be irradiated by homogeneous fuels sources, and other materials might well be added to this group.

Utilizing the core solution as an irradiation source would increase the inventory cost slightly, and this would have to be met in charges for the irradiation service. The amount of extra U^{235} cannot be calculated until a design is finally adopted. It is evident from the work of Robb et al.[80] that fission product distribution does not change significantly with irradiation time; activity yields for the fuel solution would therefore not be sensitive to the degree of burn-up or to irradiation time, where burn-up is between 0.1 to 1.0 per cent of the U^{235} present.

In many chemical processes, desired reactions might be achieved by simultaneous application of heat and irradiation. This is particularly true if irradiation helps crack hydrocarbons at moderate to high temperatures. Feed materials could be both heated and irradiated with a small stream from the core fuel solution reducing the circuitous route of reactor steam feed for heat transfer and the attendant additional equipment. Hydrocarbons would not be activated by delayed neutrons generated in the liquid metal fuel solution. Fuel from the aqueous homogeneous reactor could be used for the same purpose, but temperatures reached would be considerably lower.

REACTOR LIQUID FUELS AS FOOD IRRADIATION SOURCES

Liquid fuels for reactors might provide the gamma sources for some of the food-irradiation processes described. The fission energy distribution given in Table 3.1 (p.55) shows that of

192 Mev per fission event, 6 Mev is associated with the prompt gamma photons and 5 Mev with the gamma and beta-particle radiation from decay of fission products. Thus, $^{11}/_{192}$ or about 5.1 per cent of fission energy is theoretically available for gamma irradiation. As discussed previously, however, foods cannot be irradiated in the reactor core, because of neutron capture and resulting induced radioactivity. Also the design of a reactor would be greatly complicated and the critical size and fuel inventory greatly increased, if provision were made to convey bulky items such as foods through a reactor core. Homogeneous reactor liquid fuels might be removed from the core, and circulated through an external loop so that the fission products in the fuel solution could serve as a gamma source. The fission product gases, xenon and krypton, account for about 35 per cent of the gamma activity. Many radioisotopes of these gases have very short half-lives, and the fuel solution must be used as quickly as possible after removal from the core to get the most out of the gamma activity.

Although the fuel removed would be circulated through pipe lines and equipment small enough in size and geometry to prevent the formation of a critical mass outside the reactor core, some fission would occur in the external loop as a result of delayed neutrons. When fuel in a fissioning reactor captures neutrons, fission occurs promptly (perhaps as quickly as 10^{-14} sec), with, in most cases, the emission of new neutrons termed prompt neutrons. In some of the cases, however, fission may not occur until later, sometimes several minutes after neutron capture, and here the new neutrons emitted are called delayed neutrons. Actually, about 99 per cent of the total emitted may be considered prompt neutrons. If delayed neutrons are captured by a target outside of the core, radioactivity can be induced.

INDUCED RADIOACTIVITY

Loftness[24] has calculated for phosphorus alone the radioactivity that would be induced by the delayed neutrons in a fuel circulated outside a reactor core. With no shield between the meat and the gamma radiator a gamma surface dose rate of 2740 rep/min gave an induced P^{32} activity of 13 dis/cm^3/sec, which is about 3000 times the assumed tolerance. Using 10 cm of water as a shield between the gamma radiator and the meat, the gamma dose rate was calculated as 1140 rep/min, but the induced phosphorus activity was reduced only to 4.4 dis/cm^3/sec, still more than 1000 times tolerance. Loftness concluded that to irradiate food, a circulating homogeneous fuel as a gamma source was not feasible, because of induced radioactivity.[24]

USE OF A SHIELDED NUCLEAR FUEL

In a separate study[1] undertaken by the Internuclear Co., Inc., for the Quartermaster Corps (QMC), U. S. Army, fissioning nuclear fuel was considered as the radiation source with a water-shield 1 ft thick between the radiator and food (gamma irradiated 2×10^6 rep) to reduce the neutron dose level to 1.7×10^7 neutrons/cm^2/sec estimated tolerance dose. As an alternative, a holding tank for the liquid fuel between the radiator and the core was proposed. The longest half-life for the delayed neutron emitters in the fuel would be about 55 sec. A hold-up time of about 5 min would be required for decay of these neutron-emitting radioisotopes. It was estimated[1] that the hold-up would reduce the gamma energy output by about 30 per cent. In addition, use of the holding tank would require a large volume of liquid fuel, so that the U^{235} inventory, and hence the cost, would have to be increased. Figure 4.8 shows the schematic use of homogeneous circulating fuel.

Criticality outside the core, leakage of the radioactive fuel, mechanical equipment, and adequate shielding of the entire circulating system are some of the problems encountered if nuclear fuel is used as a source of gamma radiation. Because of induced radioactivity in foods, circulating fuel as a radiation source would not be practicable from either the economic or the engineering standpoint. Such gamma sources would be suitable, however, in promoting some of the chemical reactions discussed previously.

REACTOR FUEL ELEMENTS AS SOURCES OF RADIATION

In the operation of a nuclear reactor, fission products and transuranic elements are formed from the uranium contained in the fuel elements. When these materials accumulate to certain

concentrations, or radiation damages the fuel elements, the elements must be removed and chemically processed to recover uranium suitable for refabrication into new fuel elements. After fuel elements have been removed from the reactor, it is current practice to store them long enough for the intense radioactivity to decay. Except for limited use in experiments, the radioactivity from fuel elements is dissipated in the storage wells. This storage operation is

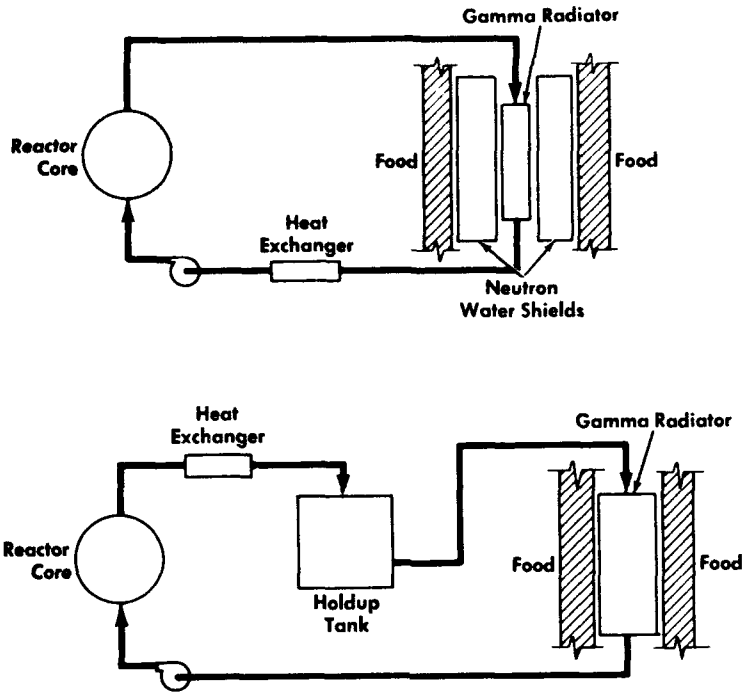


Fig. 4.8—Schematic use of homogeneous circulating fuel.[1]

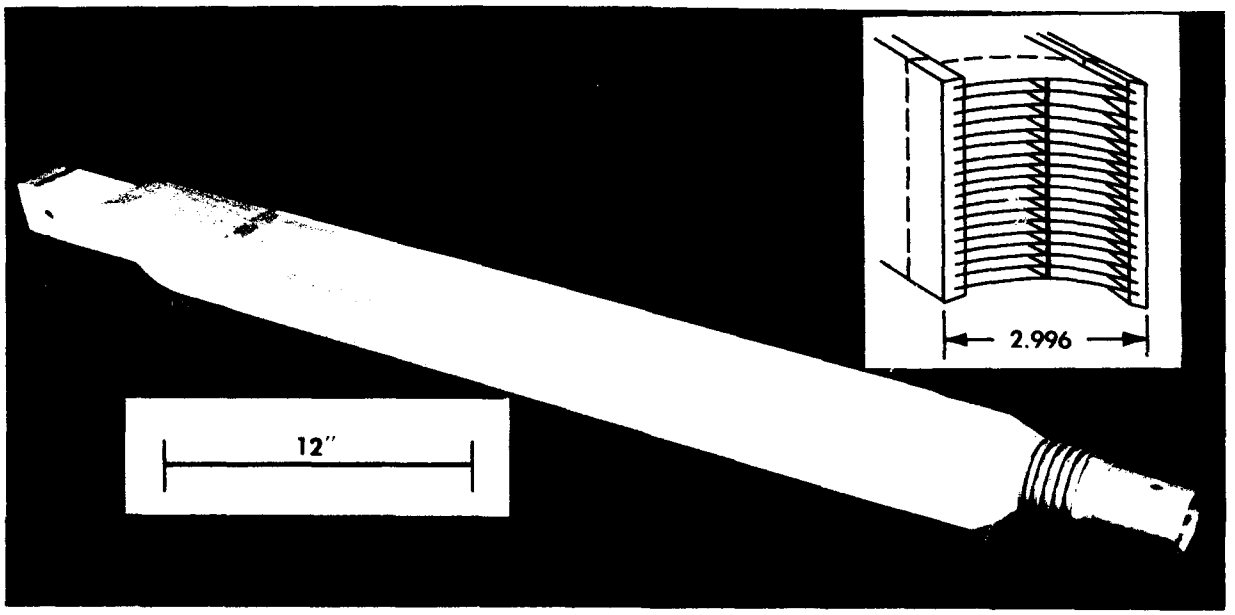


Fig. 4.9—MTR fuel element.

expensive because large inventory costs are associated with fissionable material. If some of these fuel elements were used as radiation sources, part of the expense of the delay period before chemical processing, would be defrayed. Thus, use of the fuel elements as commercial sources of radiation might be adapted conveniently and economically to the present scheme of chemical processing operations.

MTR FUEL ELEMENTS

Many reactors have been designed to incorporate widely different types of fuel elements. Eventually, to minimize cost, power reactors using fuel elements may employ a few standard designs. The most widely used fuel element now is the one designed originally for the Materials Testing Reactor, (MTR). With slight variations, the type of fuel element shown in Fig. 4.9 has been or will be used in such reactors as: Materials Testing Reactor, Low Intensity Test Reactor, Bulk Shielding Reactor, Argonne Research Reactor, Boiling Reactor Experiment No. 1, Boiling Reactor Experiment No. 2, Ground Test Reactor, Pennsylvania State Reactor, University of Michigan Reactor, Oak Ridge Research Reactor, Battelle Memorial Institute Reactor, Livermore Pool Type Reactor, Watertown Arsenal Reactor, and Naval Research Reactor.[81]

The extensively used MTR type fuel element has high neutron flux, resulting in highly radioactive spent fuel elements; accordingly this type of fuel element is the most logical for consideration as a radiation source. A detailed description including fabrication methods for the MTR fuel element was given by Cunningham and Boyle.[82]

In one possible cycle for the MTR, the reactor might be operated for 12 days at a power level of 30 Mw and then shut down for 2 days while all 23 fuel elements are discharged to a canal filled with water.[83] Figure 4.10 is a photograph showing a number of MTR fuel elements assembled in a rectangular grid in the canal.[84] Note the glow of Čerenkov radiation around the elements.

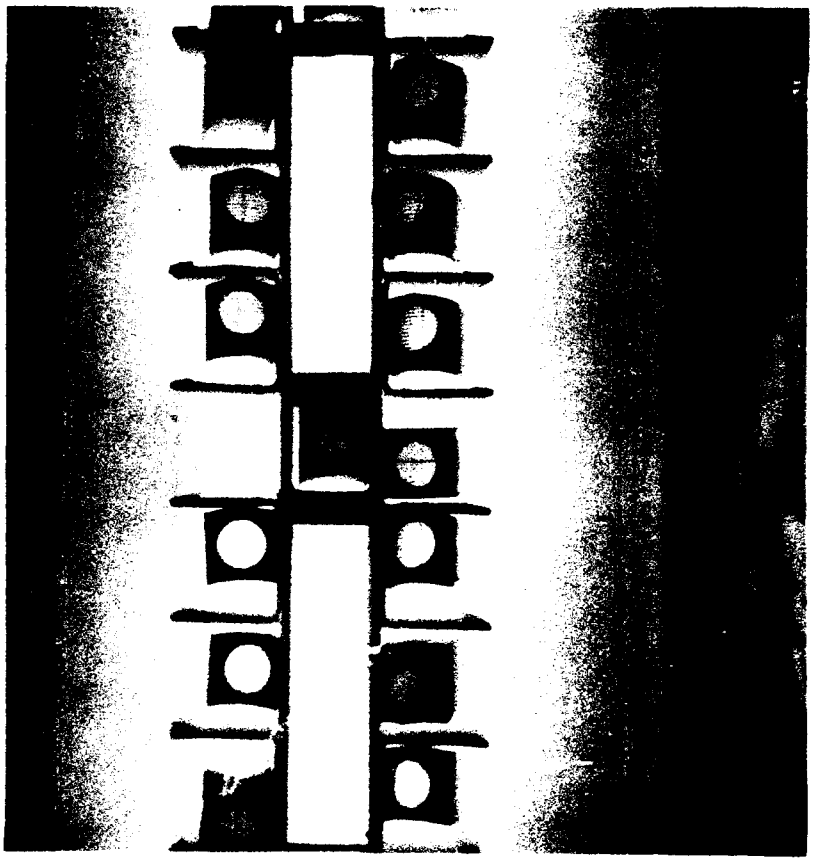


Fig. 4.10—MTR fuel elements in rectangular grid in water canal.[84]

Fuel elements are very radioactive when first removed from a reactor. They decay initially at a rapid rate, however, losing about half their gamma activity in less than 2 days. After 3 weeks, this activity has decreased by a factor of about 10. A decay curve based on a large number of measurements on a number of different fuel elements is shown in Fig. 4.11.[84] The gamma-radiation energy spectrum of MTR fuel elements cooled for 30 days is given in Table 4.7.

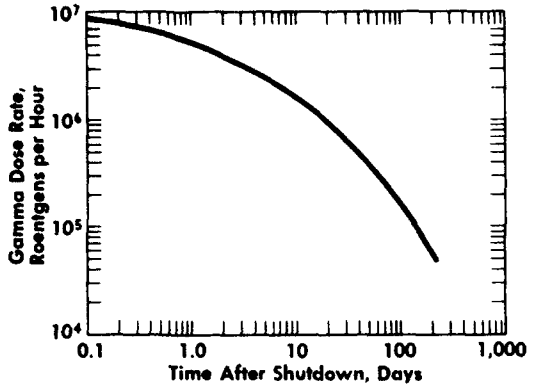


Fig. 4.11—Average gamma dose rate per MTR spent fuel element vs. time after shutdown (measured at 3 in. from source in water).[84]

Table 4.7—GAMMA-RADIATION ENERGY SPECTRUM FOR MTR FUEL ELEMENTS[84]

Group	Average energy $\bar{E}$, Mev	S*	Contribution to total activity, %
I	2.50	0.000260	0.8
II	1.75	0.005990	19.1
III	1.25	0.000026	0.1
IV	0.90	0.002720	8.7
V	0.70	0.007900	25.2
VI	0.50	0.005980	19.0
VII	0.35	0.008500	27.1

Irradiation time, 17 days; cooling time, 30 days

* S, source strength in photons per second based on one fission per second.

Groups of fuel elements in the water-filled canal at the MTR site in Idaho have been a major source of gamma radiation for studies by the U. S. Army Quartermaster Corps and its contractors. These sources of gamma radiation have been supplemented by additional irradiation facilities using fuel elements at Argonne National Laboratory and at Dugway Proving Ground and by Co^{60} sources at the University of Michigan and elsewhere. Except for Co^{60} sources, the fuel elements are now the only readily available gamma sources of high activity. Fuel elements, however, have the disadvantage of changing dose rate rapidly (see Fig. 4.11) and require frequent handling and transfer to maintain a uniform radiation field. Handling and transfer is usually performed under water in canals, or in hot cells with heavy shielding and long handled tools or slave manipulators. Such equipment is relatively expensive and is not always available.

USE OF FUEL ELEMENTS IN MOBILE FOOD IRRADIATION FACILITY

Brownell, Patterson and Purohit[85] pointed out that among products best suited to irradiation are seasonal crops. Since harvest seasons come at different times of the year in different parts of the country, the problem of providing a radiation source is geographic as well as seasonal. A mobile facility is therefore ideal for irradiating crops, at the location where they are to be stored, immediately after harvest. After the crop of one locality has been treated, the unit could be moved elsewhere to treat another crop, keeping the facility in more or less continuous use, with a consequently lower operating cost per unit of product irradiated. Or, to illustrate another advantage inherent in a mobile unit, it can be taken to a seaport or border city and used to irradiate imported foods that might otherwise be quarantined.

Due to considerations of weight, the only way to transport a large mobile irradiation source by land is to use a rail vehicle. Even when mounted on a railroad car, the complete unit must not be so heavy that the car cannot be safely moved into railroad sidings in agricultural areas. In providing adequate shielding to attenuate high-energy gammas, the weight problem becomes extreme. The shielding can be held to a minimum by keeping the volume of the irradiation chamber as small as possible.

To allow passage through railroad tunnels, car width should not exceed $8\frac{1}{2}$ ft, and its height should be no more than 14 ft above the rails. For safety reasons the center of gravity should not be more than 7 ft above the rail.

A shipping cask containing a source of usable size is heavy, in this case 25 tons. It was decided that such a container could best be handled by picking it up with a crane and lowering it through a hole in the top of the car. Cranes capable of such hoists are common along rail sidings in most areas.

RADIOISOTOPES PRODUCED BY NEUTRON CAPTURE AS FOOD IRRADIATION SOURCES

Excess neutrons produced inside the reactor can be employed to produce radioactive isotopes for gamma radiation. Assuming that an average of 2.5 neutrons are liberated per fission of uranium nucleus, 1 neutron is needed to maintain the chain reaction by causing a fission, 0.18 neutron will be absorbed in the U^{235} without producing fission, 0.25 neutron is absorbed in the structural material, and 0.25 neutron will leak out of the assembly, leaving 0.8 neutron available to produce radioisotopes. The main problems are to increase the number of neutrons available to produce radioisotopes and to decrease parasitic capture of neutrons. For efficiency, the neutrons should be absorbed uniformly. The use of highly enriched U^{235} fuel would be necessary, eliminating or minimizing resonance capture of neutrons by U^{238}. The selection of an element to absorb excess neutrons is based on the following considerations:

1. Total output of gamma energy (Mev) per neutron absorption.
2. Half-life of gamma emitter.
3. Neutron absorption cross section of parent material.
4. Spectrum of the gamma radiation.

Table 4.8—ELEMENTS CONSIDERED TO BE MOST PROMISING AS GAMMA PRODUCERS[1]

Element	Mev gamma per neutron absorbed	σ_{th} absorbed,* barns	Half-life, important isotopes	Energy of major gammas, Mev
Sodium	4.14	0.50	15.0 h	2.76, 1.38
Cobalt	2.53	37.0	10.4 mo	0.059
			5.28 y	1.33, 1.17
Lanthanum	2.44	8.9	40.0 h	2.59, 1.60, 0.82, 0.49, 0.44, 0.33, 0.27
Scandium	2.09	24.0	20 s	0.18
			85 d	1.12, 0.89
Indium	1.81	190.0	13.0 s	none (?)
			54.0 mo	2.09, 1.49, 1.27, 1.09
			50.0 d	0.19
Aluminum	1.80	0.23	2.3 mo	1.80
Gallium	1.73	2.77	14.2 h	2.50, 2.20, 0.83, 0.63, plus many others
Manganese	1.73	13.2	2.58 h	2.13, 1.81, 0.845
Cesium	1.54	29.0	3.15 h	0.13
			2.3 y	1.8, 0.61, 0.58, 0.56
Argon	1.29	0.62	109.0 mo	1.3
Tantalum	1.26	21.3	16.4 mo	0.51
			111.0 d	1.22, 1.12, 0.15, 0.06

* Thermal absorption cross section. Actually the less accurate experimental activation cross sections were used to determine the Mev of gamma per neutron absorbed in the natural element, since a knowledge of the absorption of the individual isotopes is often necessary.

Table 4.8 lists the elements that appear promising as gamma producers.[1] The element selected should have a large output of gamma radiation per neutron absorbed and a half-life of suitable length. The neutron absorption cross section should be large. Isotopes of half-life less than 1 min are considered to be impractical because of excessively rapid decay. Neutron

irradiation times of about 4 half-lives are needed for a gamma emitter to reach equilibrium value at which the rate of decay equals the rate of formation. This argues against the use of a reactor to produce isotopes having long half-lives, such as Co^{60} (5.3 years). Since sources are required in the megacurie range, a decay time of 10 to 15 or more half-lives must be allowed before maintenance, repair, decontamination, and disposal of waste can be undertaken. This tends to limit selection of the source material to radioisotopes having a half-life of only a few hours. Also, since the isotope used must be mixed with a moderator, the compound containing the isotope should have a fairly high solubility.

Consideration of the elements listed in Table 4.8 shows that indium most nearly satisfies all the requirements. It has a gamma energy output of 1.81 Mev per neutron absorbed, and its half-life is 54 min. Indium has a thermal neutron capture cross section[86] of about 190 barns, as shown in Fig. 4.12 and a characteristic main resonance peak with a much higher cross section for neutrons of 1.44 ev.

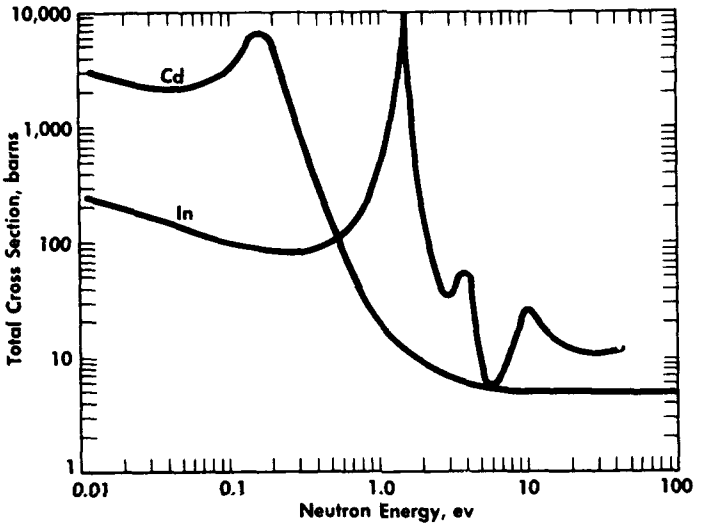


Fig. 4.12—Indium and cadmium resonance spectrum.

From the point of neutron absorption, indium has definite advantages over other elements. It may be used in the form of an indium sulfate solution which is quite soluble but is very acidic. In addition to the indium sulfate solution, manganese sulfate, liquid sodium, and solid scandium are worth consideration.

Important problems in utilizing excess neutrons are neutron leakage and neutron absorption. Excess neutrons are lost by:

1. Fast neutron leakage.
2. Thermal neutron leakage.
3. Thermal neutron absorption in:
   a. Moderator.
   b. Structural material.
   c. Control rods.
   d. Fission products.

Water-moderated reactors will be of minimum size for a given efficiency of neutron absorption. Graphite and beryllium oxide moderated reactors are particularly suitable with sodium as the coolant.

The radioisotope to be used as a radiation source can be produced in the core, blanket, reflector, and coolant of a reactor. A comparative study of core and blanket production of isotopes was undertaken by the Internuclear Co., Inc.[1] In this study, it was assumed:

1. The reactor core contained 90 to 95 per cent enriched U^{235}.

2. The absorption of thermal neutrons by the control rods, poisons such as Xe^{135} and Sm^{149}, the uranium nuclei (U^{238} and U^{236}) and the fission products, was about $1/10$ of U^{235} absorption.

The calculations were carried out using two-group multiregion diffusion theory. A heterogeneous reactor was selected containing MTR type fuel elements and moderated by ordinary water.

PRODUCTION OF GAMMA EMITTER IN REACTOR CORE

Neutron absorption efficiency in the parent material was studied[1] by calculating the effect of changing the metal-to-water ratio, reactor core size and the reflector material.

For a given core size, the neutron absorption efficiency, Z, becomes greater with the increase in metal-to-water ratio; where

$$Z = \frac{\text{neutrons absorbed in parent gamma producer}}{\text{neutrons absorbed in } U^{235}}$$

The efficiency increase is expected as a result of the decrease in fast neutron leakage. The neutron absorption efficiency also increases with the volume of parent material, which is in essence equal to the volume of the moderator. If a water reflector is replaced by graphite, Z is found to increase. In the case of values of Z much above 0.4, however, there is a positive temperature coefficient of reactivity.[1] The excess reactivity above the design value must be regulated with control rods or by changing the concentration of the parent material. Introducing control rods causes neutron absorption and results in the lowering of Z. On the other hand, to control the reactivity by changing the concentration of the parent material solution would be difficult because of the problem of developing a sensitive control with rapid response.

PRODUCTION OF GAMMA EMITTER IN REACTOR BLANKET

The "nuclear independence" of the reactor blanket from the core permits the maximum freedom of nuclear and mechanical designs if the gamma emitter is produced in the blanket. In such a design the reactor core may be a long cylinder to diminish leakage of neutron flux from the ends and to increase the radial flux leakage, L_r, to the blanket. The leakage can be increased by adjusting the core diameter and the critical mass. The radial flux depends entirely upon the core design. Once the design has been optimized, changes in the nuclear properties of the blanket do not appreciably affect the criticality of the core or the radial flux leakage. Neutrons leaking from the radial surface are fast, and must be slowed to thermal velocities before they can be absorbed. The blanket moderator may be chosen independently of the core moderator; it can be selected entirely on the basis of the production efficiency of the gamma emitter in the blanket. To increase the neutron absorption efficiency, Z, the blanket must contain an adequate amount of the parent material. To utilize all the neutrons available, the structural material (pressure vessel shell, liner for corrosion resistance, etc.) between the core and blanket must have a low thermal neutron absorption cross section and should allow the fast neutrons to pass through without attenuation. On the other hand, the structural material outside the blanket should be a good fast-neutron reflector.

Neutron absorption efficiency in the blanket is increased by increasing the blanket volume and by increasing the flux leakage L_r. This is accomplished by decreasing the volume of the core. Therefore, when a blanket is used to produce the gamma emitter, the core volume may be decreased and the blanket volume increased. For the same efficiency of neutron absorption, the volume of parent material required is smaller for production in the blanket than when the gammas are produced in the core.

Figure 4.13 shows the effect of blanket thickness on Z, the neutron absorption efficiency in the parent material, and Z_b, the neutron absorption efficiency in the light water blanket.[1]

In considering the design of a reactor for food irradiation (the FIR) for the Army, a comparative study using different types of blankets was made by the Internuclear Co., Inc.[1] The basic reactor specifications used in this comparison were as follows:

Height of core	90 cm	Reflector saving SR	4.0 cm
Metal-to-water ratio	Y = 0.7	Radial fast neutron leakage, L_r	0.65 cm
Core diameter	44.87 cm		

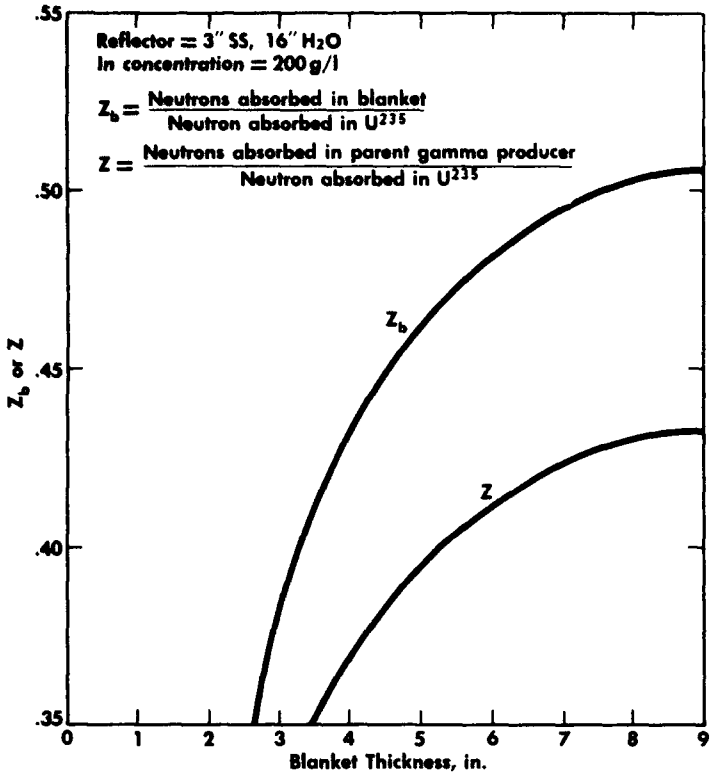


Fig. 4.13—Effect of thickness of water blanket on neutron absorber efficiency. (Courtesy of Internuclear Co.)[1]

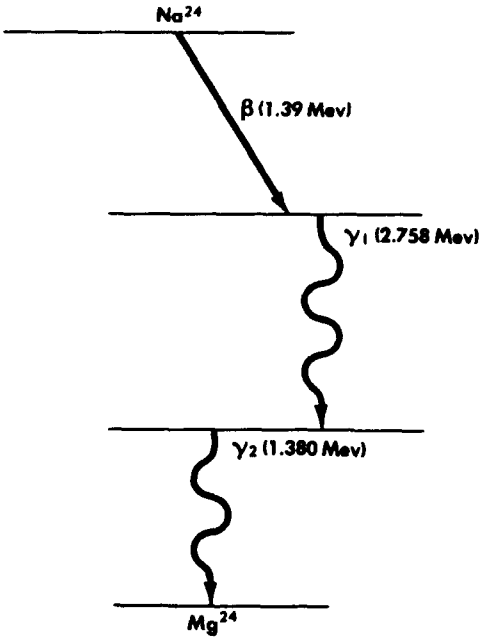


Fig. 4.14—Decay scheme of sodium.

The calculations were made on an IBM computer using two-group multiregion diffusion theory.

The blanket materials studied were:

Ordinary water + indium sulfate solution
Heavy water + indium sulfate solution
Heavy water + manganese sulfate solution
BeO + liquid sodium

Ordinary Water (H_2O) Blanket

An ordinary water blanket of 3- to 9-in. thickness containing 200 to 600 g/liter of indium sulfate was investigated.[1] The reflectors surrounding the blanket consisted in one case of 16 in. of water in a 3-in. thick stainless steel tank, and in the second case of 2 ft of graphite with a $\frac{1}{8}$-in. stainless steel plate separating the blanket from the reflector. The maximum neutron absorption efficiency that can be obtained with either of these reflectors is about 0.45 to 0.55, which gives a gamma energy output of about 0.5 to 0.6 per cent of the reactor power output.

Heavy Water (D_2O) Blanket

The absorption of neutrons in an ordinary water blanket competes with the neutron absorption by the parent material, because the thermal neutron absorption cross section of water is fairly large (σ_a = 0.658 barn). If the ordinary water is replaced by heavy water, which has a

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low absorption cross section (σ_a = 0.00282 barn), the nuclear absorption efficiency is increased for a blanket thickness greater than 9 in. If the blanket thickness is less than 9 in. (about 23 cm), the neutron absorption efficiency, Z, will be lower than in the case of ordinary water.

Beryllium Oxide (BeO) Blanket

If liquid sodium is the parent material, a BeO moderator would be used in the blanket. The use of sodium gives gamma energy equal to 1 per cent of reactor power at Z = 0.4, which is large compared to that produced by an indium sulfate solution, but neutron absorption efficiency is lower than in the previous cases. The volume of sodium required is about twice the volume of indium sulfate solution for Z ~ 0.4. Liquid sodium as both gamma emitter and coolant appears to be more promising than the use of sodium in the blanket only. This design will be discussed separately.

The discussions comparing the use of the core with the use of the blanket concluded that producing radioisotopes in the blanket is more efficient than in the core. In addition, the nuclear independence of the blanket favors its use over the core. Both ordinary and heavy water moderated blankets are more efficient for small reactors. In the case of large reactors, however, both the beryllium oxide and graphite-moderated blankets have advantages over ordinary and heavy water and would be preferred.

PRODUCTION OF GAMMA EMITTER IN SODIUM COOLANT

It appears possible that liquid sodium can serve both as coolant and parent material for the gamma emitter. Liquid sodium has a low thermal neutron absorption cross section of 0.49 barn, but a large amount of gamma energy is produced per neutron absorbed. For example, with a Z of 0.4, the energy of the gamma photons from sodium is 1 per cent of the reactor power. This energy output is high when compared to the indium sulfate solution. Sodium emits two gamma rays after neutron capture, one of 2.76 Mev and the other of 1.38 Mev, according to the decay scheme given in Fig. 4.14.

The half-life of $_{11}Na^{24}$ is 15 hr. The transformation of Na^{23} into Na^{24} that takes place is

$$_{11}Na^{23} + {}_0n^1 \rightarrow {}_{11}Na^{24} + \gamma$$

When liquid sodium is the reactor coolant, two types of moderators for fast neutrons are used, either graphite or beryllium oxide. In a reactor for food irradiation, liquid sodium might be used both as coolant and as the parent material for producing the gamma emitter in the core and blanket.

Neutron absorption in the blanket compared with that in the core was discussed for water-moderated reactors. This design would require an extra sodium loop for heat removal, if the heat produced by the reactor is to be recovered as process heat or used to generate electric power.

Beryllium Oxide-Moderated Sodium-Cooled Reactor

In this design, the core of the reactor would contain beryllium oxide blocks as the moderator. Though gamma energy output is considerably higher, the sodium-cooled, beryllium oxide system appears less efficient because a large volume of sodium is retained outside the core in the heat removal equipment. The total volume of sodium (about 500 to 900 cu ft) is about five times that of the reactor core volume. The need for large volumes of sodium indicates that beryllium oxide-moderated reactors could prove efficient, if large irradiation facilities are required.

Graphite-Moderated Sodium-Cooled Reactor

A graphite-moderated reactor using sodium requires a larger critical mass as compared with the beryllium oxide moderated reactor for a given neutron absorption efficiency, Z. For a given volume of sodium, Z is lower for graphite than for beryllium oxide. Compared with beryllium oxide-moderated reactors, graphite-moderated reactors are larger and are less efficient.

COMBINED PRODUCTION OF GAMMA EMITTER IN CORE AND BLANKET

A combination of core and blanket production of the gamma emitter at first appears attractive because of the low critical mass required and the possible increase in neutron absorption efficiency. The problems of precipitation in the fluid streams, however, in the fouling of heat transfer surfaces and the handling of large volumes of parent materials, make the combined core and blanket production less promising.[1]

REACTORS WITH REMOVABLE SOLIDS AS GAMMA RADIATION SOURCES

Parent material, instead of being used as liquid (as with indium sulfate solution, manganese sulfate solution and sodium), may be solid. The solid may be in thin plates, rods, or hollow cylinders surrounding the core. Scandium of 85 days half-life has been considered for such a source.[1] Each source plate would be cycled between the reactor and the facility about every 2 half-lives to maintain a sufficiently high level of activity.

The advantages a solid parent material compared to a liquid parent material are:

1. No leakage of radioactive material.
2. Elimination of corrosion, precipitation, and decontamination problems.
3. Simpler reactor operation.

On the other hand, the solid gamma emitter has a number of disadvantages in comparison with an indium sulfate/water system. Some of the disadvantages are:

1. Time required for the build-up of gamma "power" before the facility can be started is about 1 year.
2. Obtaining short irradiation time, which is possible in the other facility by varying the concentration, is difficult.
3. Control of the maximum and minimum dose in the irradiated product may prove troublesome.

MULTIPURPOSE REACTOR TYPES

The reactor does not necessarily require special and expensive design features to serve a variety of purposes. Homogeneous reactors using aqueous fuel solutions or liquid metal fuel could, with few or no design changes, utilize fission product gases for irradiation purposes. Small bleed streams of fuel solution could be removed continuously, perhaps as the stream is being chemically processed, and the activity in this stream used to irradiate materials. Neither operation would detract from the ability of the reactor to produce heat and electrical power.

If reactors prove feasible when used for a single purpose (e.g., as a source of electric power, process heat, or gamma radiation), two or more applications might well be combined to considerable advantage. To meet competition, the nuclear plant should take maximum advantage of the value of every product.

Various types of reactors might be suitable for this multipurpose service. Table 4.9 lists them and summarizes the possible use of each. Gas-cooled graphite-moderated research reactors, such as the Brookhaven BNL, the British BEPO, and the Oak Ridge X-10, do not achieve temperatures high enough for process steam or power. Mainly they supply neutrons for research studies and for producing isotopes.

The British CO_2-cooled reactors, such as the Calder-Hall and subsequent similar reactors, are designed both for producing power and as sources of irradiation, through direct employment of the fission product gases released almost instantaneously from the fuel solution. Heterogeneous reactors have completely "canned" or "clad" fuel elements in which fission products are immobilized so that they cannot be used until the elements have been removed from the reactor.

Aqueous homogeneous and liquid-metal-fuel homogeneous reactors, and heterogeneous reactors cooled by molten metal, can be considered for multipurpose use. Fuel solution and fission gases from homogeneous reactors can be used as gamma radiation sources. The efficiency of gamma radiation production by n,γ reaction—using the parent material in the core, blanket, reflector, and coolant—has been considered in previous sections. In heterogeneous

Table 4.9—SUMMARY OF REACTOR USES

Type	Process steam	Useful power	Irradiation
Pressurized water (STR, PWR)	Yes	Yes	*
Boiling water (Borax) (EBWR)	Yes	Yes	*
Molten metal cooled (SGR, EBR, APDA)	Yes	Yes	*†
Gas cooled (BNL, ANL, X-10)	---	---	*
Aqueous homogeneous (HRE)	Yes	Yes	Yes
Molten metal homogeneous (LMFR)	Yes	Yes	Yes

* This type of heterogeneous reactor, using separate fuel elements, would not be adaptable for irradiation service from fission product gases. The spent fuel elements could be used as radiation sources, but this would not be a primary function of the reactor.

† Molten metal coolants, such as sodium, become radioactive under neutron bombardment, and emit gamma energy. This energy could be used as an irradiation source by diverting a side stream from the main coolant stream.

reactors cooled with molten metal, the coolants emit gamma radiation as a result of the n,γ reaction. Thus, liquid sodium can serve as both coolant and gamma emitter.

Present designs for heterogeneous reactors provide little opportunity to use the great gamma energy of fission gases. The homogeneous reactor, therefore, appears more desirable for multipurpose use. Manowitz[25] suggested such multiple use in a chemical plant, and Brownell et al.[31] proposed the use of an LMFR in a rubber tire plant. If gamma radiation were combined with a process heat function, the LMFR would have the added advantage of a higher temperature cycle than that of the aqueous homogeneous reactor.

LIQUID METAL-COOLED HETEROGENEOUS REACTORS

Several reactors planned or being built use heterogeneous fuel elements cooled by a molten metal. Such reactors would include the sodium-graphite reactor (SGR) and the fast-breeder Enrico Fermi Reactor being built by the Power Reactor Development Co., Inc. at Monroe, Mich. Sodium is usually employed because of its excellent heat transfer properties and low cost. This metal becomes radioactive and emits gamma radiation because it absorbs neutrons to give the n,γ reaction mentioned earlier.

Figure 4.15 illustrates a reactor system in which two molten-metal loops are employed in the same way as in the APDA breeder reactor. In this system, the radioactive sodium from the reactor transfers its heat to a second sodium loop which then turns water in a third loop to steam. The sodium in the second loop does not become radioactive, nor can radioactive sodium in the reactor or primary loop enter the steam lines as a result of superheater or steam generator tube failure. Part of the radioactive sodium is by-passed from the primary loop through the irradiation facility. If the materials being irradiated are adversely affected by heat, some thermal barrier must be placed between the container wall and the irradiated material.

It has been reported that 0.3 curie/cm^3 is the activity expected in the sodium coolant.[87] If activity levels of about 100,000 curies are to be achieved using sodium coolant, then a volume of about 300 liters of sodium will be required in the irradiation facility. This is equivalent to the volume illustrated in Fig. 4.16 and does not appear to be unreasonable in either cost or size. The volume, however, might be excessive for radiation sources of greater activity. The hollow cylindrical configuration would be suitable for irradiation of solids or liquids where passage of material is around the outside or through the center. Slab configuration is very adaptable to irradiation of bulky solids where the materials are brought around both sides of the slab by conveyor or truck.

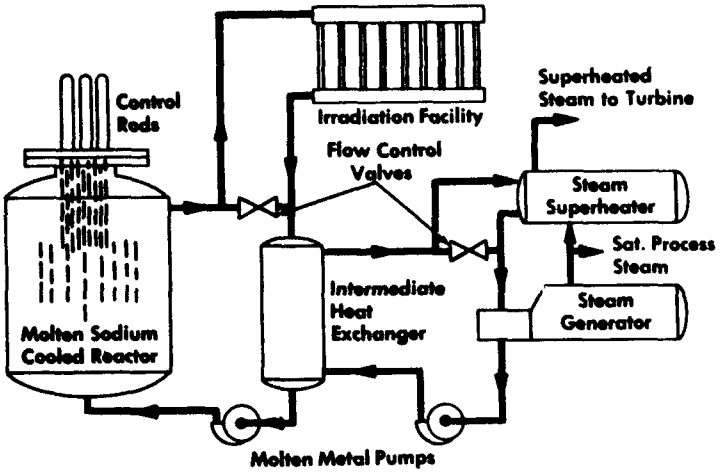
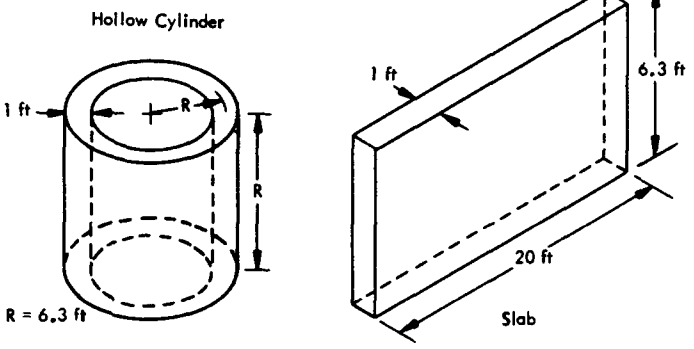


Fig. 4.15—Proposed use of a molten sodium cooled reactor for heat, power, and irradiation service.

Fig. 4.16—Configurations for radioactive sodium hold-up to achieve total activity level of 100,000 curies.



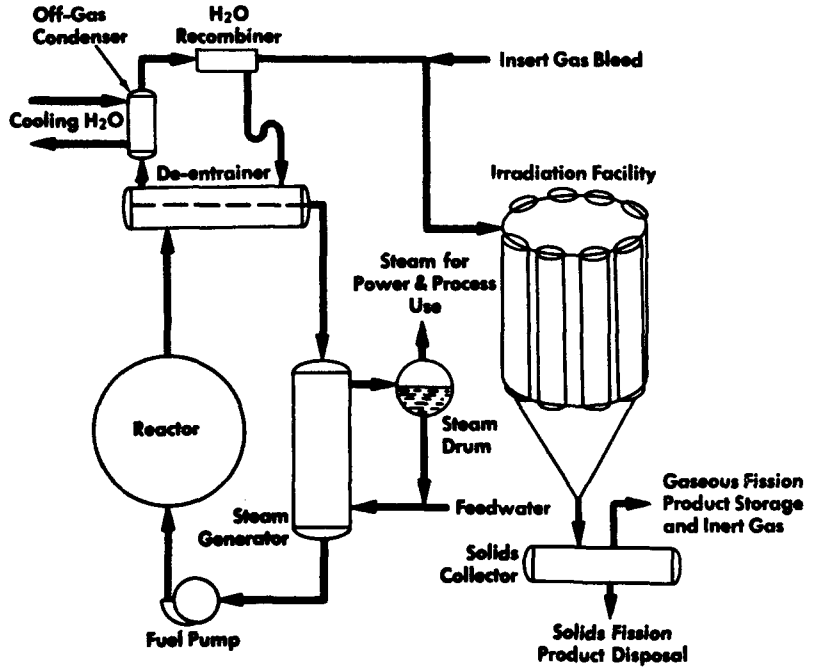


Fig. 4.17—Proposed use of an aqueous homogeneous reactor for heat, power, and irradiation service.

AQUEOUS HOMOGENEOUS REACTOR

The design proposed for a multipurpose aqueous homogeneous reactor is shown in Fig. 4.17. The illustration shows the reactor and components as far as the production of steam for use either in a turbine or as process steam, and the method of utilizing the gaseous fission products as an irradiation source. Most gaseous fission products decay into solid isotopes in relatively short times. The irradiation facility should therefore include means to drop these decay products into a solids collector, from which solids would be removed periodically by dissolving them in an aqueous reagent or by flushing them with water to a more appropriate storage area.

The system illustrated in Fig. 4.17 could be adapted to a reactor operating at any power level. Since the volume of gaseous fission products evolved is small—about 3.2 liters/100 g of U^{235} fissioned (see Table 4.3)—the size of the irradiation facility is not determined by any internal volume limitations but by the intended use of the facility. The time of irradiation and dose level required will thus greatly influence the size of the irradiation source, but they will have small effect on the reactor power level.

From the data in Table 4.3, the total gamma radiation available from such an irradiation facility with a reactor operating at a power level of 100 Mw would be 6.4 Mev-megacuries. Assuming the efficiency of the facility to be around 25 per cent, it is obvious that very high dose rates and consequent high capacity production of irradiated material can be achieved. Cooling the radioactive gas container will be necessary, since about 1.3×10^5 Btu/hr will be liberated as heat when the gaseous fission products decay.

LIQUID METAL FUEL HOMOGENEOUS REACTOR

The LMFR has a number of ideal characteristics for a multipurpose reactor that is to provide electric power, heat for processing, and gamma radiation. A study of the feasibility of the use of a LMFR (75 Mw) in a rubber tire plant was made at the Fission Products Laboratory of The University of Michigan by Brownell et al.[31] A combination of steam and radiation vulcanization, using fission gases as the gamma radiation source, was proposed. The core fuel solution, however, might also be used as radiation source.

The LMFR has a number of advantages over a heterogeneous reactor.[88–90] These are:

1. There is no radiation damage to liquid metal fuel, which permits unlimited burn-up of fuel; radiation damage to solid fuel elements limits the burn-up of fuel.
2. The LMFR has high specific power.
3. High temperatures are produced, giving high thermodynamic efficiency.
4. Low vapor pressure in the LMFR core reduces the problems of the core vessel.
5. Liquid fuel can be handled easily by pumping; it does not require complicated mechanical devices, as do heterogeneous reactors.
6. Preparing liquid fuel is simpler since fuel elements need not be fabricated. Absence of cladding reduces the number of neutrons captured, thereby increasing the neutron economy.
7. Reactor control and operation is less complicated because of simplicity of the reactivity control. In the event of mechanical failure, the fuel can be pumped out rapidly to a noncritical storage assembly.
8. Fission product removal is continuous and relatively cheap.
9. Capital cost is low.

CHARACTERISTICS OF CARRIER METAL

The carrier liquid metal for the uranium fuel must have several characteristics. These include: low vapor pressure, low viscosity, high thermal conductivity, and low neutron capture cross section. It must also be capable of dissolving sufficient uranium to provide a critical mass. Low neutron capture cross section is the most important. Lithium-7, Pb^{208}, and bismuth are suitable but the very high cost and low uranium solubility prohibits the use of Li^7 and Pb^{208} Liquid bismuth has a low neutron capture cross section for thermal neutrons, $\sigma_c = 0.030$ barn. Its melting point, although low, is enough to dissolve a sufficient amount of uranium (0.5 per cent of uranium in bismuth at 500°C). The solubility curve of uranium in bismuth is shown in Fig. 4.18.

A disadvantage of using bismuth is that in liquid form it is corrosive, and dissolves most structural materials. Moreover, it dissolves metal at the high-temperature side of a temperature cycle and deposits at the low-temperature side, resulting in plugging. By adding inhibitors, such as small quantities of zirconium and magnesium, it is possible to minimize the mass-transfer problem.

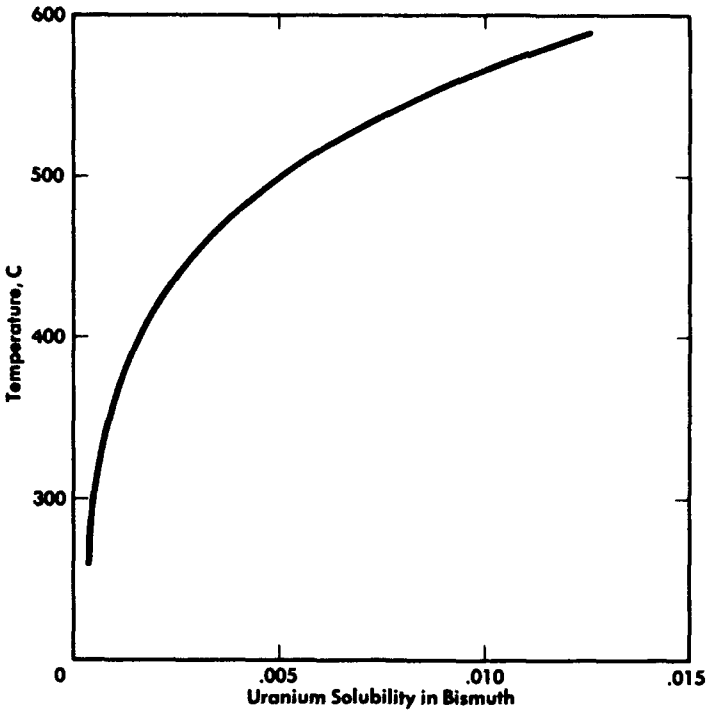


Fig. 4.18—Solubility of uranium in bismuth.

Since bismuth is a heavy element, it causes velocity erosion because of the large kinetic energy of moving bismuth. Dwyer[91] has proposed to limit its linear velocity to 10 ft/sec.

Bismuth is converted into RaE by the neutron capture process, the latter radioisotope decaying into polonium by emission of a beta particle. Polonium is toxic and difficult to handle and therefore presents a problem of removal.

REACTOR CORE

The corrosiveness of liquid bismuth severely limits the choice of structural materials. They must not only be corrosion-resistant but also should have low neutron capture cross section. Graphite fulfills these two requirements, but is a poor material for construction. Chrome alloy is reported to be satisfactory if inhibitors are added to the liquid bismuth.[91] Thus the inside layer of the core could be of graphite, with the outer shell of chrome alloy. In order to utilize the leakage of neutrons from the core and to provide for the replenishment of the spent fuel, a breeder blanket surrounding the core is suggested. The use of the breeding material would make the plant self-sufficient with regard to fuel supply. The proposed reactor core is 5 in. in diameter, contains a graphite moderator, and has channels for the passage of the liquid metal fuel. The fuel enters at the bottom and leaves at the top of the core. A spherically domed degasser is provided at the top of the core to separate the gaseous fission products.

The advantages of external cooling, however, must be considered with two major problems which need special attention.

1. The whole system must be leak-tight.
2. The whole system must be adequately shielded.

The presence of a leak in the fuel loop can be hazardous, since fission gases may leak into the atmosphere. Atmospheric oxygen may diffuse in the system through the leak and form oxides which may be deposited, immobilizing the fluid and lowering the efficiency of the mechanical pumps. It may also reduce the heat transfer rates by decreasing the flow rate of the fuel. Because the pipelines carry radioactive fuel, the whole system, including pumps, valves and accessory equipment, needs adequate shielding.

FLOW DIAGRAM

A schematic flow diagram of the multipurpose reactor radiation facility as proposed by Brownell et al.[31] is presented in Fig. 4.19 and includes an LMFR (75 Mw) and a radiation chamber. The main components of these two items are:

1. Reactor
   a. Core with graphite moderator
   b. Liquid metal fuel loop (Bi + U)
   c. Liquid metal breeder loop (Bi–Th)
   d. External secondary coolant loops
   e. Fission gas transfer loop
2. Radiation Chamber
   a. Radiation source of fission gases
   b. Steam line
   c. Monorail

The radiation chamber and reactor are shown, for convenience, side by side in Fig. 4.19, while in Fig. 4.20 the actual elevation is shown with the reactor below ground level and the radiation chamber above it.

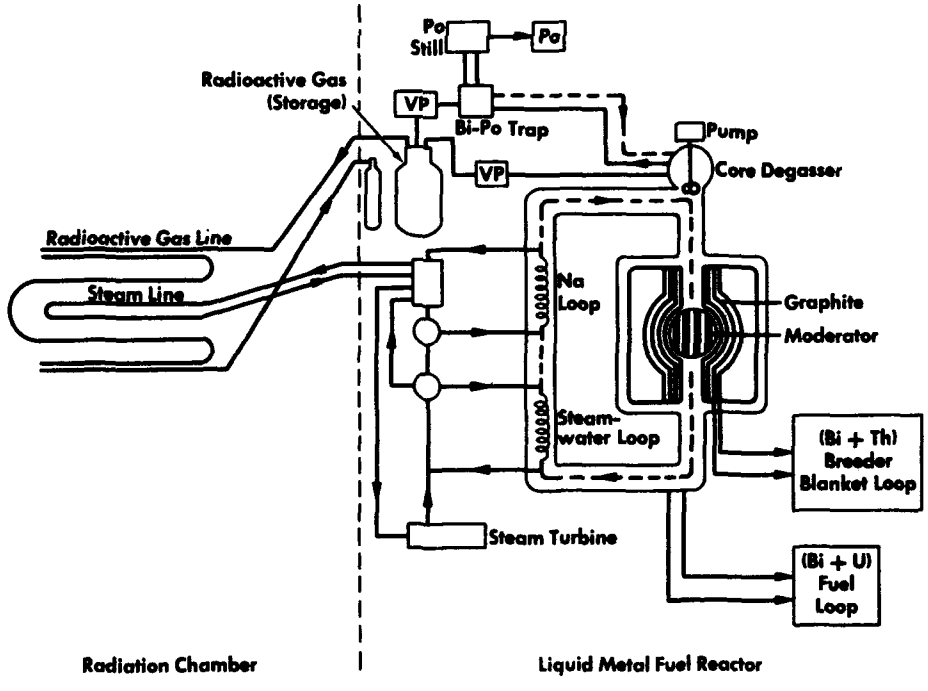


Fig. 4.19—Multipurpose liquid metal fuel reactor.[31]

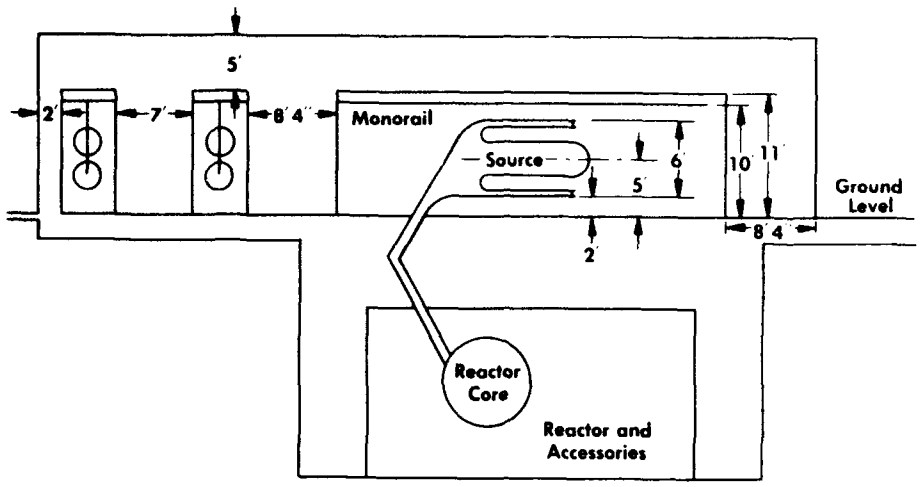


Fig. 4.20—Elevation of irradiation chamber and underground reactor.[31]



Fig. 4.21—Cobalt-60 source submerged in well of water at Goodyear Research Laboratories; note Cerenkov glow. (Courtesy of S. D. Gehman, Goodyear Research Laboratories, Akron, Ohio)

MULTIPURPOSE USE OF THE LMFR IN A TIRE PLANT

VULCANIZATION OF TIRES WITH GAMMA RADIATION

The possibility of improving the quality of rubber tires by vulcanization with gamma radiation has stimulated considerable research. Most of the larger companies in the industry possess gamma radiation facilities and have active programs in this field. Figure 4.21 shows a photograph of the Co^{60} source, with accompanying Čerenkov glow, in the Radiation Laboratory of the Goodyear Research Laboratories at Akron, Ohio. Irradiation is performed in air in a "cave" designed for 10,000 curies of Co^{60} or its equivalent. The gamma source is lowered into a well of water below the floor to permit research personnel to enter the radiation cave. Experimental samples and equipment can then be arranged for irradiation. After all personnel leave the cave, the source is raised to the perforated aluminum container shown in the center of the tire. Some experimental results of tests performed at the Goodyear Research Laboratory[34] and at the Wright Air Development Center[35] on irradiated rubber are described in Chap. 6.

Tests on rubber tires vulcanized with gamma radiation have been reported by Stockman et al.[92] of the B. F. Goodrich Co., Brecksville, Ohio. Table 4.10 lists the stocks used in these studies.

Table 4.10—STOCKS USED AT B. F. GOODRICH CO. IN GAMMA-VULCANIZED TIRES[92]

Material	Parts
Tread	
GR-S 1500/1501	100
HAF black	47.5
Processing oil	10
Carcass and liner	
Hevea blend	100
FEF black	30
Processing oil	7
Fatty acid	2
Bead	
GR-S 1002	50
Hevea blend	50
FFF black	20
SRF black	100
Processing oil	12
Fatty acid	3

The tire analysis given in the table was for a tubeless tire for a passenger car. The tire had a carcass of hevea (natural) rubber and a tread of GR-S synthetic rubber. GR-S is the synthetic rubber most often used for tire tread; it is a copolymer produced from a mixture containing 22 per cent styrene and 78 per cent butadiene. In conventional tire manufacture some butyl rubber may be used in the tire liner but, since this rubber degrades rather than cross links as a result of gamma irradiation, it was not included in the test tire. Also all antioxidants, chemical accelerators, and curing agents normally used were omitted. Dacron cord was used because it has greater resistance to radiation damage than nylon and the tire was inflated with nitrogen at 200 psi rather than with air to minimize the possibility of oxidation during irradiation.

In this test the source of gamma radiation consisted of spent MTR fuel elements with the ends cropped. The test tire was irradiated under water in the canal at the MTR site long enough to give the tire bead a radiation dose of about 4×10^7 r (see Fig. 4.22). Since it was known that the water in the canal contained some radioactive contamination, a mold with a smooth tread was used to permit removal of the tire from the mold after only preliminary decontamination.

Inspection of the tire after irradiation showed small rubber pips indicating cold flow of the raw stock in early stages of vulcanization. Hardness measurements of the tread indicated satisfactory vulcanization. Abrasion tests on tread buttons given the same radiation dose indicated that abrasion resistance had increased about 10 per cent over that for thermal-vulcanized tire tread stock.[92]

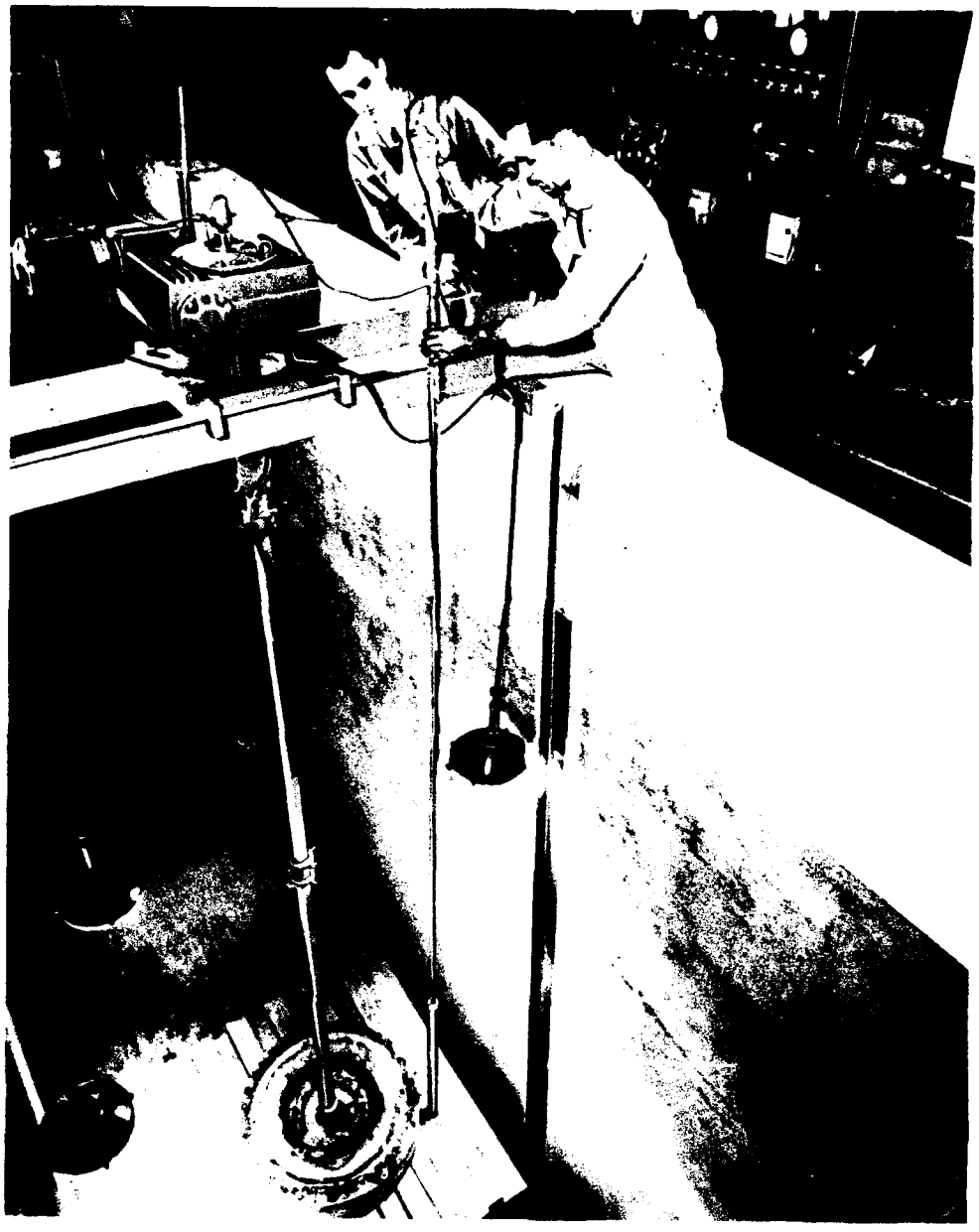


Fig. 4.22—Molded rubber tire ready for irradiation. (Courtesy of C. H. Stockman, B. B. Goodrich Co. Brecksville, Ohio)[92]

Probably the most dramatic results were in the accelerated aging test in which tire "life" is measured by the time required to reduce an initial stress to 50 per cent of its original value. (This test gives a measure of the rupture of the chain in the rubber molecule, a type of degradation important in oxidation of rubber.) The gamma-vulcanized tire proved to have a life 200 to 300 per cent longer than that of thermal-vulcanized tire stock.

From the above, and also from road tests which were in progress, it was concluded that gamma-vulcanized tires have greater resistance to oxidation and will age and wear as well or better than thermal-vulcanized tires.[92]

Stockman et al.[92] estimated that the cost of irradiating tires with a 40 mrep dose, using an electron beam from a resonant transformer, would be $7.91 per tire, not including an allowance for absorption of radiation in the mold. This cost is much too high for the highly competitive passenger-tire market. It might be lowered by reducing both the dosage required and the cost of the radiation.

Mesrobian reported that when 15 per cent styrene is incorporated in the rubber, the radiation dose for gelling rubber solutions can be reduced to less than 1 mrep.[92] This reduction by a factor of 40 would result in an approximately proportional reduction in radiation cost if it could be applied to rubber tires. However, use of radiation in the form of high speed electrons from a resonant transformer of present design would not be practical because of the great electron absorption in the mold. It is difficult to obtain uniform dosage in such a bulky object as a tire. The more penetrating gamma radiation would be desirable if intense sources were available at low cost. Such sources might be obtained from the gamma radiation of the fission gases removed from homogeneous fuel of a multipurpose reactor.

PROPOSED COMMERCIAL IRRADIATION OF TIRES

Brownell et al.[31] considered the possibility of using an LMFR in a rubber tire plant. The rubber industry requires large amounts of steam and electric power in tire production. A small modern tire plant needs about 60 Mw of thermal energy in the form of electric power and steam to produce 10,000 tires per day. (These figures apply to the smallest tire plant in which it is more economical to produce rather than purchase electric power.)[3] Large quantities of steam are required in curing the rubber. A 75-Mw reactor would have the capacity needed for power production in large plants and would produce about 75 megacuries of fission gases. This activity is estimated on the basis of the approximate equilibrium value of 1 curie/watt given by Donaldson[26] (Fig. 4.2). To utilize this high gamma activity, the fission gases must be separated from the fuel and transported to the radiation facility as rapidly as possible. The activity output given in Table 4.3 indicates that if 10 min are allowed to elapse, the total activity will drop to about 8.28 megacuries. The flow diagram of the proposed facility is given in Fig. 4.19 while the diagram of the fission gases radiation source and the flowsheet of the removal of fission gases are given in Figs. 4.3 and 4.4, respectively.

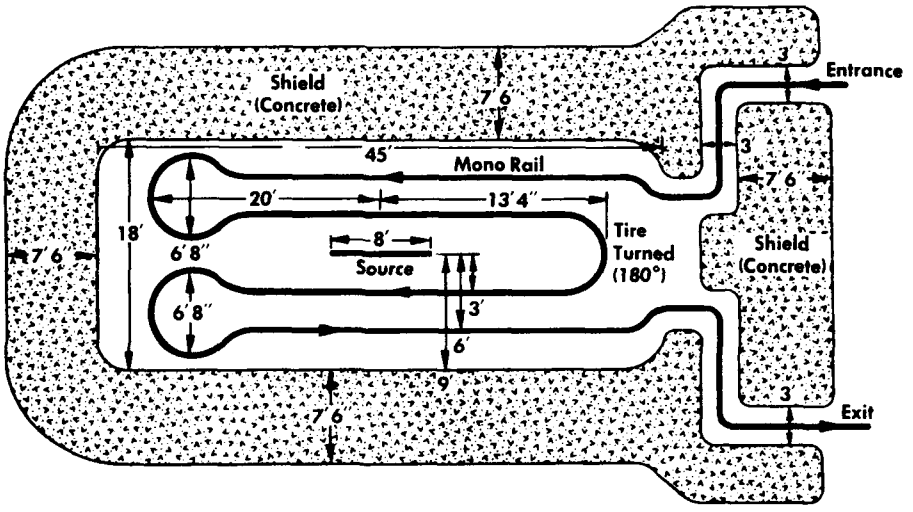


Fig. 4.23—Plan view of radiation chamber for rubber tires.[31]

A plan view of the radiation chamber is shown in Fig. 4.23. In this proposed design, rubber tires make two passes on each side of the source, at distances of 3 ft and 6 ft from the plane of the source. Tires are carried along a monorail through a 3-ft-wide labyrinth in the concrete

shield. The dimensions of the chamber are 45 by 18 by 12 ft. Chamber height is arbitrary; space permitting, it should be as high as possible to economize on the shield above the roof. The concrete shield is placed at a distance of 9 ft from the plane of the source. Total travel distance inside the radiation chamber is about 150 ft. When tires reach the source-plane axis, they are turned through 180° to insure uniform dosage. In processing 6-ft-diameter tires, one mold is placed every 6.5 ft. Tire width is assumed to be $2\frac{1}{2}$ ft and it is assumed that 14 small tires can be accommodated in the space occupied by one 6-ft-diameter tire.

The economics of a multipurpose reactor in a rubber tire plant are far from simple to estimate. Reactor and fuel processing costs, the major contributors to capital cost and operating expenses, are both difficult to predict. Published cost data are badly distorted because much research and development cost has been included. Liquid metal reactors are still in the developmental stage, and even anticipated cost data are scarce.

At present fuel processing appears to be the principal factor retarding economical use of atomic energy for power. Present method is to dissolve the fuel elements with acid prior to the reclaiming steps. This has been found costly. Pyrometallurgical processing is currently undergoing intensive study. If this yields satisfactory results, atomic energy should be competitive for power generation. However, economics of the reactor-radiation facility combination for a rubber plant appear quite favorable.

Although this facility was considered specifically for producing rubber tires, plants producing other types of high polymers, especially polyethylene, might use a multipurpose reactor to advantage.

SUMMARY

It has been shown that it is probably not feasible to utilize the prompt gamma radiation of fission and that the use of the very intense gamma radiation from the gaseous fission products, is premature. Apparently reactors for commercial processing with gamma radiation may not come into use until multipurpose or homogeneous fuel reactors are developed for this purpose.

At present, only research reactors have been designed and constructed primarily as sources of nuclear radiations. A brief description of a few of these research reactors follows.

RESEARCH REACTORS AS SOURCES OF RADIATION

Most reactors in operation before the first Geneva conference in August 1955 were research reactors. Nearly all of these reactors have ports to permit their use as sources of radiation. A number of such reactors are described in Chap. 3 and further information can be obtained from the references at the end of this chapter.

X-10

One of the early research reactors in the United States was built at Oak Ridge National Laboratory and is known as the Oak Ridge graphite-moderated reactor or X-10. This reactor operates with thermal neutrons and is air cooled. For many years it has been used as a source of neutrons for radioisotope production.[93] Figure 2.6 shows the loading face of this reactor.

BEPO

One of the best known research reactors abroad is the British BEPO (British Experimental Pile). This reactor also operates with thermal neutrons, is gas-cooled, graphite-moderated, and is similar in these and other respects to the X-10 and to the BNL (Brookhaven National Laboratory research reactor) at Upton, New York.

NRX

Canada's National Research Experiment Reactor (NRX) has been used extensively as a source of nuclear radiations. It differs from those previously mentioned in that it is both cooled and moderated with heavy water.[94] A cross sectional view is shown in Fig. 4.24

MTR

Many experimental reactors built in the United States are nuclear radiation sources. Each has been differently designed and has its own special features. Only the MTR (Materials Testing Reactor), however, will be described here, because this reactor is considered to be perhaps the best example of a U. S. research reactor as a source of nuclear radiations.

The MTR was designed primarily to test materials and components under high intensity nuclear radiation. Built at the National Reactor Test Site near Idaho Falls, Idaho, the MTR first operated at the full-power level of 30 Mw in May 1952.[85] Power level was subsequently raised to 40 Mw with an average neutron flux of 2×10^{14} and a peak of 5×10^{14} thermal neutrons/cm^2/sec.

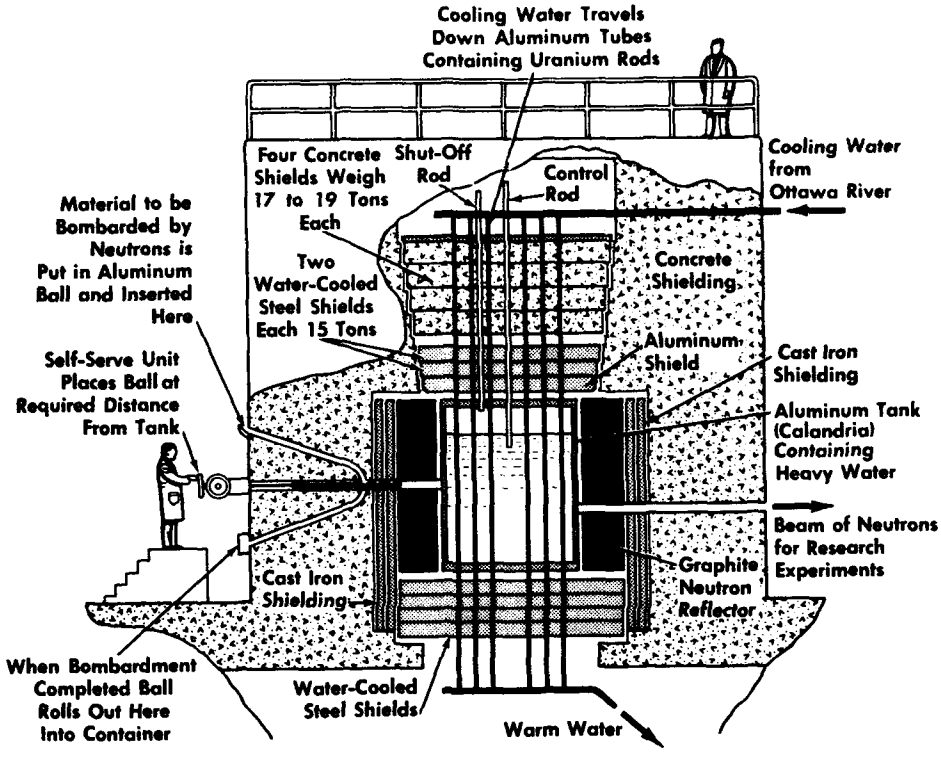


Fig. 4.24—Cross section of Canadian Nation Research Experiment Reactor (NRX). (Courtesy of Atomic Energy of Canada Ltd.)

The MTR is a thermal reactor, i.e., the majority of its fissions are produced by thermal neutron reactions. It uses fuel elements of enriched uranium and a beryllium reflector, and uses ordinary water as moderator and coolant. The core is 60 cm high and 40 by 70 cm in cross section. It is made up of fuel elements about 3 in. square held at top and bottom by grids. Figure 4.9 shows a photograph of an MTR fuel element. The small core lattice is surrounded by a primary reflector of beryllium, 3 ft high and perforated to allow passage of cooling water. Figure 4.25 shows the beryllium reflector. Only one stainless steel jacketed control rod, $1\frac{1}{2}$ in. in diameter and containing cadmium to a height of 23 in., is required for control; however, a standby rod is also provided. Eight spaces are provided for "scram" rods to shut down the reactor quickly in case of emergency.

The assembly of fuel elements, grids, control rods, and beryllium reflector fits into an aluminum tank $54\frac{1}{2}$-in. ID. This tank also contains the water moderator and coolant. Outside the aluminum tank is a "pebble zone," 9 ft high by $7\frac{1}{2}$ ft square containing graphite pebbles 1 in. in diameter. The pebbles provide additional reflector volume; they can be removed, if damaged, through a discharge chute. Outside the pebble zone, 4- by 4-in. graphite bars are stacked $9\frac{1}{2}$ ft high and 12 by 14 ft in cross section to provide a permanent reflector zone. Out-

side the graphite is a thermal shield of two layers of steel, each 4 in. thick. This shield is about 14 by 16 ft in cross section and 12½ ft high. A dense (barytes) concrete shield about 9 ft thick outside the steel shell reduces the radiation level to about $^1/_{10}$ tolerance. The resulting outside dimensions are roughly those of a 32-ft cube.

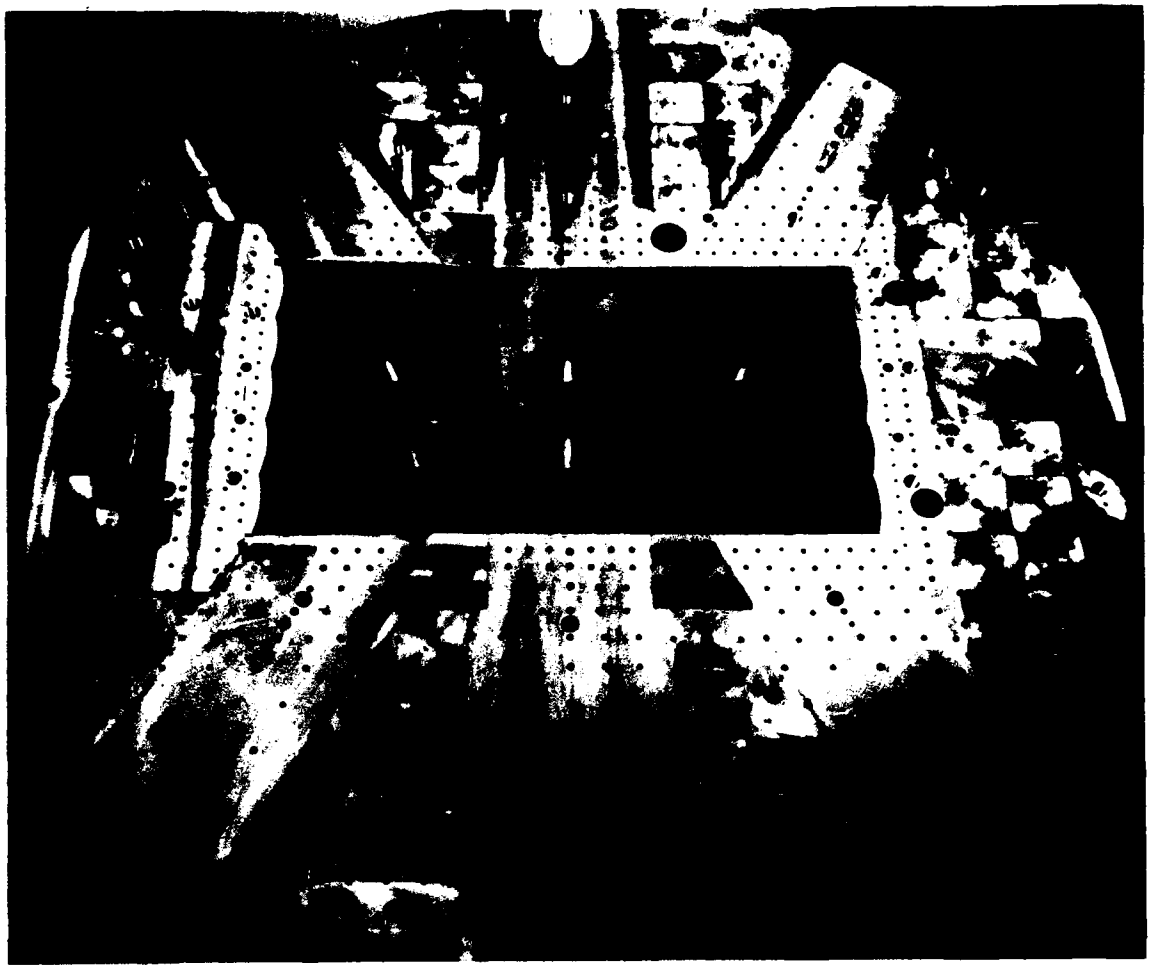


Fig. 4.25—Horizontal section of beryllium reflector for MTR.

Access Openings in MTR

Since this reactor was designed primarily for testing effects of nuclear radiations on various materials and systems, it has about 100 openings for inserting specimens.

In addition, two 4- by 4-in. holes and one 9- by 15-in. hole pass horizontally through the thermal column to the wall of the reactor tank. Thermal column and ports provide convenient facilities for studying phenomena associated with neutrons moderated to the thermal-energy range.

The MTR contains two horizontal pneumatic shuttle tubes with an inside diameter of 1 in. These are "through tubes" that pass within 1 in. of the lattice of fuel elements and are used for short irradiation times of from about 1 sec to a few minutes. For longer irradiations, 5 min to several days, there are vertical hydraulically operated shuttle tubes.

The beam holes and tubes are closed with long plugs that fit closely to prevent escape of radiation. Experimental material is placed in the selected reactor location in a new plug by adapting the plug to accept the sample.

Maximum neutron fluxes exist at the reflector positions, the horizontal beam holes, and the shuttle tubes. In the reflector position, sections of the beryllium reflector can be removed

from inside the reactor tank and test items can be placed in the cavities for irradiation. The reactor contains 17 primary experimental holes that are accessible from the exterior concrete face. Penetrating to the highest flux areas in the reactor, each has a special radiation door to shield personnel during loading and unloading. One horizontal hole, termed a "through hole," extends across the reactor from face to face. There are four additional experimental through holes in the graphite.

In addition to the horizontal ports, the MTR has 71 vertical holes, for experimentation and instrumentation, that are accessible from the top of the reactor. Most have diameters of 2 to 4 in. In loading, the experimental material is inserted into the reactor during shutdown. Heavy lead-shielded casks weighing about 20 tons, called "universal doffins," are used for removing the horizontal plugs because the parts of the plugs that are inside the thermal shield are highly radioactive. The plugs are carried in the lead coffin to a storage facility outside the reactor building.

During shutdown, radioactive materials and spent fuel elements are taken from the reactor by removing the top plug after flooding the reactor with a 20-ft column of water to provide shielding. The irradiated materials or fuel elements are then moved to a hole in the beryllium reflector that leads to a water-filler discharge tube emptying into a water canal 18 ft deep. During reactor operation, this hole in the beryllium reflector is closed with a beryllium plug.

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Chapter 5

USES OF RADIATION IN INDUSTRY

The production of radioactive isotopes in quantity as a result of nuclear reactor operation has greatly stimulated industrial interest in these newly available materials. The use of microcurie amounts of radioactive isotopes in tracer studies is discussed in Chap. 6. At the other extreme, radiation sources of megacurie strength have potential applications in the chemical and food industries and are described in detail in Chaps. 8 to 10. Now we will consider the intermediate ranges. Sources of millicurie to kilocurie strength are readily available and new applications of these sources are constantly being explored by industry.

Many uses have already been found. Radiation can be directed at or through a material to gain information about the material. The requirements are: (1) A radioactive source, (2) the material, and (3) a detector to record the radiation transmitted through, scattered by, or reflected from the material. Interactions of radiation with materials may lower the energy of the radiation as in the case of beta particles and secondary photons. All interactions decrease the intensity of the primary beam. The thickness of material, its density, its atomic number, and its structure have different effects on radiation. Major applications of this principle are: (1) Industrial radiography, (2) gauging of thickness, density, and composition, and (3) measuring the levels of various materials.[104]

The amount of radioactive material required for a gauge is usually in the millicurie range. Radiography requires quantities ranging from 1 to 1000 or more curies. Radiation from radioisotopes also is used to produce: (1) Ionization of air for static elimination, (2) excitation of phosphorescent light sources, and (3) electric current.

The application of radiations to transform nuclear energy directly into electrical and mechanical energy is being steadily pursued. Atomic batteries capable of delivering microwatt power have been constructed and designs are being improved.

This chapter begins with a discussion of industrial radiography, and the factors to be considered in selecting suitable radioisotopes and apparatus. Gauging by absorption and by scattering of radiations and the advantages of these methods over other gauging techniques are considered. Alpha- and beta-excited phosphors and the use of radioactive sources as static eliminators are discussed. The chapter ends with a summary of present and possible future techniques of directly transforming radiation energy into electrical energy.

The particular type of radiation involved in these applications depends upon the nature of the problem under investigation. Some applications may use charged as well as uncharged radiations, while others are based exclusively on the specific use of either alpha-particle, beta-particle, gamma, or neutron radiation.

INDUSTRIAL RADIOGRAPHY

Gamma radiography originated with Becquerel's discovery in 1896 that radiations from compounds containing radium will darken photographic plates.[1] Radium and mesothorium sources have been used on a limited scale for radiography but require long exposures because of their low activity. Before World War II radioisotopic radiography was quite expensive. Tenny used a number of artificial radioactive sources (including Na^{22} and Cu^{64} produced by the

Ohio State University cyclotron) as early as 1943. In recent years the construction and operation of a large number of nuclear reactors has produced comparatively cheap artificial radioisotopes.

USE OF RADIOGRAPHY

Radiography has been used for inspection of a wide variety of materials and products but has proven particularly useful in the inspection of steel products fabricated by welding. Today welding is the standard method of fabrication for pressure vessels, storage tanks, ships, and a variety of other types of equipment. Success in the use of the welding process has been due to many factors including the development of improved welding techniques, rigid qualification of welders and welding methods, stress relieving, and radiographic inspection.

Numerous failures in the past of storage tanks, pressure vessels, and ships at sea have been traced to brittle fracture of the steel. Hutton of the Newport News Shipbuilding and Dry Dock Company states "Many brittle failures in ships have been shown to be associated with welding defects. The quality of ships' welding has been very materially improved through the application of spot radiography."[115]

Failure by brittle fracture rupture can be initiated by the presence of a small crack in a section under stress. Radiographs can be used to check for the presence of cracks in welded joints. These cracks may be caused by inclusions or incomplete penetration of weld metal and might result in catastrophic failure in service.

The U. S. Navy has the most complete specifications of any agency for radiography of welds in ships. As many as ten exposures per plate may be made in highly stressed portions of a surface ship. More than 4000 separate radiographs were used for the *U.S.S. Forrestal*.[115]

The American Bureau of Shipping Rules require either trepanning of plugs from hull welds or radiography to determine soundness of hull welds. Although no specific requirements of radiography such as the Navy specifications exist for merchant ships, radiographic inspection of the welds is used extensively. Over 2500 radiographs were made of welded joints on the superliner *United States* and about 500 exposures are used for tankers or cargo ships of the mariner class.[115]

In the Soviet Union inspection of weldings and castings by gamma radiography is at present compulsory according to the rules and regulations of the Boiler Inspection Agency and other ministries and agencies.[105]

The weight of the steel used in the welded pressure vessels may be reduced 10 to 15 per cent as a result of radiography of the welded joints.[115] This is possible because of higher permissible joint efficiencies in the design of the vessel if the joints are to be radiographed.[116] Since the tonnage of steel for pressure vessels used in the chemical, petroleum, and allied industries is very large, the savings incurred by the use of joint radiography are also large.

In the past X-ray machines were the chief source of radiation for radiographs. Precious radium was sometimes used for sections which were inaccessible to these machines. The availability of artificial radioisotopes since World War II has greatly increased the versatility and lowered the cost of radioisotopic radiography. Rhoten[117] points out that "isotopes as a source of radiation do not compare to machine radiation for production line work or where a great number of exposures per day are required. Isotope exposures that may run 20 to 60 minutes could be done in from one to two minutes with high-energy machines. Although the isotopes require much longer exposure time, their cost is only a small percentage of that of a high voltage X-ray machine." These considerations plus unique advantages of radioisotopes in certain applications make it desirable to consider a variety of cases.

The small compact size of most radioisotopic sources makes it possible to radiograph joints where there is not sufficient room for X-ray equipment. Also the portability of the radioisotopes has an advantage when it is necessary to take the radiographic equipment to a welded joint, casting, or forging rather than moving the part. The fact that no electric power is required for radiography with radioisotopes is a great advantage in field work.

Over 500 U. S. firms now use radioisotopes in industrial radiography resulting in estimated annual savings of $65 million. These savings to industry are exceeded only by those obtained as a result of the use of radioisotopic gauges in industrial gauging.[104]

In the radiographing of cylindrical or spherical vessels the 360° solid angle of the radiation field produced by radioisotopes may be used to unique advantage. For example, Stewart[118] describes the radiographing of over 2000 ft of welded joints in a spherical vessel 52 ft in diameter with a single overnight exposure. Similarly the complete circumference of circumferential welds in large cylindrical storage tanks may be radiographed in one exposure. Great numbers of these tanks are used in the oil industry. Radiography of the welds of such tanks becomes increasingly important with the use of higher strength and less ductile steels which are more susceptible to failure by brittle fracture.

SELECTION OF RADIOISOTOPES

Radioisotopes available cover a wide range of energies; Table 5.1 lists some considered useful by Eastwood.[2] Radium occurs in nature, but the other radioisotopes are either produced by the n,γ reaction in a nuclear reactor or recovered from fission products. A review of the low-energy gamma emitters has been reported by West.[3] In selecting a suitable gamma emitter the following criteria are considered: half-life of the isotope, energy of gamma radiation, specific activity, ease of production and the quantity that can be produced, and cost.

Table 5.1—GAMMA RAY SOURCES FOR RADIOGRAPHY[2]

Isotope	Half-life	Radiation energy
Na^{24}	15h	1.4, 2.8 Mev
La^{140}	40h	2.5, 1.6 Mev
Ra^{226}	1590y	Av. 0.8; max. 20 Mev
Co^{60}	5.22y	1.17, 1.33 Mev
Ta^{182}	111d	1.2 Mev
Ir^{192}	74d	Up to 0.6 Mev
Cs^{137}	30y	0.66 Mev
Cs^{134}	2.3y	Up to 0.7 Mev
Tm^{170}	129d	84-kev X rays
Am^{241}	470y	60-kev X rays
Eu^{155}	1.7y	87-kev X rays

The amount of gamma radiation energy selected depends on the thickness and density of the material: with energetic radiation, small defects in the material cannot be detected, as absorption will be so slight that such imperfections will not change the intensity sufficiently to produce a visible change in the radiograph; with less penetrating radiation, excessive attenuation limits the thickness of material under examination. The energy of gamma radiation selected therefore, involves a compromise depending on the thickness and density of the material to be radiographed.

Morgan[119] states that Ir^{192} is most satisfactory for radiographing steel plate in the thickness range of $\frac{1}{4}$ to $1\frac{1}{2}$ in., Cs^{137} in the range $1\frac{1}{2}$ to 3 in., and Co^{60} for thickness greater than 3 in. For materials other than steel these thickness ranges are altered. Cesium-137 gives radiographs similar in contrast to an X-ray tube operating at about 1.25 million volts peak. A sensitivity of 2 per cent is obtained with cesium over a range of $\frac{3}{4}$ to $3\frac{1}{2}$ in. of steel.[117]

In 1959 Behal[106] reported on the results of a study of radiography of steel sections, containing holes or grooves of known dimensions. The observations are summarized as follows:

1. The maximum thickness of steel sections suitable for radiography with Co^{60} is 12 in. for 2 per cent sensitivity in the radiograph.
2. Steel sections less than 3 in. in thickness should only be radiographed with a fine-grained film if Co^{60} is used as the source.
3. Good radiographs with 2 per cent sensitivity can be obtained using Co^{60} and a fast coarse-grained film with steel sections 4 to 10 in. thick.
4. The optimum thickness of steel sections for Co^{60} radiography is 5 to 6 in.
5. For sections having a thickness greater than 6 in., "edge loss" is extensive and masking is required. If the source has less than 1000 curies of activity, the long exposure times and

extensive masking required make radiographing these sections difficult. With sources having greater than 1000 curies and a sufficiently small size (high specific activity) this difficulty can be partially overcome.

6. If the steel sections have a thickness greater than 5 in., better sensitivities can be obtained as the film is located closer to the radiographed section.

7. The shape of a defect in the section produces differences in sensitivities. Grooves are more readily discernable than holes of identical size.

As of 1959 the highest capacity radioisotope machine installed in the United States is capable of handling a radiation source of 1500 curies of Co^{60} used in the form of small wafers. According to the producers the machine can radiograph steel sections[107] having thicknesses up to 12 in.

The half-life of the gamma emitter should be fairly long, to reduce the number of replacements required in a given time and to ensure uniform intensity of radiation during its use. The radioisotope should have high specific activity (curies per gram), giving a source of fairly concentrated strength with small size. Suitable sources usually have activities at least equivalent to 2 or 3 mg of radium per cubic millimeter. The cost of the radioisotope must be less than the cost for renting a radium source of equivalent strength—radium may be rented for $2.50 to $3.00 per milligram per year.[4] The source should be fairly strong. A radiograph through 1 in. of steel can be made using a source having a strength equivalent to 500 mg of radium by exposing a fast film for 6 hr with a distance of 30 in. between the source and the film.

Clarke[5] has discussed the characteristics of radioisotopes used in radiography, comparing their efficiency, performance, and cost with equivalent X-ray generators.

Table 5.2—*RADIOISOTOPES USED IN RADIOGRAPHY*[5]

Isotope	Curies*	Size,† in.	Half-life	Output, rhm	Equivalent X-ray energy, kv	Half-value thickness in steel, in.	Cost,‡ $
Ra^{226}	½	¼	1600y	0.42	2500	1	10,000
Co^{60}	½	1/16	5y	0.67	2500	1	200
	10	3/16	5y	13.5			600
	50	5/16	5y	67.5			2,000
Ir^{192}	50	⅛	75d	27.5	700	½	400
Cs^{137}	10	¼	30y	3.6	1400	¾	2,500

*Source strengths are sufficient for radiographing in a reasonable time.
†Sizes are approximate diameters of right cylinders.
‡Costs are representative for encapsulated sources.

Table 5.2 gives some of the important characteristics[5] of Ra^{226}, Co^{60}, Ir^{192}, and Cs^{137}. These radioisotopes have been extensively employed in the radiography of steel and heavier metals. For aluminum and other light metals and thin sections of heavy metals,[6] Tm^{170} is recommended. Two other isotopes, Ce^{144} and Eu^{155}, would appear to be promising for thin-section radiography but have certain disadvantages. Cerium-144 (half-life ~280 days) emits a heterogeneous spectrum: a 134-kev gamma ray and hard gamma rays of ~2.19 and 1.48 Mev. The more energetic hard rays pass through the target material without much absorption taking place. They therefore tend to fog the film and obscure the contrast created by the less energetic rays which are absorbed to a greater degree. Europium-155 (half-life 1.7 year) would be ideal for low-energy radiography (since it emits gamma photons of 87, 105 and 130 kev) if it were not always contaminated with long half-life isotopes emitting hard gamma rays. Beta-particle excited X-ray sources such as Pm^{147} tungstate produce radiation having a suitable energy spectrum.

A comparison of the relative costs of radiography with radioisotopes and with high-voltage X-ray machines is in Tables 5.3 and 5.4. These tables show that the total capital investment and annual cost per unit for X-ray generators is very large compared to those for gamma-ray units.

Table 5.3 — COST ASSUMPTIONS FOR COMPARISON OF ISOTOPES AND X-RAY GENERATORS[5]

Items	Co^{60} sources		High voltage generators	
	10 curies	100 curies	1 Mev	2 Mev
Equipment and source cost	$ 2,500	$ 5,000	$ 30,000	$ 90,000
Enclosure	3,000	6,000	25,000	50,000
Total capital	5,500	11,000	55,000	140,000
5 year amortization and interest	1,260	2,500	12,600	32,000
Maintenance	100	100	1,600	2,000
Annual cost/instrument	1,360	2,600	14,200	34,000
Labor and overhead: (1 man, 2,000 hr/year)	12,000	12,000	12,000	12,000
Daily cost, first machine	53	58	105	184
Additional cost per machine	5.60	10.40	105	184

Table 5.4 — COMPARISON OF ISOTOPE COSTS[5]

	Handling equipment	Source	Enclosure	Annual cost
½ curie radium	$ 300	$10,000	$1,500	$1,010
½ curie cobalt	300	200	1,500	460
10 curies cobalt	1,900	600	3,000	1,260
10 curies cesium	1,900	1,500	1,500	875
10 curies iridium	1,900	1,200/year*	1,500	1,980

*This source has a half-life of 75 days.

Radiography using Co^{60} for the 1-Mev range, Cs^{137} and Ir^{192} in the intermediate range (600 to 700 kev), and Tm^{170} in the low-energy range is being developed rapidly. Gamma-radiation units used in conjunction with low-voltage X-ray generators for specific purposes may replace costly high-voltage X-ray generators for testing and inspecting manufactured products. Besides the pure gamma emitters, beta-particle excited X-ray sources (Sr^{90}) of intermediate energy have been reported by Reiffel et al.[7] Promethium-147 tungstate sources producing low-energy X radiation have been reported by Brownell, Coleman, and Fox.[102]

In addition to the developments in the United States, gamma radiography is being widely employed in nondestructive testing and inspection of manufactured good in Canada, the United Kingdom, France, and the Soviet Union.

At the Atomic Industrial Forum, Wiggin[9] reported a saving of $9000 resulting from the radiographic testing of a 50-ft diameter sphere of steel using a gamma-radiation unit which cost about $3000. Inspection of slag pots, wing sections of airplanes, weldings, and steel casting has been carried out at Ford plants[9] using Co^{60}, Se^{75}, Cs^{137}, and Ir^{192}.

A study has been made covering the use of slightly radioactive welding wire and brazing materials as a means of nondestructive examination of uniformity and penetration in a weld with a radiation monitoring instrument.[108] Metals considered were alloys of iron, aluminum, and silver. Filler wire added during welding becomes indistinguishable from the material of the weld. The use of filler wire tagged with radioactivity has the advantage over radiographic,

fluoroscopic, and ultrasonic testing of showing weld penetration. Penetration is not shown by other methods which are used to locate various kinds of defects in the weld. In the procedure developed, the intensity of gamma or beta radiation from the underside of the welded joint is used as a measure of the penetration of the weld. The nuclides considered most suitable are iron and cobalt for steel welds and silver and zinc for aluminum welds. Activities of 0.10 to 0.20 μc/in. of wire is believed to be ample.

The characteristics of some of the individual isotopes are worth consideration. Figure 5.1 shows the relative exposure times for radioactive cesium, cobalt, and iridium sources compared with radium.[5] The curves extend over the working range of the isotopes. Considerable exposure time is saved by using artificial radioisotopes as compared with the exposures necessary for a radium source. It is apparent that the "softer" radioisotopes permit radiography of thinner sections of steel than would be possible with radium.

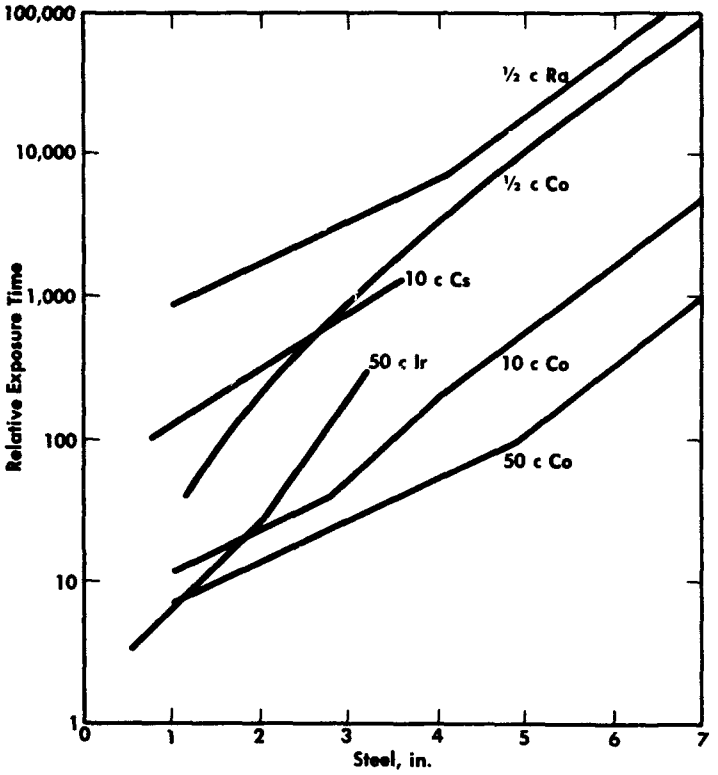


Fig. 5.1—The relative exposure times for radioactive cesium, cobalt, and iridium sources compared with radium.[5]

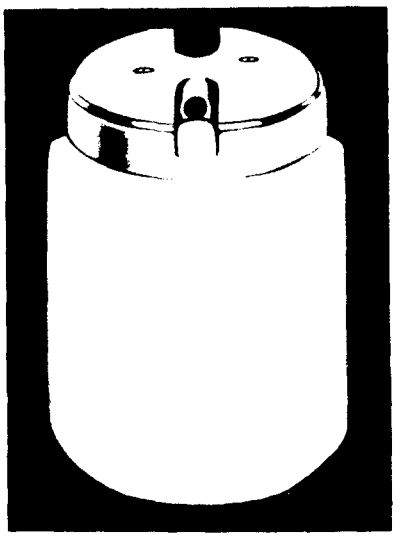


Fig. 5.2—A Co^{60} capsule used in radiography. (Courtesy of Atomic Energy of Canada Ltd.)

COBALT-60

Cobalt-60 is produced in a nuclear reactor by irradiating the natural isotope, Co^{59}, which undergoes the n,γ reaction[1]

$$_{27}Co^{59} + {}_{0}n^{1} \rightarrow {}_{27}Co^{60} \tag{5.1}$$

$$_{27}Co^{60} \rightarrow {}_{28}Ni^{60} + \beta + \gamma + \gamma \tag{5.2}$$

Cobalt-60 decays into Ni^{60} by emitting a beta particle of 308 kev energy, followed by two gamma photons in cascade with energies of 1.17 and 1.33 Mev.

The half-life of Co^{60} is fairly long, about 5.3 years. Decay results in a loss of about 5 per cent activity every 5 or 6 months. For radiography, the source strength is usually recalculated about twice a year.

Cobalt-60 sources are small and are encapsulated to prevent contamination of surfaces; the capsule is stored in a lead-shielded container. Figure 5.2 shows such a Co^{60} capsule used in radiography. The capsule is $\frac{3}{4}$ by $1\frac{1}{8}$ in., hexagonal (for ease in holding with suitable wrenches), and is fabricated of lightweight dural to minimize absorption of the radiation. The closure has a magnetic cap useful in handling. The maximum activity in this capsule is 10 curies of Co^{60} in the form of 1 mm pellets. Figure 5.3 shows a similar capsule of Co^{60} being used

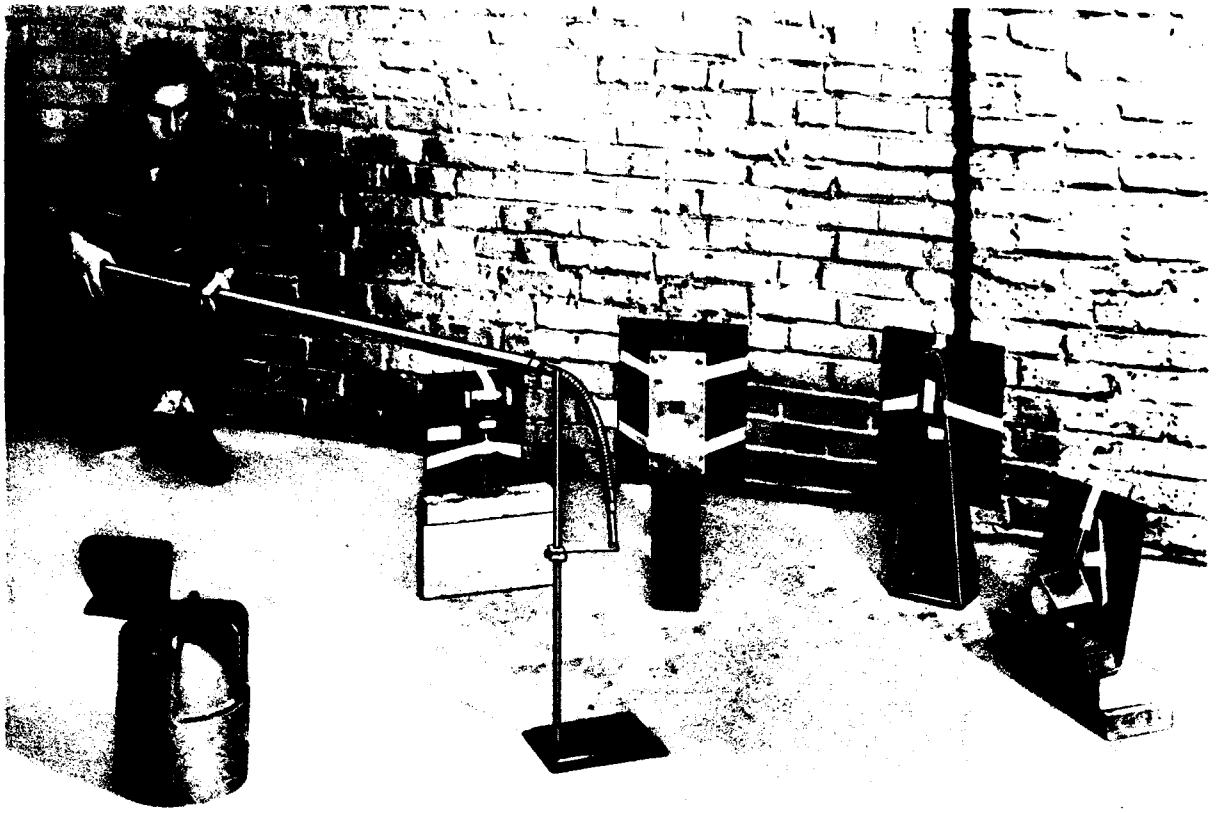


Fig. 5.3—A similar capsule of Co^{60} being used for making radiographs of four industrial parts. (Courtesy of Tracerlab, Inc.)

for making radiographs of four industrial parts. After removing the cobalt capsule from the lead shield, the operator is employing a remote-handling tool to place the capsule centrally among the parts, which are backed with plates holding radiographic film. With sources of relatively large activity, special mechanical equipment is required to handle the radioisotopes. Equipment usually consists of a cranking mechanism for remotely transferring the source from its container to the place of radiography, as shown in Fig. 5.4. The Co^{60} source is cranked from the shielded storage container (center) to the weldment (right) on which film cassettes are attached.[5] Pneumatic systems of transfer are also used.

Industrial gamma sources may also be used in a mobile shield having a port to emit a collimated beam of radiation. Such an industrial radiography unit, having a maximum capacity of 750 curies of Co^{60}, is shown in Fig. 5.5. This gamma radiography machine (GRM) model of Atomic Energy of Canada Limited (AECL) can radiograph up to 9 in. of steel.

The radiation beam from the unit shown in Fig. 5.5 emerges as a cone which at 50 in. from the source has a field diameter of 22 in. The unit requires no external power, an advantage not possessed by an X-ray machine. All operations are manual.

CESIUM-137

Cesium-137 is a typical isotope for radiography in the intermediate energy range of gamma radiation. The long half-life of about 30 years makes it a very promising source. It emits 1.2-

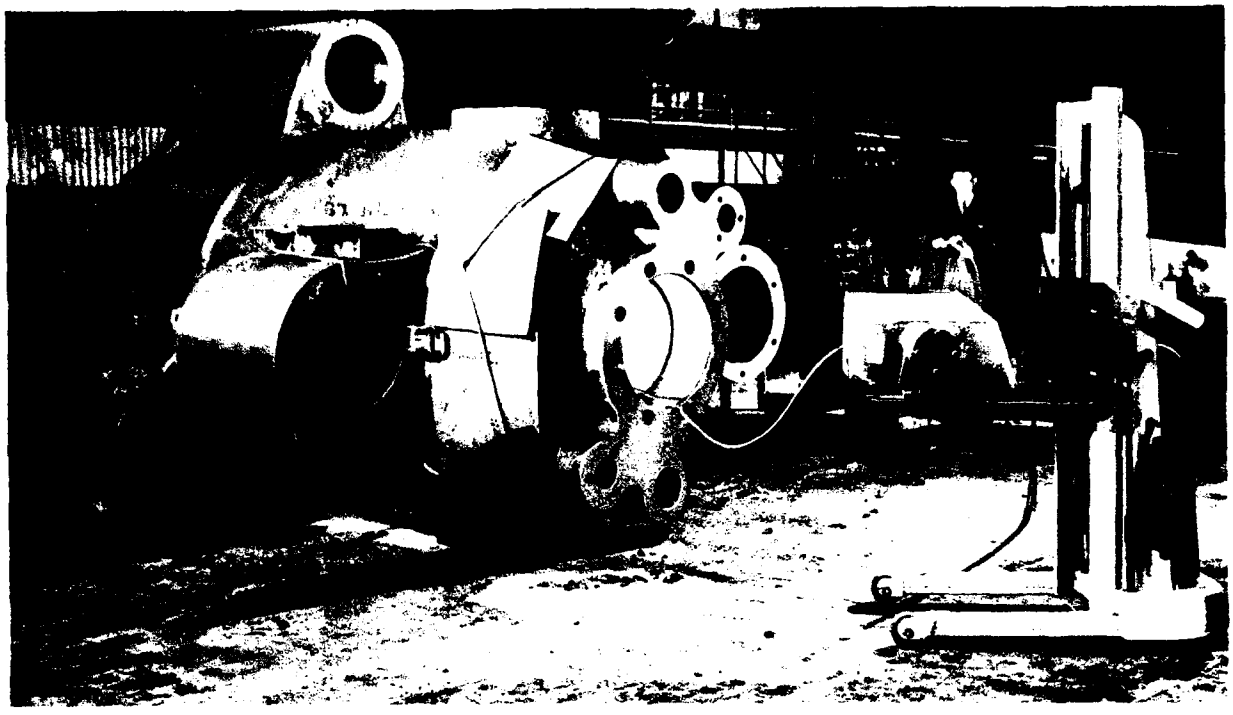


Fig. 5.4—Remotely transferring the source from its container to the place of radiography. (Courtesy of C. F. Braun & Co.)

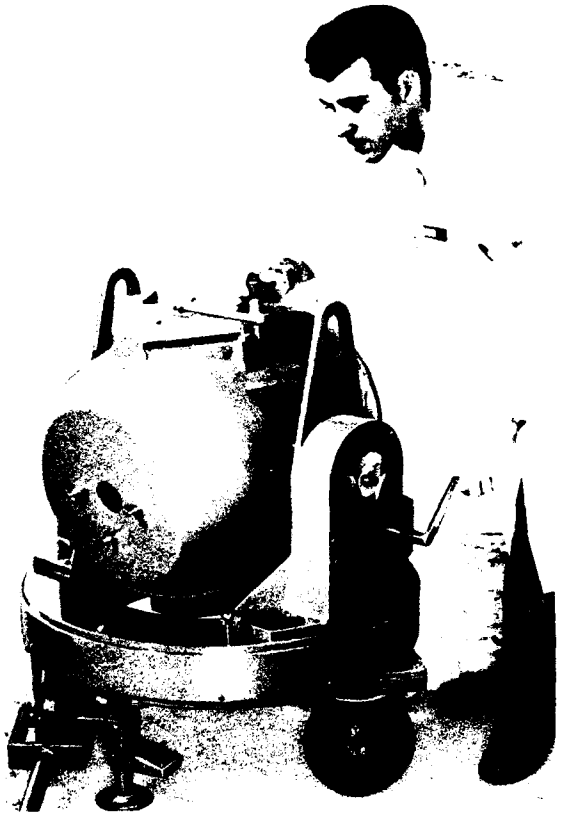


Fig. 5.5—Industrial radiography unit, having a maximum capacity of 750 curies of Co^{60}. (Courtesy of Atomic Energy of Canada, Ltd.)

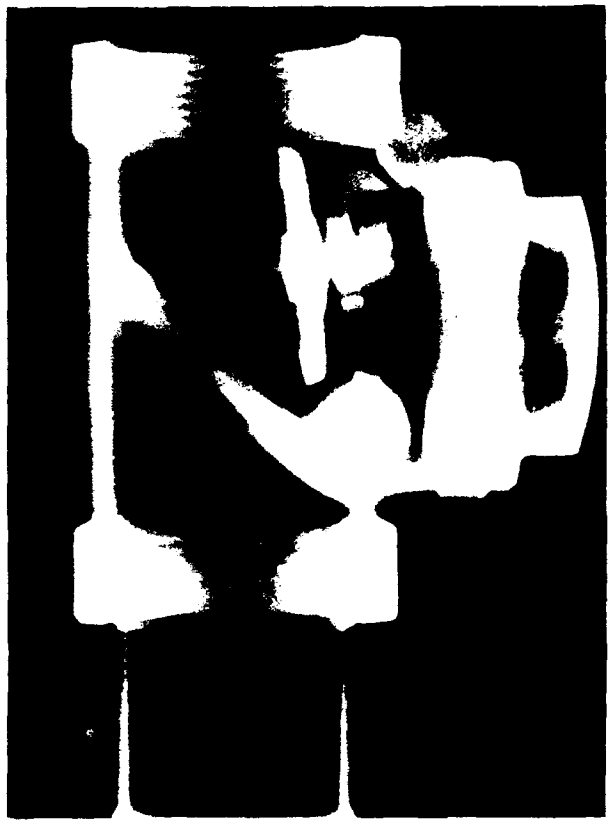


Fig. 5.6—A radiograph of a brass check valve assembly, using a Cs^{137} source.[10]

Mev beta particles in 5 per cent of the disintegrations, while 9.521-Mev beta particles are emitted followed by 0.66-Mev gamma photons in 95 per cent of the disintegrations. Cesium-137 is a fission product and is recovered in a mixture of two other isotopes, Cs^{133} and Cs^{135}, in approximately equal parts. Cesium-133 is not radioactive, while Cs^{135} is a beta-particle emitter. It has been estimated theoretically that a maximum specific activity of 23 curies/g can be obtained for cesium sulfate.[10] Cesium-137 gamma radiation is similar in performance to radiation from a 1000-kv X-ray unit.

As early as 1953, Dutli and Taylor[10] made a comparison of Cs^{137}, Co^{60}, and machines and concluded that "Cesium-137 is very suitable as a source of gamma radiation for industrial radiography .... The predicted low cost coupled with high specific activity and long half-life make it a very attractive radiographic source."

In 1957, Rhoten[117] made a comparison of Ir^{192}, Cs^{137}, Co^{60}, and machines. He states "in choosing an isotope, cesium-137 answers the demand for a source that will cover the most widely used range of thicknesses. Giving 2 per cent sensitivity on a range of $\frac{3}{4}$- to $3\frac{1}{2}$-inches of steel, with its long half-life, it is less expensive than any other source of radiation."

Rhoten[117] concludes that "As we become more familiar with cesium and its uses and as it becomes more readily available it is an isotope that is sure to become very popular." There appears to be no question about the advantages of Cs^{137} in industrial radiography.

Figure 5.6 shows a radiograph of a brass check valve assembly obtained with a Cs^{137} source of $7\frac{1}{2}$ curies. For comparison, radiographs of a darkroom timer produced with Co^{60} and Cs^{137} sources are shown in Fig. 5.7.

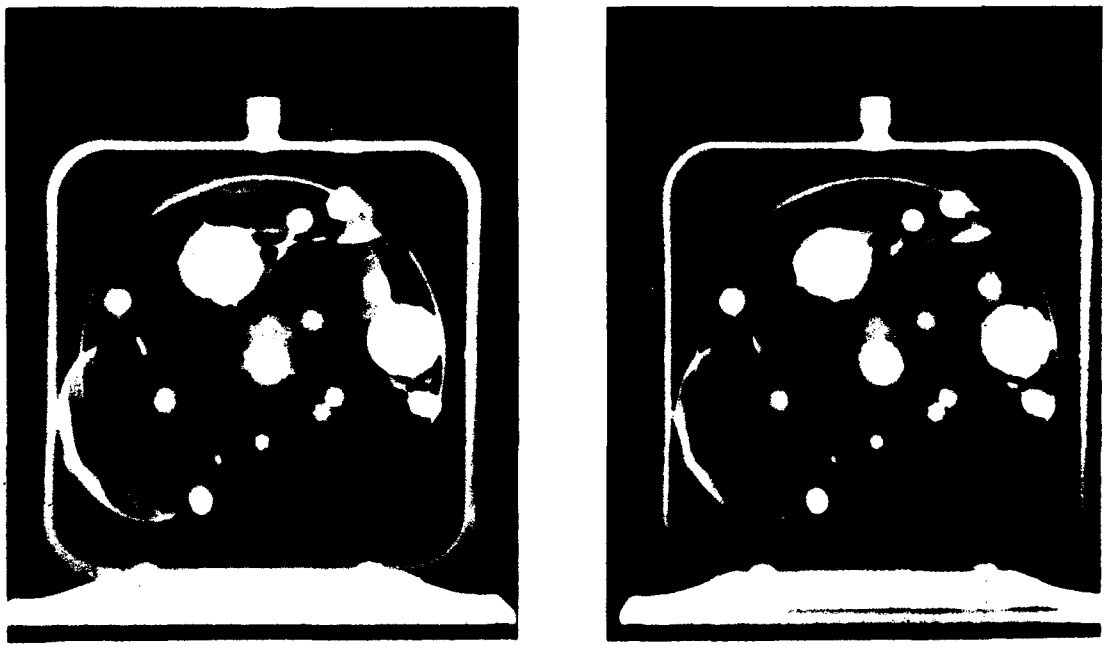


Fig. 5.7—Comparative radiographs of a dark-room timer produced with Co^{60} and Cs^{137} sources.[10]

IRIDIUM-192

Iridium-192 emits gamma radiations with energies ranging from 100 to 700 kev. The average is about 350 kev. This radioisotope is available at about $10 per curie when purchased in large quantities. It can be obtained in a very high specific activity—~300 curies/g. Its intermediate energy range makes it serviceable for radiographing a wide range of thicknesses of different metals.

Iridium radiography machines have all the advantages of other isotope radiography; low cost, safety, low weights of about 60-135 lb. Versatility of the iridium radiography units make them attractive when compared to X-ray machines. A 50-curie iridium radiography unit re-

quires a smaller exposure time than a 250-kv X-ray machine for radiographing the same thickness of steel. Figure 5.8 gives the decay curve for Ir^{192}. Figure 5.9 shows a commercial iridium radiography unit known as the Iriditron, manufactured by the Budd Company, Philadelphia. When the aluminum source cap is replaced by a source tube and the short cable by a long one, the machine can be used for panoramic exposures, in which the source is kept at the center of a large number of parts to be radiographed. This attachment is also used within an object, around which the photographic film is wrapped. X-ray machines cannot usually be employed in such a versatile manner.

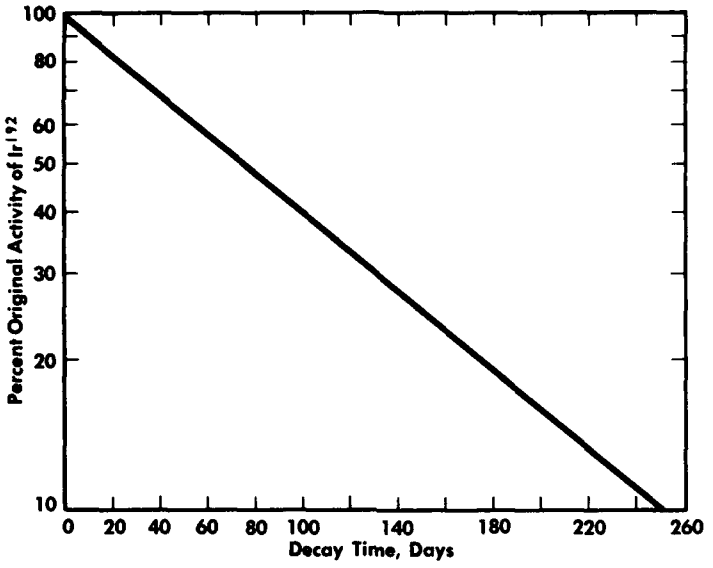


Fig. 5.8—The decay curve for Ir^{192} (semilog scale).

CERIUM-144

Cerium-144 is one of the fission products of uranium having a heterogeneous gamma-ray spectrum.[11] It emits hard gamma photons of 2.19 and 1.48 Mev as well as weaker gammas of about 134 kev. The total gamma-ray output is due to 5 per cent of the disintegrations; the other 95 per cent of the disintegrations are beta-particle emissions. The half-life of Ce^{144} is about 280 days. The heterogeneous gamma-ray spectrum makes it useful for simultaneously investigating thick as well as thin sections of metals. Limited studies carried out with sources of about 50 mc indicate that for radiographing 12 in. of steel sections, about 10 curies are required, and for 15 in. or more of steel, about 100 curies of Ce^{144} would be required. Although great amounts of this radioisotope exist in the fission products, separated sources of Ce^{144} of large strength are not yet available.

THULIUM-170

Thulium-170 emits gamma radiation of 84-kev energy.[6] This source has been used with X-ray films in medical diagnosis. The early studies were carried out by Mayneord[12] and Spangenberg.[13]

BETA-PARTICLE EXCITED X-RAY SOURCES

Besides gamma emitters, isotopes emitting beta particles may be used as radiography sources by employing the X radiation emitted from targets bombarded by beta particles. The pure beta-particle emitter Sr^{90} produces 2.2-Mev beta particles and has been used to excite bremsstrahlung and X-rays from lead targets with a conversion efficiency of about 10 per cent. The efficiency of bremsstrahlung photon production is directly proportional to the square of the atomic number of the target element; it is also proportional to the energy of the beta particles.

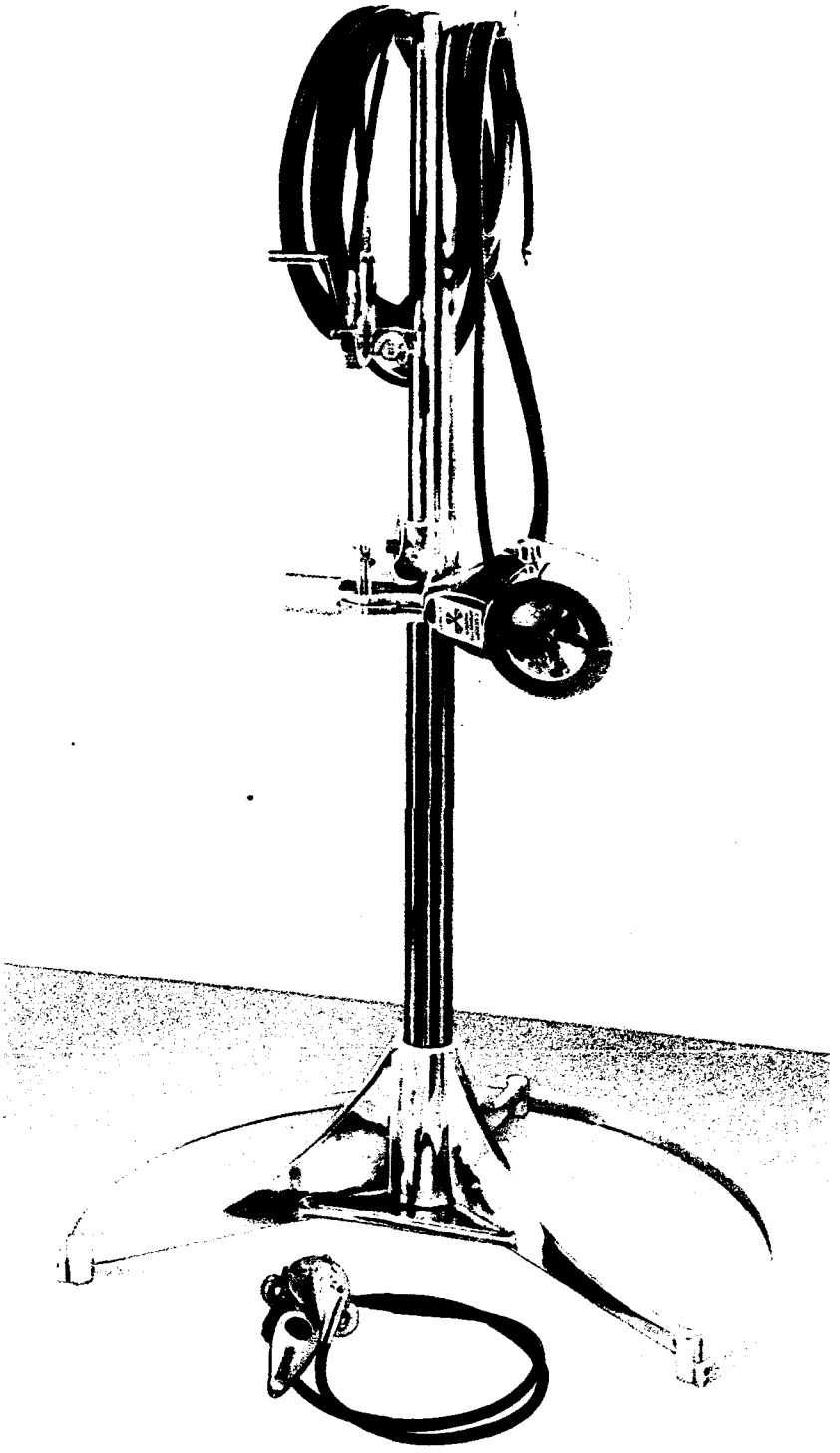


Fig. 5.9—A commercial iridium[192] radiography unit known as the Iriditron. (Courtesy of Budd Company)

Beta-particle excited X-ray sources using transmission or reflection targets have been developed at the Armour Research Foundation and reported by Reiffel and Humphreys.[7] Excellent radiographs showing grid wires (about 0.002 in. diameter) in radio tubes have been taken using 10 mc of Sr^{90} as a beta source with a lead target.

Beta-particle excited X-ray (bremsstrahlung) sources, whether of the transmission or the reflection type, have a definite size limitation. As the amount of radioactive material is increased to give increased X-ray output, more and more of the beta particles are absorbed in the source material itself. X rays produced there have a different energy from those that are produced in the target. This increase in the energy spectrum may or may not be desirable.

In case thin sections are to be radiographed, soft (low energy) X radiation is required and one energy or a narrow band of energies will produce fine detail on the radiograph. Brownell, Coleman, and Fox[102] have studied the use of source-target mixtures to produce beta-particle excited X-ray sources having the most suitable X-ray energy range for use in medical diagnosis. In the course of these studies some radiographs using a Pm^{147} tungstate source-target mixture were prepared (see Fig. 5.10) which gave very fine detail of objects of low absorption.

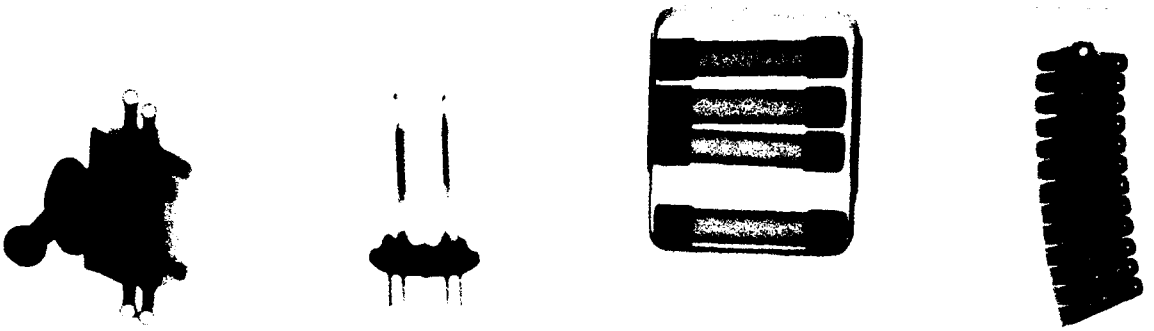


Fig. 5.10—Very fine detail of objects of low absorption by using a Pm^{147} tungstate source-target mixture.[102]

Although the spectrum of the Pm^{147} tungstate source is more satisfactory than that of other sources for certain thin-section radiography, the efficiency of X-ray production should be improved to make exposure times practical for industrial (or medical) radiography.

Figure 5.10 is a reproduction of a radiograph of various objects of different materials and densities. Unfortunately the reproduction does not show the fine detail of the original radiograph: the detail of a portion of the flexible watch strap is excellent; the mica supporting discs in the electron tube are clearly shown; and the inner construction of the switch and the fuses is shown quite well. Brownell, Coleman, and Fox also reported absorption studies of the promethium tungstate sources using aluminum and stainless steel. Figure 5.11 is the absorption curve for stainless steel using a 1-curie Pm^{147} tungstate source.

INDUSTRIAL RADIATION GAUGES

A large number of radiation gauges based upon the principles of transmission, absorption, and scattering of nuclear radiations have been designed to solve a wide range of problems in industry but have been employed primarily to measure density and thickness of materials. The operation of density gauges is based upon the absorption of nuclear radiations. Gauges have been used to determine the density of slurries, to measure and control sulfuric acid concentration over a specific gravity range of 1.50 to 1.10, to measure liquid level in high-pressure autoclaves, to detect the position of an interface (between two samples of the same liquid differing in density or samples of two different liquids), to standardize the amount of tobacco in a cigarette, and in numerous other ways.

Several thousand radiation thickness gauges are being employed in industry. Some of these gauges make use of scattering; others are based on measuring how much nuclear radiation is absorbed in one pass through the material. Product thicknesses are measured and automatically controlled by thickness gauges coupled with servomechanisms using the feed-back principle.

In the continuous casting process used for steel, molten metal is poured into a mold in a tower about 45 to 60 ft high. The solidifying metal ingot passes down through cooling zones and is guided by drawing rolls until the final product is completely solid. It would be valuable to know the depth and profile of the liquid metal inside the solidifying ingot precisely, because it

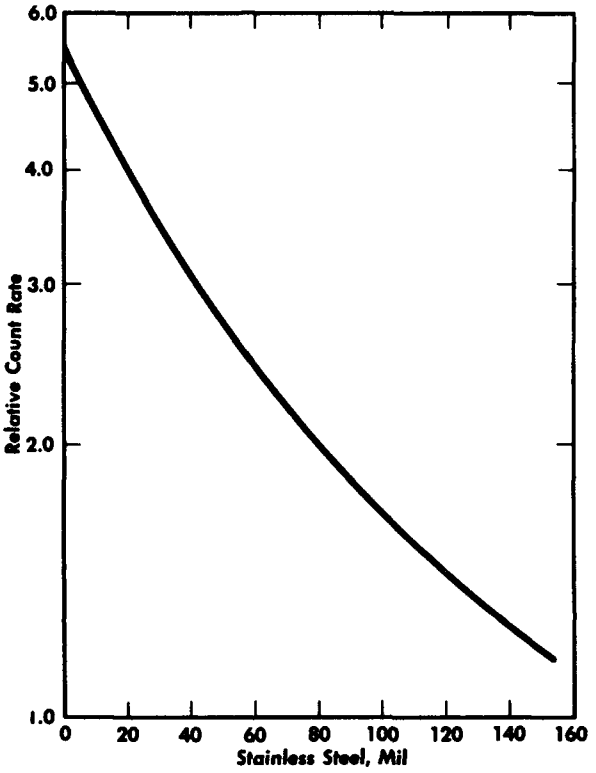


Fig. 5.11—The absorption curve for stainless steel[102] using a 1-curie Pm^{147}.

then would be possible to cut the ingot accurately as soon as it is completely solid without loss of liquid metal. In a Hungarian steel plant a method using isotopes was developed to locate the liquid-solid interface.[107]

In the Hungarian procedure a float is used which remains in the pasty transition zone. The float rises as the steel solidifies but stays just below the liquid zone. The float consists of a ball of tungsten 15-20 mm in diameter with inserts of 10 μc of Co^{60} wire sealed into the ball. For use a tungsten ball is thrown into the steel melt and a radiation counter is placed next to the surface of the ingot mold to detect the depth to which the ball settles.

ADVANTAGES OF RADIATION GAUGES

In general, the use of radiation gauges has resulted in large savings to industry. For example, a manufacturer of coated abrasives[14] reported a saving of $100,000 after installing five radiation-thickness gauges on the production line. As another example, a process used only 95 per cent of the rubber formerly used because better control eliminated the need to use extra material to take care of process variations (see Table 5.5).

Radiation gauges are also being used for routine inspection and continuous monitoring of finished products on fast-moving production lines. The measurements are rapid and reliable,

and no contact, tearing, or marking of the material under investigation is required. Additional advantages, which have made these gauges important tools in industry, follow:

1. In many applications, gauge components (source and detector) can be located outside the system under examination. The conditions of the system — high or low temperature, abrasion, corrosion, viscosity, and agglomeration — do not affect the measurements. Installation of the gauge is simple, economical, and flexible, and can be carried out without disturbing the system.

2. The radiation gauges are long-lived and rugged and require low maintenance and operational cost.

3. Turbulence and variations in flow rates do not affect the measurements, as long as density and thickness do not change.

4. Sampling tubes are not required, since the sample is measured directly in the process system.

5. Radiation gauges can be used in the food industry since they do not touch the material being measured.

6. There is no danger of contamination from a properly sealed source.

7. The gauges are extremely stable during operation, but the calibration should be checked in case of dust collection on the source and changes in temperature and humidity. However, calibration must be periodically corrected for the decay of the source. This limitation can be overcome by using two identical sources and the "null" method of detection.

Table 5.5 — COMPARISON OF PROCESS CONTROL IN VARIOUS INDUSTRIES USING RADIATION GAGES[15]

Industry	Application	Per cent of material outside specifications		
		Conventional control*	Radiation gauge, manual control	Radiation gauge, automatic control
Paper	Fourdrinier machine	± 12	± 10	± 3
Metal	Tandem cold-roll mill	± 10	± 7	± 1.5
Rubber	Sheeting calender	± 10	± 6	± 3.2
	Tire fabric calender	± 6.5	± 5.5	± 3.5
Plastics	Calender	± 7	± 3.5	± 2.5
Paper processing	Saturator dip coater	Automatic control 2.5 times better than conventional control		
Abrasive	Abrasive maker	Automatic control twice as good as conventional		

*Conventional control is normally by a sampling method.

Various radiation sources, types of radiation detectors, principles of operation, and specific applications of absorption gauges and scattering gauges are discussed briefly in the following section.

SOURCES FOR RADIATION GAUGES

Four types of nuclear radiations are being extensively used in radiation gauges — gamma, alpha-particle, beta-particle, and neutron radiations. X rays have also been used in some applications. Gamma and beta-particle radiations are obtained from radioisotopes; those used most frequently are Cs^{137} and Sr^{90}, respectively.[16] Table 5.6 lists these and other gamma- and beta-particle emitters employed in radiation gauges.[110] In general, the sources should have high specific activity and long half-life. If possible, pure beta-particle emitters should be used to avoid the problem of shielding from gamma radiation. Beta-particle sources usually consist of foils containing 5 to 20 mc of activity sandwiched and sealed between thin sheets of silver.

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Table 5.6—RADIOACTIVE SOURCES USED IN GAUGING[110]

Source material and symbol	Type of radiation	Half-life	Form of activity and quantity	Measured material thickness, mg/cm^2	Gauge application
Carbon-14 C^{14}	Beta	5500y	Various; 1 mc	0–5	Early pliofilm gauges
Promethium-147 Pr^{147}	Beta	2.6y	Powder; 5–50 mc	0–15	Thin films and tissues
Krypton-85 Kr^{85}	Beta	10.3y	Gas; 350 mc	0–70	Light paper and thin plastics
Thallium-204 Tl^{204}	Beta	4y	Crystalline; 100 mc	0–75	Same as Kr^{85}
Cerium-144 Ce^{144}	Beta	290d	Crystalline; 10–50 mc	0–200	Short half-life limits use
Cesium-137 Cs^{137}	Beta	30y	Crystalline; 10–50 mc	0–130	Papers and foils
Radium-D and Radium-E Pb^{210} and Bi^{210}	Beta	22y	Powder; 1–10 mc	0–250	Paper, plastic and light foil
Strontium-90 Sr^{90}	Beta	25y	Crystalline; 20–200 mc	0–600	Heavy paper, boxboard, thin metal, rubber
Thulium-170 Tm^{170}	Gamma	127d	Powder; 1–100 mc	100–150	Metal foils and strip
Ruthenium-106 Ru^{106}	Beta	1y	Crystalline; 10 mc	200–1200	Rubber, plastic laminates, heavy web material
Strontium-90 Sr^{90}	Bremsstrahlung	25y	Crystalline; 0.25–1 curie	500–1000	Heavy metals, steel, copper, aluminum
Iridium-192 Ir^{192}	Gamma	74.4d	Metal; 50–500 mc	500–1000	Limited by short half-life
Cesium-137 Cs^{137}	Gamma	30y	Crystalline; 50–500 mc	1000–20,000	Level and density gauging of liquids
Cobalt-60 Co^{60}	Gamma	5.27y	Wire of pellets; 1 mc–1 curie	2000–200,000	Dense materials 1-in. steel
Radium Ra^{226}	Gamma	1620y	Crystalline; 1–50 mc	2000–200,000	High-density materials

Neutron sources utilize the α,n reaction. Alpha particles are obtained from polonium, radium, or plutonium. In the case of radium-beryllium, extreme care must be taken to seal the neutron source to prevent escape of radon, a radioactive daughter-product of radium decay. Since radium emits gamma radiations as well as alpha particles, shielding is required. Polonium-beryllium or plutonium-beryllium sources are preferred because they produce no gamma radiations. Sources containing 5 mc of polonium have been found satisfactory in most applications.

Beta-particle excited X-ray sources mentioned in the section on industrial radiography have also been used in some thickness gauges.

RADIATION DETECTORS

Process measurement and control requires detecting instruments with rapid response. Photographic films, though inexpensive and simple to use, have been found impractical because of the time lag between a measurement and its evaluation. Detecting instruments should be simple and rugged in construction and cover a wide range of radiation levels. Electronic instruments used with nuclear radiations satisfy many of these requirements, but must be selected on the basis of low "noise" level, and, once calibrated, must be stable.

Usually ionization chambers are preferred for detecting beta radiation because of their lower cost; the more sensitive scintillation counters are better suited for gamma and X radiation. For neutron detection, boron fluoride proportional counters or foils and scintillation counters are usually employed. Some density gauges operate with radioactive cells (such as the contact potential difference type discussed later in this chapter) for the detection, in preference to ionization chambers, since they do not require the high-voltage power supply needed for ionization chambers. Two metallic electrodes, separated by gas, supply the driving force for the movement of the ions constituting the electric current. The radioactive cell has a larger signal-to-noise ratio than does the ionization chamber. Also, the cells do not need guard rings employed in the ionization chambers to reduce leakage resistance errors.

ABSORPTION GAUGES

Absorption gauges have been constructed for use with either gamma, beta-particle, or alpha-particle radiations. Absorption of gamma radiation in the material being gauged is exponential and involves photoelectric, Compton, or pair-production processes. Beta-particle radiation absorption and loss of energy occur as a result of interactions with electrons of the material being gauged.

Gamma Absorption Gauges

The intensity of gamma radiation traversing a homogeneous material of thickness, t, is attenuated exponentially according to the equation

$$I = I_0 \, e^{-\mu t} \tag{5.3}$$

where μ = linear absorption coefficient.

Since the absorption coefficient, μ, is not constant for all materials for a particular energy of radiation, the gauge must be recalibrated whenever a new absorbing material is used.

A simple gamma radiation gauge with Co^{60} as the source was designed to measure the depth of snow.[17] In the gauge described by Doremus 40 *mc of* Co^{60} are placed underground in a lead tube used for collimating the radiation.[17] Gamma radiation passing through the snow is detected by a Geiger-Müller (G-M) tube. The output pulses from the G-M tube are transmitted to the recording station. After calibration, this gauge measures directly and with great accuracy the amount of water at a remote station.

In an early application for the Hawaiian sugar industry, a gamma radiation gauge using Cs^{137} as the source was used to measure the weight of "bagasse" (crushed and pressed sugar cane) moving on a conveyor belt.[18] Cesium-137 gauges using 500 mc have also been used for continuously monitoring steel plate up to 2 in. thick.

Many gamma-radiation gauges are employed in industry to measure and control levels of liquids or density of slurries. Figure 5.12 shows a radioactive gauge installation for measuring density of slurries in an industrial processing plant.[19] The gauge is mounted on an 8-in. pipe carrying a slurry. The source holder and the measuring cell are mounted outside the pipe on opposite sides. Output pulses are amplified and recorded on a chart.

Figure 5.13 shows the diagram of the gauge installation shown in Fig. 5.12. The measuring cell is of the type that converts radiation directly into electrical energy. Another "atomic battery" is used in the compensating cell to produce a small but highly stable current which almost balances that of the measuring cell. This arrangement gives greater sensitivity since the difference between the two currents is fed to the amplifier and control system. The compensating cell contains a small amount of a radioisotope on the end of an adjusting screw. By turning the screw the position of the source may be altered so as to adjust the current corresponding to a predetermined gravity specification.[120]

There are many ways of using gamma-radiation gauges to measure and control liquid levels in tanks and process equipment. Radiation gauges have the advantage of continuous recording, freedom from corrosion or temperature effects of the fluids being measured, measuring on a closed system, and adaptability to automatic control.

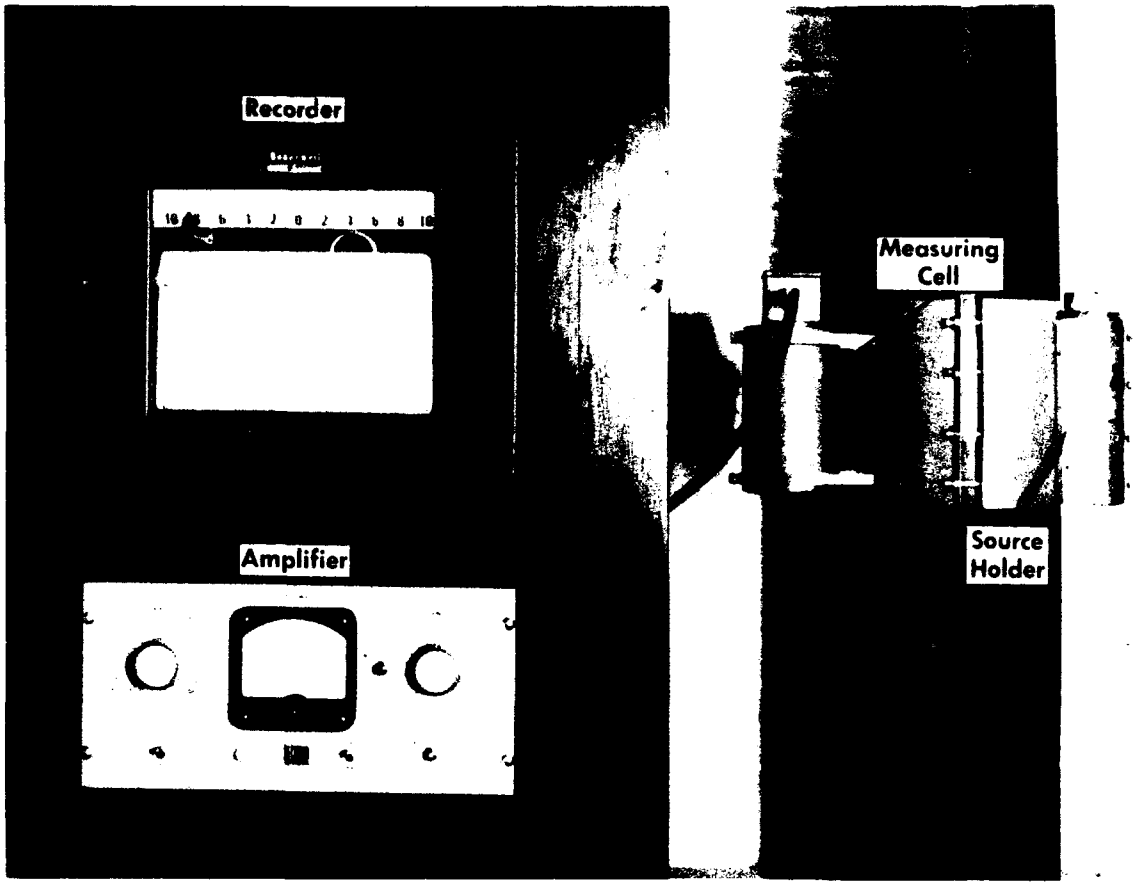


Fig. 5.12—Gamma radiation gauge installation for measuring density of slurries in an industrial processing plant. (Courtesy of Ohmart Corp.)[19]

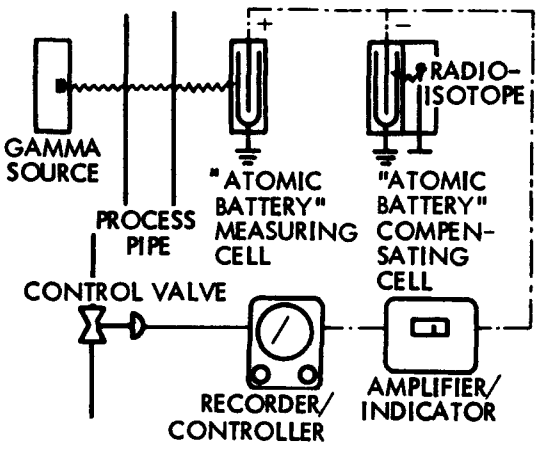


Fig. 5.13—The diagram of the gauge installation shown in Fig. 5.12. (Courtesy of Ohmart Corp.)[120]

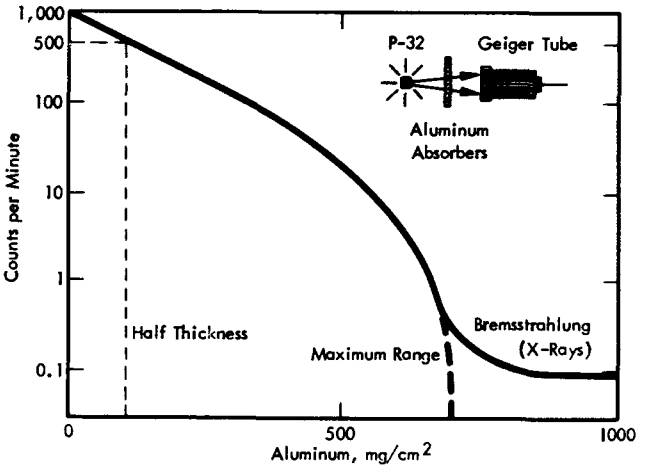


Fig. 5.14—Variation of detector response with thickness of aluminum sheet for P^{32} beta particles.

A specific example of an early application of a gamma-radiation liquid-level gauge at the Ford Motor Company is for measuring the height of different materials in a cupola.[14] A gamma source was placed outside the cupola, and a Geiger counter placed 180° away on the opposite side of the cupola. The source and sensitivity of the counter were adjusted to determine levels of liquid metal, slag, scrap, and coke. In another type of system to measure liquid level, a radioactive source is placed in a float whose height can be determined by a radiation detector.

Another example of a gamma gauge application was the inspection of a high-pressure steam line.[14] In this case a 4-in. high-pressure steam line from a power house had ruptured. The question arose as to whether corrosion was so great that the entire steam plant should shut down for replacement of corroded steam lines. However, inspection of the lines with a gamma-radiation gauge located the hazardous portions of the system and they were quickly replaced without major shutdown. This inspection located one badly eroded section that was highly dangerous.

X-ray (radioisotope) Gauges

In addition to radiation from gamma emitters, X radiation produced by K-capture (capture of a K shell electron in the nucleus of atoms of a target material) or bremsstrahlung from beta-particle emitters has been used in radiation gauges.

One such application of X radiation from K-capture is that developed to analyze petroleum products for sulfur.[15,20,21] This takes advantage of the resonance absorption by sulfur of X radiation from manganese — sulfur selectively absorbs this radiation 20 times more than carbon and 450 times more than hydrogen. Five millicuries of Fe^{55} are placed in a source holder and the sample to be analyzed is between this source and a G-M tube connected to a scaler and timer. After calibrating the equipment, a sulfur analysis that is accurate to ±0.05 per cent sulfur can be made in about 5 min. This saves about ½ hr per analysis in comparison with older methods. The analysis can be performed by nontechnical operators.

X rays excited by beta-particle radiation sources have also been used in thickness and density measurement systems.[7] A system for measuring thickness and density with a secondary X-ray source uses a highly efficient radiation detector, usually either a scintillation or a proportional counter.

Bremsstrahlung radiations, from external targets with Sr^{90} beta-particle sources, are employed in industrial gauges for measuring steel thicknesses. There is also a bremsstrahlung gauge in the shape of a U for measuring silt density in lakes, rivers, and harbors. When silt builds up excessively, it has to be dredged. Determining the depth of silt and its density is important in planning dredging operations. The silt gauge, which can be less than a foot wide, is lowered from a boat and moved through the silt at a selected depth, making a continuous record of silt density. This gauge uses 20 mc of Sr^{90} source and a lead target to produce bremsstrahlung radiation. It is accurate, does not require the collection of samples, and gives the desired measurements rapidly.

Beta-Particle Absorption Gauges

Beta-particle gauges are extremely versatile; they serve in many industries to measure mass, density, or thickness. Products for which these gauges are suited include many items such as: sheet metals, foils, floor tile, paper, plastic sheets, boxboard, tire fabric, artificial leather, ceramics, ink films, cigarettes, and adhesive tapes.

Beta-particle gauges may operate either by absorption or by reflection (back scatter). The basic elements required for a beta-particle absorption gauge are radioactive source, detector, voltage supply, and a meter or recorder. The material to be gauged is placed between source and detector. Absorption of radiation can determine thickness or density of the material. (See Fig. 5.14, which shows variation of detector response with thickness of aluminum sheet.) In a gauge the detector signal is shown on a calibrated meter or recorder, or operates a controller through relay circuits. An arrangement of beta-particle radiation gauges for measuring the thickness of a continuous sheet is shown in Fig. 5.15.

Gauging continuous sheet is one of many industrial problems now being solved by radiation gauges. Earlier methods were very unsatisfactory, involving the use of contacting gauges that were both inaccurate and costly to maintain. The automotive industry is now using beta-absorption gauges to measure the thickness of sheet steel during rolling.[14] The gauges are ruggedized versions of those shown in Fig. 5.15 and consist of a high-intensity, high-energy

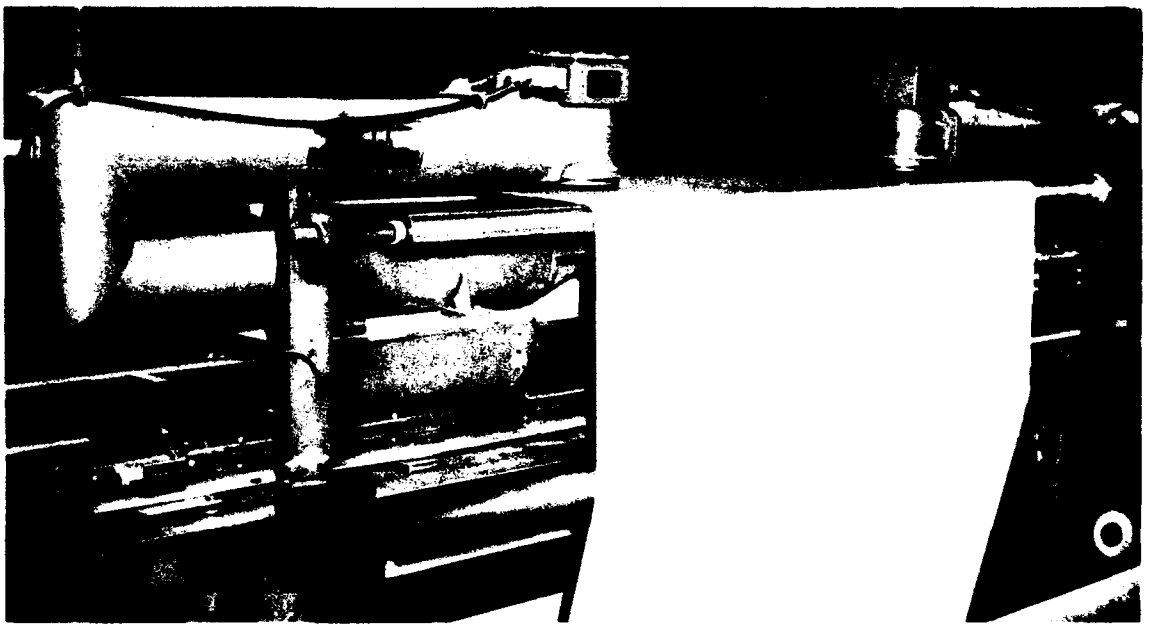


Fig. 5.15—An arrangement of beta-particle radiation gauges for measuring the thickness of a continuous sheet. (Courtesy of Tracerlab, Inc.)

Table 5.7—APPROXIMATE RANGE OF THICKNESS MEASURED BY Sr^{90} BETA-PARTICLE GAUGES[15]

Material	Density, g/cm^3	Thickness, in.
Aluminum	2.7	0.0002–0.075
Asbestos	2.5	0.002–0.075
Copper	8.9	0.0006–0.020
Glass	2.7	0.002–0.075
Paper	1.0	0.005–0.200
Plastics	1.4	0.004–0.145
Rubber	1.0	0.005–0.200
Steel	7.8	0.0006–0.120
Textiles		5 to 150 oz/sq yd

beta-particle source on one side of the sheet steel and a radiation detector on the opposite side. The attenuated radiation produces a signal shown on a strip-chart recorder; this signal governs a control mechanism for the rolls of a three-stage tandem cold mill in the steel division. Thickness is controlled within ±0.001 in. on 0.035-in. sheet; previous gauging techniques showed four times this tolerance.[14]

Beta-particle absorption gauges are also used for paper, plastics, and nonferrous metals. Such gauges more often contain Sr^{90} than other beta-particle sources, because Sr^{90} emits high-energy beta particles, has a long half-life, and is in ready supply. Table 5.7 gives the approximate range of thicknesses that can be measured by Sr^{90} gauges.[15]

Absorption of beta-particle radiation is primarily due to scattering as a result of interaction with the orbital electrons of the absorbing element. The difference between beta-particle

attenuation by hydrogen and by carbon atoms has led to design of the density gauge for measuring hydrogen-carbon ratio (H/C ratio) in hydrocarbons.[15] This ratio is determined by making two measurements, one of beta-particle absorption, the other of specific gravity.

One design (see Fig. 5.16) of a commercial hydrogen-carbon ratio gauge using a beta-particle source, requires no correction for decay. The equipment is operated by balancing currents from the two ionization chambers, by adjusting an absorption wedge, and by using a source common to both ion chambers.[15] If the chambers are identical, the currents will be balanced regardless of source activity, providing the same amount of absorption occurs before the radiation enters both chambers. With this procedure, measuring the thickness and specific gravity of a test sample, the hydrogen content can be determined in about 15 min within an accuracy of about 0.05 wt.%.

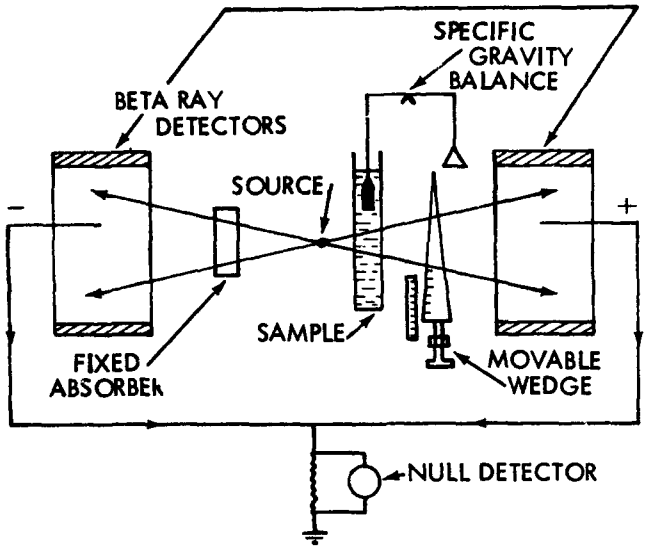


Fig. 5.16—Hydrogen-carbon ratio gauge using a beta-particle source.[15]

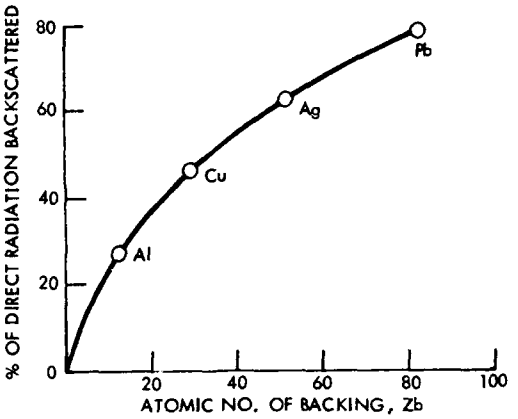


Fig. 5.17—The percentage of beta particles backscattered increases with the atomic number of the absorber having "infinite thickness."

Low-energy beta-particle emitters such as C^{14} may be used to gauge very thin films of material with low atomic number. Plastic films (cellophane, polyethylene, saran, and polymylar), as well as paper and photographic film, are examples of such products. The average deviation was reduced from 0.015 lb/sq yd (without control) to one-third this value with radiation thickness gauges.

An application of beta-particle measuring that has received considerable publicity is the gauging of cigarettes. The amount of tobacco in a cigarette affects both the smoking quality and the cost of the cigarette. In manufacturing cigarettes, carded tobacco is compressed on a narrow belt, transferred to the cigarette paper, and a continuous cylinder emerges to be cut into lengths. A beta-particle gauge is used to check the firmness of the continuous cylinder emerging from the cigarette machine. The tobacco industry employs approximately 3000 such gauges in the United States.

Two Sr^{90} sources are used rather than one, but both have the same decay rate and give the same result as would a common source. The difference in readings of the response of two ion chambers, one reading radiation passing through the standard cigarette and the other reading the radiation passing through the continuous cigarette cylinder, shows whether the cylinder is lighter or heavier than the standard. The desired density of 200 mg/cm^3 is controlled within a maximum variation of 1 mg/cm^3, by automatically adjusting the tobacco feed at a rate proportional to the amount and duration of the error. After the adjustment is made in the feed rate some time is allowed before a new correction is accepted to prevent "hunting" of the control system. The radiation system is much more stable and sensitive than the previous gauges.

Some industries require multiple gauges for process control. In one application of this type a series of five (Sr^{90} beta-particle) absorption gauges are employed on a machine making abrasive paper.[15]

SCATTERING OR REFLECTION GAUGES

Different gauges are used to scatter or reflect different types of radiation. The various gauges employed are discussed in the following paragraphs.

Beta-Particle Scattering

Scattering gauges are employed where only one side of the material is accessible. The backscattering of beta-particle radiation has been extremely useful in measuring the thickness of paint or other coatings on base material, such as tin plate on steel.[21]

The interaction of beta particles with the inner orbital electrons of the atoms of an absorber results in recoil or "backscatter" of some of the particles. If the absorber is a thin film most of the beta particles may pass through the film (see Fig. 5.14). As the thickness of the absorber is increased the quantity of recoil electrons increases until a "saturation" value is reached. Beyond this value an additional increase in absorber thickness produces no additional recoil particles outside the absorber because of absorption. The critical thickness is sometimes termed the "infinite thickness" because it produces the same backscatter as an absorber of infinite thickness. It is approximately equal to half the maximum range of beta particles of a given energy in the absorber (see Fig. 5.14). The percentage of electrons backscattered also increases with the atomic number of the absorber as shown in Fig. 5.17.

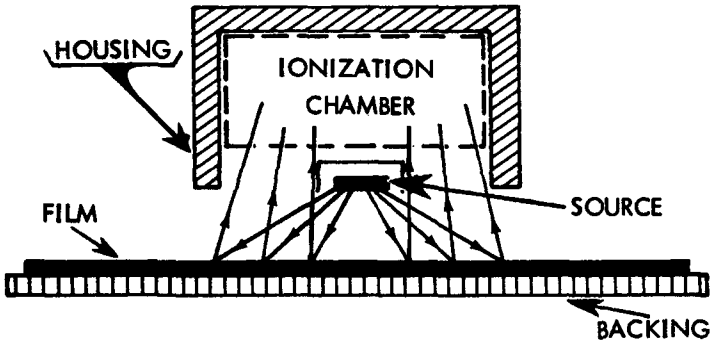


Fig. 5.18—Beta-particle backscatter gauge for measuring thickness of films.

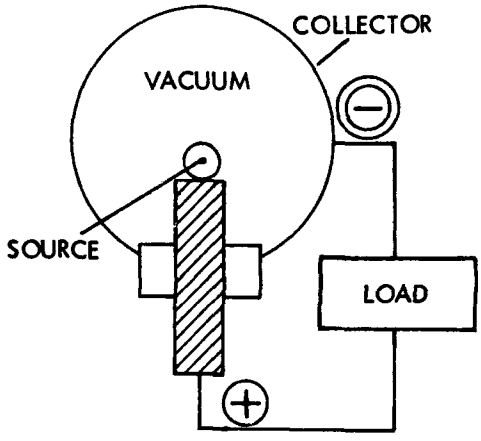


Fig. 5.19—Direct charging type of atomic battery based on the collection of charged particles on a metallic electrode.[121]

This difference in the backscatter produced by materials of different atomic number can be used to measure film thicknesses less than the "infinite thickness" of the beta particles in the film material. To calibrate the gauge the percentage backscatter for an "infinite thickness" of the backing material (zero film thickness) is measured and the gauge adjusted to zero. The change in gauge reading as different thicknesses of film are inserted between the gauge and the backing can be used for the calibration.

The source of radiation and the detector are mounted in the same container, as shown in Fig. 5.18, but the detector is shielded from the direct rays of the source. Commonly used sources are Sr^{90}, Co^{60}, Cs^{137}, Ru^{106}, and S^{35}. The backscattering gauge continuously measures the thickness of the zinc coating in the galvanizing process.[22]

The sensitivity of this type of gauge depends on the quantity

$$\frac{Z_b^n}{Z_c} - 1 \qquad (5.4)$$

Z_b refers to the atomic number of the base element and Z_c to that of the coating. According to this expression, a higher sensitivity of detection is possible when the difference between Z_b and Z_c is large.

Gamma Scattering

Gamma scattering gauges, based upon differential Compton scattering and the differential photoelectric absorption for materials of different atomic numbers have been designed. Gamma rays from Tm^{170} (85 kev) have been applied experimentally in sorting of coal and shale.[23] It was found that the pure coal (of mean atomic number of 6) scattered back more gamma radiation than the shale (of mean atomic number of 13). A gauge based upon the above experimental finding is being developed to sort coal automatically.

Neutron Scattering

Neutron scattering gauges have many important specific applications, one of which is to determine the moisture content of soil. In soil, a fast neutron is slowed or moderated primarily by a series of elastic collisions. Hydrogen is effective in slowing fast neutrons since it has a large cross section for elastic scattering. By collision with a hydrogen atom, a neutron may lose most of its kinetic energy. The amount of moderation caused by other elements in the soil is small; in normal soils most of the hydrogen present is in the moisture. The detector counts slow neutrons, and a boron fluoride proportional counter or scintillation counter probe is often used. Another alternative is a G-M tube counter wrapped with silver foil. The silver is made radioactive by neutron capture and its radioactivity is detected by the G-M tube. Such instruments can measure moisture content with an accuracy of ±5 per cent within a period of 6 min.[24] The gauge can also determine moisture in grain, timber, and stones. By using a gamma-scattering gauge with the neutron-scattering gauge density may also be determined.

In determining moisture content and density in a depth of soil, grain, or other material, an access tube of about $1\frac{1}{4}$ in. in diameter permits lowering the probe into the soil or grain. The moisture probe containing the neutron source and the detector is lowered through the access tube to permit quick determination of moisture at various depths. The neutron source consists of 5 mc of polonium-beryllium. Slow neutrons reflected to the probe activate the 3-mil silver foil, producing Ag^{110} with a half-life of 24 sec. Three minutes are required to permit the Ag^{110} to come to equilibrium before reading the activity.

The density probe contains a gamma source of Co^{60} and a detector. Gamma radiation is scattered primarily by the Compton effect. If the density of the medium is first increased, the probability of scattering by single collisions increases and raises the count rate of the meter. However, additional density places more absorber atoms between the detector and the point of interaction, which increases the absorption of secondary radiation returning to the detector. Therefore, as the density is increased over a wide range, the counter response first speeds up sharply, then decreases slowly.

Emission of prompt gammas from neutron capture has also been utilized in studying the composition of underground strata during the logging of bore holes. An instrument was designed at Harwell, England, to obtain information from neutron-capture nuclear reactions.[25] The neutron source (polonium-beryllium) and the scintillation detector are lowered in a long tube into the bore hole. The neutrons are absorbed by the surrounding medium and the prompt gammas characteristic of the elements present in the medium are emitted. These are detected by a counter with a linear response, and the output pulses are fed into an electronic pulse height analyzer or "kick sorter" to record the spectrum of emitted gammas. From examination of the various peaks, a qualitative estimate of the nature and relative frequency of the elements capturing the neutrons can be made. By this procedure it has been possible to distinguish between crude oil and brine in sandy strata.

ALPHA-PARTICLE GAUGES

Alpha particles have very limited penetration and therefore alpha-particle emitters have not been used extensively for gauging. However, an alpha-particle transmission gauge has been designed to measure very small thicknesses (up to 2 mg/cm^2). Alpha-particle pressure gauges have been developed[16] to measure gas pressures from 1 to 500 μ. The alpha particles in the gauge ionize gas molecules remaining in the partially evacuated system, and the ions are collected on electrodes as in an ionization chamber. The ionization current is amplified and meas-

ured to indicate the concentration of gas molecules and therefore the pressure in the chamber. Gauges of this type have been used for vacuum furnaces, vacuum metallizing equipment, cathodic etching, and laboratory applications.[14]

The automotive industry has used alpha-particle gauges to measure accurately the air flow through the carburetor of an automobile during a road test.[14] Foils containing radium were placed on plates and the plates installed in the air line of the engine. Air entering the engine was ionized by alpha particles from the radium source, the ions were collected, and the ionization current determined to give the air flow (which is proportional to ionization current). The gauge has also been used on air-conditioning equipment and on experimental brake-cooling studies.[14]

LUMINESCENT COMPOUNDS, PAINTS, AND LIGHT SOURCES

Radiation is used extensively in the production of luminescent compounds, paints, and light sources. The following paragraphs discuss the use of alpha and beta particles for this purpose, how the compounds and paints are applied, and the problems associated with their production.

MECHANISM OF ALPHA-PARTICLE ACTIVATION

Since the discovery of radium, one of its industrial applications has been in the production of luminescent compounds and paints. For this purpose radium bromides and sulfates are used to excite the phosphors of zinc sulfide. Besides radium, other alpha-particle emitters such as mesothorium and polonium, and other phosphors such as sulfates of zinc and calcium, tungstates of calcium and cadmium, and sulfides of strontium and cadmium have been employed. The zinc sulfide phosphor shows the maximum efficiency for converting energy.

The effect of alpha particles in exciting phosphors appears to be a mechanical effect similar to "triboluminescence." Each alpha particle causes an individual flash of light which can be readily observed under magnification. Millions of flashes produce a uniformly luminous surface.

The maximum brightness obtained from alpha-particle activated phosphors is about 100 microlamberts. This light is in the range of 5200 to 5700 A for green or yellow zinc phosphors. Deterioration of phosphors by alpha-particle bombardment reduces the maximum brightness by a factor of $\frac{1}{2}$ during a period of 6 months.

MECHANISM OF BETA-PARTICLE ACTIVATION

Beta-particle emitters are also used to activate large quantities of phosphors. The effect of beta-particle radiation on the phosphor is both electrical and electromagnetic—the mechanism is not the same as that of alpha-particle radiation. Even with a high-resolution microscope, no flashes are observed for beta-particle activation of the phosphor. The problem of phosphor deterioration is eliminated when beta-particle radiation is used. It has been found that the maximum brightness of the light output of phosphors activated by beta radiation is about 1000 microlamberts in the green or yellow range and 100 microlamberts in other ranges. No decrease in brightness of beta-particle-activated phosphors has been reported over a period of 5 years. Efficiency of light production may be improved by increasing the beta-particle energy of the activator. Some activators react with the phosphors and change their responsive properties, discoloring the crystals; this effect is most pronounced with the isotopes of heavy metals.

The ratio of light production efficiency for alpha-particle compared to beta-particle radiation varies with the type of beta-particle-emitting radioisotope. Table 5.8 lists a number of radioisotopes, their beta-particle energies, and the ratio of the efficiency of light production for alpha particles compared to beta-particle radiation. Efficiency also varies with the method of processing luminous compounds.[26]

When self-luminous compounds are used, the beta-particle activators should be properly shielded for both the primary beta-particle radiation and the secondary bremsstrahlung radiation. Glass and plastic shields have been employed; however, decomposition of plastics by beta-particle radiation is reported to be a serious problem.

ADVANTAGES OF BETA-PARTICLE EMITTERS.

There are numerous advantages in using beta-particle activators rather than alpha-gamma activators:

1. Increased useful life of the phosphors.
2. Higher light intensity.
3. Availability of beta-particle emitters of suitable half-life and energy.
4. Freedom from gamma radiation minimizes the problem of shielding.
5. Greater range of colors at practical brightness levels.
6. Absence of the radioactive gas deposits encountered in some of the alpha-particle activators.
7. Relatively lower cost of the activators.

Table 5.8—LIGHT PRODUCTION EFFICIENCY RATIOS FOR VARIOUS BETA-PARTICLE SOURCES[26]

Radioisotope	Energy, Mev	Light production efficiency ratio, alpha/beta
H^3	0.0189	1:5000 to 1:7000
Pm^{147}	0.223	1:40
Tl^{204}	0.78	1:12
Sr^{90}	0.54	1:14
Y^{90}	2.18	

TYPICAL USES

Many applications of beta-particle activated luminous compounds have been reported. These include safety markings in commercial aircraft, mines, and photographic dark rooms, and other places where electrical failure may occur or where there are no other sources of illumination. Some examples are signalling equipment for railroads, life rafts, and markers on channel-indicating buoys.

Krypton-85 has been used as the beta emitter for activating such light sources. Selection of Kr^{85} as an exciter provides three important operating features:

1. Kr^{85} has a half-life of ten years. Luminous intensity varies directly with the activity of the exciting source. Thus a lamp activated by Kr^{85} requires ten years for its brightness to decrease by one-half.
2. Kr^{85} is a chemically inert gas and has a relatively soft emission with little penetrating radiation. There is little danger of radioactive contamination if the lamp were damaged because the gaseous krypton would be rapidly dissipated by the surrounding air.
3. The lamp is explosion proof because it generates essentially no heat and utilizes no electricity.

Such lamps as produced by one company (United States Radium Corp.) consist of a hermitically sealed light capsule and a housing. The housing protects the light capsule from breakage and also serves as shielding. The light can be provided in two forms: (1) as a concentrated, collimated beam providing a narrow intense beam for long distance viewing, or (2) as a bright diffuse surface which is visible for wider angles.

The lamps cannot, however, compete with daylight and are not visible during the day. They function best as a directly viewed light source in a dark area or in an area whose ambient light level is no greater than those encountered at dusk. Such a lamp designed as a signal lamp for railroads can be seen for 500 yd at night.

Self-luminous compounds are also used in calibrating photometric equipment, as comparison sources in light meters and in activating photocells.

Another application is in low-level illumination of instrument dials and panels. These sources are also employed to illuminate cross hairs in optical equipment and to identify by color equipment operated in darkness.

PROBLEMS REQUIRING ADDITIONAL STUDY

Further research must be undertaken if the full potentialities of radiation activation of phosphors are to be realized. The problems requiring such study are the mechanism of beta-particle activation of phosphors, the ultraviolet and infrared emitting compounds, improvements in transparent shielding materials, and determination of variables affecting efficiency.

STATIC ELIMINATION

Normally air is one of the best electric insulators. Actual conductivity of gases is so small that it may be measured only by extremely sensitive apparatus. However, a charged gold-leaf electroscope gradually loses its charge at a rate slightly higher than is explained by leakage through the insulating material. Years ago this was thought to be due to minute leakage currents through the air of the order of 10^{-18} amp/cm^3 of air present. To check this possibility an experiment[27] was performed in which the original potential was maintained on the outside of the insulator. In this way, any leakage occurring across the insulator would tend to keep the electroscope charged. The electroscope still discharged, indicating that the charge was lost through the conductivity of the gas.

The phenomenon can be explained by the presence of ionizing radiation, which accelerates discharging an electroscope by current leakage through air. The rate of discharge of a well-insulated electroscope, remote from any source of ionizing radiation, is a direct measure of ionizing radiation present due to cosmic rays and to local residual radioactivity.

Lord Kelvin[28] ionized air with uranium, stating that "two polished metallic surfaces connected to the sheath and the insulated electrode of an electrometer, when the air between them is influenced by the uranium rays, give a deflection from the metallic zero, the same in direction, and of about the same amount as when the two metals are connected by a drop of water."

With the use of X rays, radioactive materials, and other sources of ionizing radiation, it became industrially possible to dissipate an undesirable accumulation of electric charge by ionizing the surrounding air.[29]

Moving dielectrics in industrial plants, such as leather belt conveyors, tend to accumulate static charge until sparking potentials are reached; this can be dangerous if an explosion or fire hazard exists in the plant.

ALPHA-RAY STATIC ELIMINATORS

The dangers of many explosions and fires in plants can be eliminated by placing an alpha-particle source near the charge collector to reduce the conductivity of the air between it and a "ground," permitting excess collected charge to dissipate through the air.

Static eliminators employing the intense ionizing power of alpha-particle radiation have been proved feasible and dependable by years of industrial usage. However, such sources require the utmost care in preparation and use, because of the extreme health hazard involved in the escape of alpha-particle radioactivity into the environment.[30]

For alpha particles to escape from the source with enough energy to produce ionization, the source container must have an extremely thin window. This window, although only a few milligrams per square centimeter thick, must be strong enough to withstand handling and abrasion. Also, in cases were radioactive gases such as radon tend to accumulate, the container must retain these gases.

The automotive industry uses ionized air from an air gun containing a polonium alpha-particle source, to remove dust and lint from surfaces to be painted.[14] This is a particular problem in painting plastics because their high dielectric constant results in the build-up of large static charges which attract and hold small particles on the surface. Normal air will not remove all these particles but ionized air neutralizes the charge and blows off the objectionable particles.

BETA-PARTICLE STATIC ELIMINATORS

Both beta- and alpha-particle emitters can eliminate static electric charge by ionizing the surrounding air; this is an important industrial application of radioisotopes. Because the range

of beta particles in air is substantially greater than that of alpha particles, beta-particle static eliminators can be packaged more solidly and placed farther from the operation, thus tending to reduce the radiological hazard. Also, if radioactivity accidentally enters the body, the greater range of beta particles in tissue allows the same amount of energy to be absorbed in a greater volume of body tissue and reduces the degree of local physiological damage.

One disadvantage of beta-particle static eliminators, as compared to alpha-particle static eliminators, is the tremendous reduction in ionization intensity produced per particle. An alpha particle produces about 100 times as many ion pairs per millimeter of path as does a beta particle of comparable energy. This great difference in ionizing power limits the use of beta emitters for static elimination.

Henry[31] discusses the need and describes the use of electrostatic eliminators in the textile industry. The advantages and disadvantages of alpha-particle and beta-particle emitters for use in static eliminators are compared. The use of a 2-mc source of Tl^{204} as an eliminator is described and a drawing and a calibration curve are given. Henry considers the health hazard of using radioactive static eliminators near weavers in textile mills, and also comments that such units have successfully prevented "fog-marking" on acetate rayon and nylon.[31]

Precautions against explosions that might result from the ignition of anaesthetic gas mixtures are described by Quinton.[32] Special attention is given to eliminating charges of static electricity by passing a stream of air over a radiothallium source. Designs and photographs of apparatus are described. The ionized air so produced is stated to be effective in removing electrostatic charges at distances up to 6 ft.

TRANSFORMING RADIATION ENERGY ATOMIC BATTERIES AND ELECTROSTATIC GENERATORS

Converting nuclear energy into other useful forms of energy has been a dream of nuclear scientists since the discovery of radioactivity. The first serious attempt to convert nuclear energy into electrical energy was made by Moseley[33] in 1913. He obtained an electrical potential of 150 kv from 20 mc of radon in an evacuated metallic sphere. The method was based upon collecting charged particles. Since radioactive materials were not available in large quantities before the atomic era, the experiments carried out before about 1946 were largely academic.

The beta-particle emitters of greatest interest are Sr^{90}, H^3, Pr^{147}, and Ce^{144}. The properties of 1 curie of Sr^{90} and H^3 are summarized[34] in Table 5.9. Polonium-210, an alpha-particle

Table 5.9— PROPERTIES[34] OF Sr^{90} AND H^3

Property	Sr^{90}	H^3
Amount	1 curie	1 curie
Disintegration rate per sec, dps	3.7×10^{10}	3.7×10^{10}
Current, amp	6×10^{-9}	6×10^{-9}
Average voltage, volts	5×10^{5}	6×10^{3}
Power, watts	3×10^{-3}	3.5×10^{-5}
Half-life, years	20	12.4
Weight, g	5×10^{-3} (100% pure)	

emitter, is also a suitable source of charged particles, but radiation damage is caused by the high mass of alpha particles it emits. Conversion of gamma and other uncharged radiations is also being studied. However, gamma radiation must be "soft" to be of practical value. For this reason, the use of Co^{60} is not promising, but Am^{241} may be useful when it becomes available in large quantities.

It has been estimated that by 1965 the total output of fission products in the United States may be about 6×10^6 g. Although this estimate now appears optimistic, it is interesting to note

that if combined into a single source, this would be equivalent to a source strength of 3×10^9 curies, capable of producing 6×10^6 watts of power.[35]

Investigations are proceeding on four main methods of conversion: direct charging, contact potential, thermopile, and semiconductor.

DIRECT CHARGING BATTERY

This battery is based on the collection of charged particles on a metallic electrode (see Fig. 5.19). Moseley's experiment was based upon this principle. In 1952, Christian and Linder[36] were able to obtain an electrical potential of 365 kv and a power of 0.2 mw at 20 per cent efficiency using 25 mc of Sr^{90} as a source. The current obtained was only 10^{-9} amp while the impedance was about 10^{14} ohms. Coleman[37] and Rappaport and Linder[38] eliminated the problems associated with the high vacuum technology by substituting a dielectric for the vacuum between two metallic electrodes. This design is simple and reliable. It is possible to produce several thousand volts but at low current and high impedance. The current is approximately proportional to the amount of radioactive material used. Figure 5.20 shows a sectional sketch

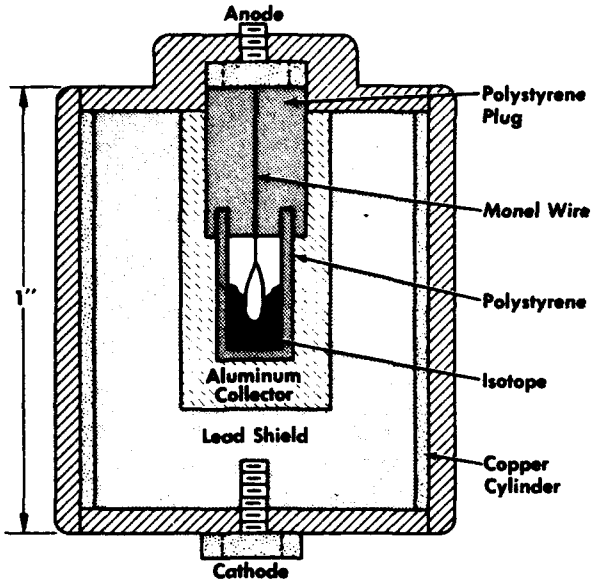


Fig. 5.20—Primary current collecting type of atomic battery with a solid dielectric of polystyrene.[39]

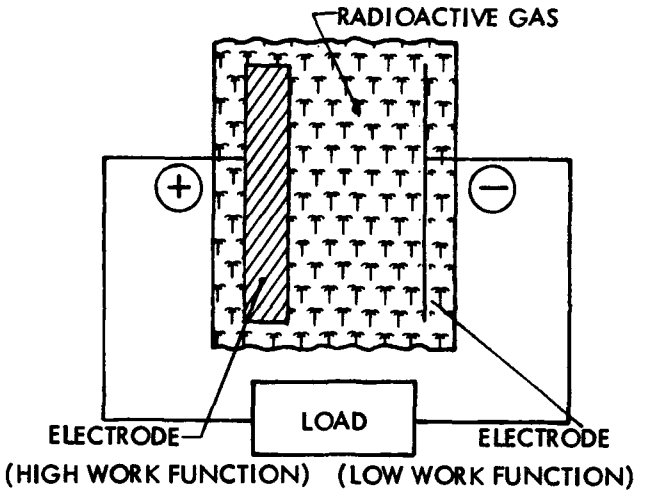


Fig. 5.21—Contact potential type of atomic battery.[121]

of a primary current collecting battery with a solid dielectric of polystyrene. The voltage, V, developed by this battery can be given as

$$V = RI_p\,(1 - e^{-t/RC}) \tag{5.5}$$

where R = insulation resistances
C = capacitance
t = time
I_p = primary current

For large $t \gg RC$ (time constant), $V = I_pR$ (5.6)

For small $t \gg RC$, $V = I_pt/C$ (5.7)

Schwarz[40] introduced a secondary emitter (MgO_2) having a high secondary emission and a collector electrode of graphite coated with aluminum to reduce the bremsstrahlung and backscattering, and to increase the total current.[39]

Both the primaries and secondaries contribute to the current. The design would be of practical value if the current could be increased and the impedance lowered, but this is possi-

ble only if a large amount of radioactive material is used. However, the submicrowatt of power available can be used in instruments such as dosimeters and radiation meters and to power transistors in midget radios and other electronic equipment.

CONTACT POTENTIAL BATTERY

This method of converting radiation to electrical energy makes use of the movement of the charged particles in the electric field between two dissimilar metallic electrodes (see Fig. 5.21). Kelvin[28] used the basic principle to measure the contact potential difference between two electrodes by ionizing the gas between them with a uranium source. Kramer,[41] Ohmart,[42] Thomas,[43] and others have extensively investigated problems connected with this type of device. A contact potential difference (CPD) battery, designed by Thomas et al., is a multicell battery with a magnesium and lead oxide electrode couple filled with a mixture of argon, hydrogen, and tritium.[43] The tritium is the radioactive source.

A device known as SNAP III (derived from the title, System for Nuclear Auxiliary Power) was developed as an auxiliary power source for space vehicles. It has a rated capacity of 5 watts for 140 days at 8 to 10 per cent efficiency and then 3 watts at 5 to 6 per cent efficiency.

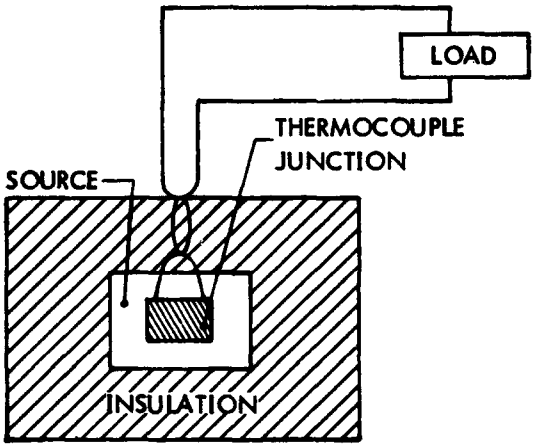


Fig. 5.22—Thermopile type of atomic battery.[121]

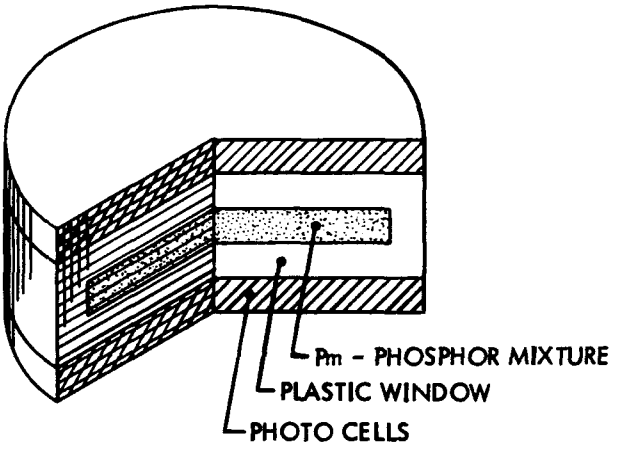


Fig. 5.23—Core of Elgin-Kidde scintillator-photocell type of atomic battery.[114]

In another research project the feasibility of using Sr^{90} is being studied as a heat source in a thermoelectric generator designed to have an unattended life expectancy of 10 years under the most extreme environmental conditions.[111] Containers are being designed for absolute containment of Sr^{90} for 500 years in both land and sea environments. The generator uses strontium titanate. This material exhibits the desirable characteristics of insolubility, high thermal conductivity, fabricability, high compound density, high strontium density, and structural stability. The compound is formed by the reaction of strontium carbonate or oxalate with TiO_2, then calcining, pressing, and sintering. The calcined powder is pressed at 5 tons/sq in. and "fired" at 1400 to 1500°C for 2 hr. Strontium titanate has a cubic crystal structure.

Cermets composed of metal powders and the strontium titanate are also being studied. Chromium appears to be the best metal for cermet use. Also, various protective claddings are being investigated and for this use Hastelloy C appears to be most satisfactory.

This battery can produce a fraction of a volt per cell and a current of 10^{-9} to 10^{-10} amp. It can convert any type of ionizing radiation, charged as well as uncharged, provided the radiation energy exceeds 30 ev, the energy required to produce one ion pair in a gas. This battery holds promise similar to that of the direct charging type; its principle use would be in instrumentation. The density gauge shown in Figs. 5.12 and 5.13 operates using two contact potential cells. Problems connected with the effect of radiation on electrode surfaces need additional investigation.

THERMOPILE BATTERY

The thermopile battery, based on the thermoelectric effect used in thermocouples (in which the difference of temperatures between two junctions causes a potential difference) can also convert nuclear energy to electrical energy. One junction is kept at a constant low temperature, while the other is heated by radiation. This method applies to both charged and uncharged radiations. The thermopile battery can be rugged, compact, and long lived (see Fig. 5.22).

Jorden and Birden[44] in 1954 devised a thermopile battery with a Po^{210} source. The power, delivered from radiation sources of 57 and 146 curies, was found to be 0.1 and 0.2 per cent, respectively, of the heat power. The maximum power output was 1.8 and 9.4 mw, respectively. By using reflectors it may be possible to increase efficiency appreciably.

More recently a battery based on the thermoelectric principle has been developed which combines a heat source and two dissimilar conductors to achieve hot and cold junctions.[111] As the heat source, the experimental battery incorporated the radioisotope Po^{210} which is an alpha emitter. If such batteries are produced in quantity less expensive Ce^{144} may be used instead. In this battery "p" and "n" type lead telluride forms the dissimilar conductors. This material was chosen because it satisfied requirements of high thermoelectric power, high electrical conductivity, low thermal conductivity, and good resistance to deterioration.

THERMIONIC CONVERTERS

Thermionic converters are similar to contact potential batteries but take added advantage of temperature differences to produce higher efficiencies than either the contact potential or the thermoelectric cells.[112] Typical efficiencies are 25 and 30 per cent for thermionic, 10 per cent for thermoelectric, and less for the contact potential cells. This performance for the thermionic converter requires a heat source operating at least at 1100°C. A thermionic converter consists of 2 electrodes in a vacuum. One electrode (the emitter) is kept at a temperature sufficiently high to cause the emission of electrons "thermionically" and the other electrode is kept at room temperature. Electrons "boiled" from the hot electrode flow to the cool one and out through an external circuit which returns it to the hot electrode. In principle, any diode will work as a thermionic converter, however, to obtain highest conversion efficiencies, a thermionic energy converter must take advantage of the difference of "contact potentials" between the two electrodes. For an electron tube, this contact potential is defined as the difference between the surface work functions of the emitter and collector.

Table 5.10—MATERIALS FOR THERMIONIC CONVERTERS[112]

Materials	Work function, volts	Temp., °C
Collectors		
BaO/SrO on Ni	1	
Cs on AgO	0.75	
Cs on WO	0.71	
Emitters		
Ba impregnated W	1.7	900–1200
Th on W	2.55	1800–2000
Cs on W	1.7	1400–1600

Table 5.10 lists some electron emitter materials that can operate in the temperature range 1000 to 2000°C. At still higher temperatures (2000 to 3000°C) refractory metals such as tantalum and tungsten may be used. Typical electrodes would have thicknesses of a few millimeters.

For satisfactory operation of a thermionic converter a reduction of the space charge is necessary to prevent limiting the current flow, because of the mutual repulsion of the electrons migrating between electrodes. The introduction of positive ions into the system will cause the formation of a plasma consisting of a mixture of positive gas ions and free negative electrons.

This gaseous plasma is a better thermoelectric medium than a vacuum because of its high electrical conductivity combined with low thermal conductivity. The simplest method of obtaining these positive ions in a thermionic converter utilizing some radioactive material would be through the ionizing effect of the radiation acting upon some gas, such as cesium vapor, injected between the electrodes.

A limitation of thermionic converters is that they are low voltage devices. A 1-kw power plant may, for example, deliver 1000 amp at 1 volt. Methods of overcoming this low d-c voltage problem are: (1) connecting thermionic cells in series, and (2) using a grid between the electrodes to introduce a modulation to the flow of electrons. This produces an a-c output, which by using transformers can be adjusted to match any external impedance.

In experimental tests[113] on a thermionic converter a plasma thermocouple operating in the core of a Los Alamos research reactor generated closed-circuit currents as high as 35 amp and an open-circuit voltage of 3.8 volts, indicating a power output through a matched load of the order of 30 watts. In this device the hot electrode is made of a solid solution of ZrC : UC with the uranium of the lower half of the rod enriched to 94 per cent. The collector electrode is stainless steel with copper cooling fins. Cesium was introduced after evacuating the container. The cesium would become highly dissociated and form the conducting plasma. When the device was inserted into a reactor core, the emitter rod heated to incandescence (about 2700°K) due to the fission reactions taking place. Cooling oil maintained the collector electrode at 1700°K.

SCINTILLATOR-PHOTOCELL BATTERY

Another method of obtaining electrical current from the energy of radiation is through the production of electrons from a photocell excited by light emitted from a mixture of a phosphor and a radioisotope capable of exciting the phosphor.[114] In the design of such a battery several of the important factors to be considered are: (1) half-life of the radioisotope, (2) decay scheme of the radioisotope, (3) efficiency of the phosphor in transforming the radiation energy into emitted light, (4) spectrum of light emitted by the phosphor, (5) response and efficiency of a photocell material to the light spectrum produced by the phosphor, and (6) cumulative radiation effects on all materials involved in the construction of the battery.

An example of such a battery is known as the Elgin-Kidde nuclear battery[114] and uses a Pm^{147} soft-beta source in combination with a cadmium sulfide phosphor and a silicon-diffused-junction photocell (see Fig. 5.23). This battery has a volume of only about 0.02 cu in. and initially will deliver 20 μw at 1 volt. Environmental tests of this battery indicate that temperatures up to 100°C have little effect on output.[114]

SEMICONDUCTOR BATTERY

The semiconductor battery is receiving considerable attention. Rappaport[46] in 1954 reported electron voltaic effect using a semiconductor having a p-n junction.[34] Silicon and germanium are usually employed (see Fig. 5.24).

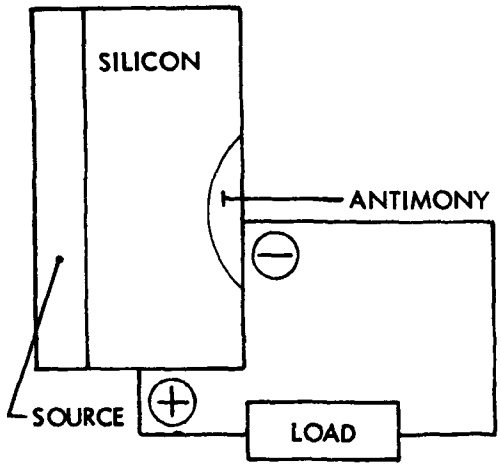


Fig. 5.24—Semiconductor type of atomic battery.[121]



Fig. 5.25—Experimental transistorized audio oscillator (left) powered with 0.050 curies of Sr^{90} in a semiconductor type of atomic battery. (Courtesy of E. G. Linder)[34]

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The semiconductor has three regions: p type, n type, and the p-n junction. Each region has three characteristic energy bands: filled, forbidden, and conduction. Normally electrons are in the filled band. Incident radiation creates electron pairs, removing the electrons in the conduction band while leaving behind the "holes" in the filled band. These charged particles diffuse to the junction region, where under the influence of the electric field they constitute the electric current.

The saturation current produced by the absorbed radiation is given as

$$I_s = E\ GL \tag{5.8}$$

where G = rate of generation in carriers per second
L = diffusion length
E = electronic charge

while the current which flows back to the junction is given according to the rectifier theory of Wagner[47] as

$$I_j = I_0\ (e^{EV/kT} - 1) \tag{5.9}$$

where kT = thermal equilibrium energy
V = electric potential

The net output current is given as the difference of two currents

$$I = I_s - I_j \tag{5.10}$$

This type of battery gives high current and low impedance, but is damaged by radiation. However, it has been found that if the energy of the radiation is below the threshold energy for the material, radiation damage can be prevented. The threshold energies are 0.3 Mev and 0.5 Mev for silicon and germanium, respectively.[48]

For a 0.3 cm^2 unit, using 50 mc of Sr^{90} as a source, a current of 10 amp at 3 per cent efficiency was obtained. The current varies linearly, while the efficiency varies logarithmically with the input power. If 7.5 curies were used, theoretical values for the power input saturation current and efficiency are P = 0.1 watt/cm^2, I_s = 5 ma, n = 6.6 per cent.

In comparison the experimental values for the solar energy conversion are: I_s = 8 ma; n = 5.95 per cent as reported by Pearson.[49] Recently an efficiency of 11 per cent was reported. Thus the semiconductor device holds promise and is being used in transistorized equipment. An example use is for an audio oscillator of 1000 cycles/sec powered by a radioactive battery consisting of 0.050 curies of Sr^{90} with a silicon junction 0.5 cm in diameter (see Fig. 5.25).

The following remarks can be made about all these methods of converting radiation to electrical energy:

1. The radioisotope should be cheap, abundant, a "soft" radiation emitter, and of long half-life. Nickel-63 with a half-life of 85 years fulfills all requirements but the first one. Some of the fission products fulfill the first but not all of the other requirements.

2. The problems common to all methods of conversion are: (a) Radiation shielding, (b) radiation damage, and (c) low efficiency.

3. The application of all these methods lies in the microwatt region. Instruments such as radiation meters, dosimeters, timing circuits, current and voltage references, transistor equipment, etc., can be supplied with microwatt radioactive batteries.

Two review articles on this subject were presented at the Geneva Conference, 1955, by W. Shorr[39] and Linder, Rappaport and Loferski.[34]

IRRADIATED ELECTROLYTIC CELL

Österlundh and Haeffner[50] reported studies on an electrolytic cell consisting of an electrode system in a chloroform-alcohol solution under gamma irradiation. The cell, which produces an electric current both during and after irradiation, consists of a Pyrex vessel holding

an electrolyte; a graphite electrode is fastened to the middle glass tube and surrounded by a perforated lead electrode. A gamma source is located in the center well.

Irradiating the chloroform-alcohol solution produces a number of decomposition products, including HCl. These decomposition products react with suitable electrodes to produce the electric current. The investigators[50] state that "a reaction between the electrodes and the decomposition products seems to be necessary for the cell to function." The current depends upon both the initial radiation chemistry reaction and the electrochemical reaction that follows.

The energy absorption in the cell was calculated to be 2.9×10^{-4} watt, whereas the greatest measured output was 2.5×10^{-6} watt (0.9 per cent conversion).

ELECTROSTATIC GENERATOR

Electrostatic generators using radioactive sources may be designed to convert nuclear energy directly into mechanical energy. Keller et al.[51] have suggested using such electrostatic devices in the Swiss watchmaking industry, as oscillators (regulating devices) and mechanical drives for springs or microsprings. Patek Philippe Laboratory[51] designed a miniature nuclear engine, which converted the electrostatic energy from the radioactive source directly into mechanical energy. A large number of problems still exist, such as insulation difficulties, degassing, deposition of radioactive material, and design of the electrodes.

HIGH-PRESSURE HYDROGEN-OXYGEN CELL

Hydrogen and oxygen from radiolysis of water have so far been considered as troublesome decomposition products of the water used as a fuel solvent, or for cooling or shielding of a reactor. Table 11.3 lists G values of about 2 for the radiolysis of water, which means that about two molecules of water will decompose per 100 ev of radiation energy absorbed. The high pressure hydrogen-oxygen cell developed by Bacon[52–56] may hold promise of converting these products into useful quantities of power. Table 3.2 shows that of the 182 Mev of energy from fission of one uranium nucleus, 6 Mev from high energy prompt gammas are lost—mostly in the thick concrete shield. Replacing the concrete with a water shield and using the radiolysis products in a high-pressure hydrogen-oxygen cell permits converting fission energy into power more efficiently. The Bacon cell has an efficiency of 66 to 75 per cent in converting the chemical energy of hydrogen and oxygen into electrical energy.[54]

Figures 5.26 and 5.27 show the principle on which the Bacon high pressure cell operates.[52–54] An experimental Bacon cell 9 in. high[52] and capable of producing 150 watts is illustrated in Fig. 5.28. This cell has been operated continuously for 1500 hr.

An important future source of battery power may lie in this cell. As more and more nuclear reactors operate, billions of curies of radioactive material are accumulating in their fission products. If the longest-lived products, Sr^{90} and Cs^{137}, could be separated completely from the reactor wastes, the remaining fission products could be dumped after a few years of storage, thus solving the disposal problem. A potentially large market for Cs^{137} has been found as a gamma source for radiation therapy in hospitals for industrial radiography, and as industrial gamma sources, but little use has been found for large amounts of Sr^{90}. Underground storage tanks containing aqueous solutions of Sr^{90} and producing hydrogen and oxygen in quantity by radiolysis of water may be one answer to Sr^{90} storage, with the possibility of considerable amount of battery power if the hydrogen and oxygen can be separated and subjected to large-scale conversion into power by high pressure Bacon cells. These permanent battery sources may be able to produce power in commercial quantities. Further study and research could well lead to an interesting addition to world power sources. Further information may be found in the references on industrial radiography,[57–66] conversion of radiation energy to electrical energy,[67–71, 121] beta-particle and gamma gauges,[72–84] neutron gauges,[85–96] self-luminescent materials,[97,98] and radioactive static eliminators.[99–101]

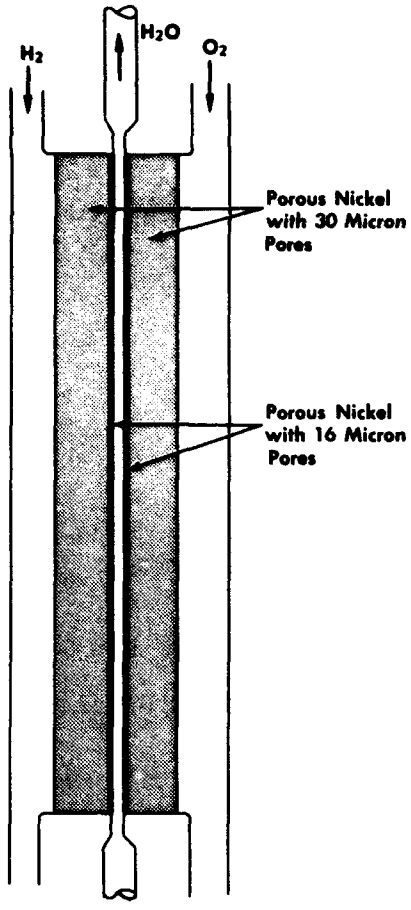


Fig. 5.26—Schematic section of porous-nickel hydrogen–oxygen fuel cell.[53]

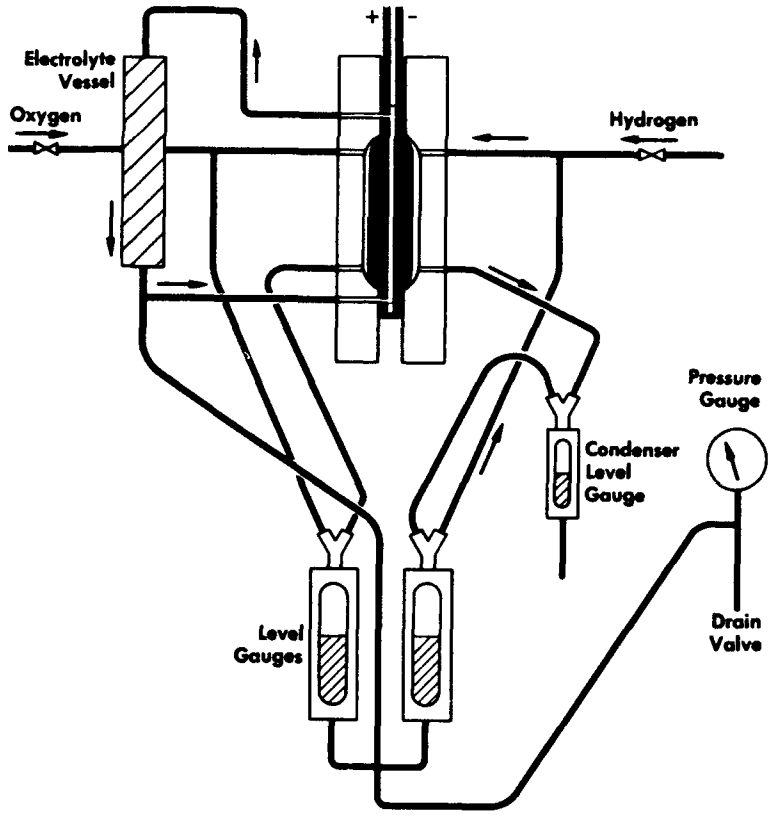


Fig. 5.27—The principle of the Bacon high-pressure hydrogen–oxygen fuel cell.[53]

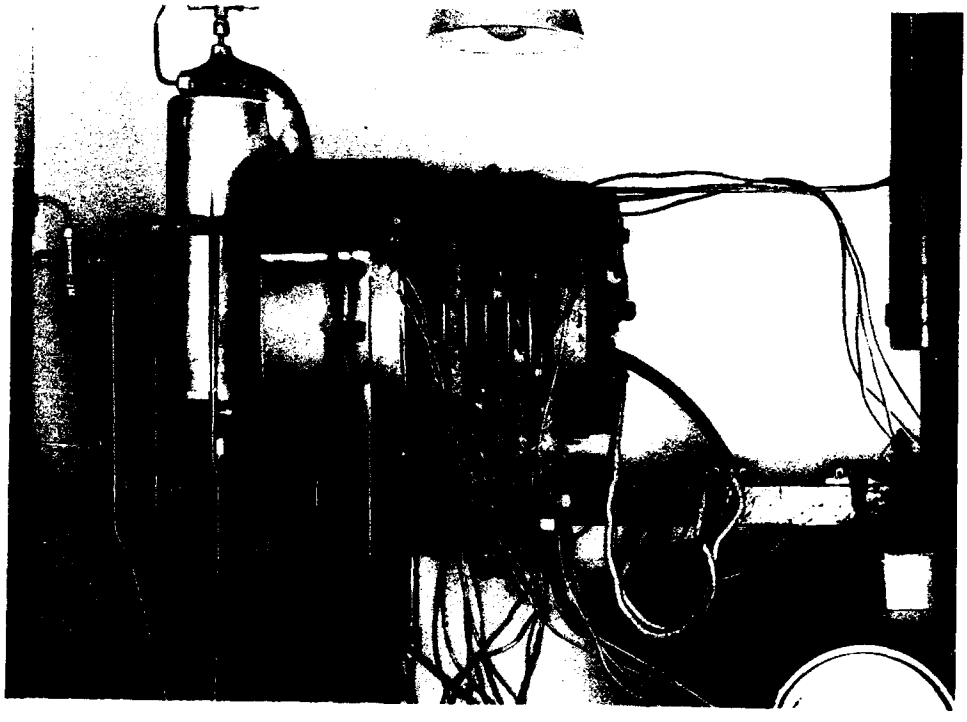


Fig. 5.28—Experimental Bacon battery of six hydrogen–oxygen cells in series giving an output of 150 watts. (Courtesy of F. T. Bacon)[52]

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Chapter 6

APPLICATIONS OF TRACER STUDIES AND AUTORADIOGRAPHY

In the field of research no other tool since the invention of the microscope has been as useful as radioactive tracers in extending knowledge. Many important results have been obtained during the few years that radioactive tracers have been used and many more will be obtained in the years to come. During the past 20 years, and particularly since World War II, thousands of reports have been published on the use of tracer amounts of radioisotopes for diagnosis in medicine, for control of variables in industrial processes, and for research studies in science. Most of these reports are technical and describe particular applications in specific and widely different fields. It is difficult, therefore, to comprehend the great importance of the new tool of radioactive tracers. The greatest use of radioisotopes over a 10-year period from Aug. 2, 1946, to Jan. 1, 1957, was in the field of medicine, with industrial use close behind. The uses at colleges, universities, and the state, federal, and institute laboratories include studies in fundamental research and in agriculture as well as uses in medicine and industry.

This chapter discusses selected applications and results in industry, research, and agriculture including wear studies and transfer of small masses, metallurgical applications, techniques used in the oil industry such as interface labeling in pipelines, tracer study of catalytic cracking and "acidizing." Road construction, evaporation, leak detection in underground piping are additional commercial uses discussed. In the field of pure research, a number of chemical reactions are considered, together with water analysis and diffusion studies including sulfur diffusion in rubber. Agricultural applications included are photosynthesis, fertilizer uptake, methods of fertilizer application, and animal husbandry problems.

TRACER METHODS OF ANALYSIS

The principle of analysis using radioactive tracers has received much publicity but the story of G. Hevesy, the originator of the method, may not be as familiar to the average scientific investigator, even though Hevesy received the Nobel award in 1943 for this important contribution to science.

In 1911 Hevesy was working with Rutherford in England where he attempted to separate RaD from lead by chemical means. At that time, it was not known that these two materials were isotopes and therefore could not be separated by chemical means but only by physical means. As a result of these studies Hevesy correctly concluded that RaD participated in every chemical reaction in which lead was involved. In 1913 at the Vienna Institute of Radium, Hevesy used this information to measure the solubility of nearly insoluble lead salts, such as the chromate and sulfide, using lead having some natural radioactivity.[1]

In addition to his studies with inorganic chemical systems, Hevesy also was the first to use this tracer technique in a biological system. The only radioisotopes available in these early days, however, were the naturally occurring radioisotopes. These have atomic numbers of lead and higher and involve isotopes that occur in biological systems to such a limited extent

that tracer studies with natural radioisotopes and biological systems in most cases are of only academic interest. Nevertheless, in 1923 Hevesy reported a precise study of the absorption of lead in plants, giving the percentage (of that originally present in the culture solution) going to the root, stem, leaf, and fruit.[2] He used the radioisotope known then as ThB, now called Pb^{212}. He measured the radioactivity of the ash from the various plant components by means of an electroscope, since reliable electronic counting equipment such as we have today was not then available.

As a result of his studies in the use of radioisotopes in chemical analysis, Hevesy in 1932 proposed the technique of analysis by isotope dilution.[3] Two years later the Joliots reported artificial radioactivity in elements exposed to alpha-particle radiation from natural radioisotopes.[4] This was followed by the development of particle accelerators such as the cyclotron and then the nuclear reactor, which made available new radioisotopes covering the entire range of atomic numbers. This opened the door to the use of the tracer method which became a new tool in the study and analysis of many different systems.

With the availability of a means of producing artificial radioactivity, Hevesy in 1936 also pioneered the development of another tracer technique known as activation analysis.[5] Seaborg and Livingood in 1938 reported studies on the technique of activation analysis.[6] The method of isotope dilution is still the widely used tracer technique, however. Other methods such as reverse isotope dilution, derivative dilution, activation analysis, and autoradiography have greatly increased the range of application of radioactive tracers. Most of these methods are discussed by Hevesy[7] in his book, *Radioactive Indicators*, published in 1948 and the principles of these methods will be briefly described here before describing selected applications.

ISOTOPE DILUTION METHOD

Analytical problems in chemistry, which might otherwise be impossible, may often be solved by the isotopic dilution technique. Suppose a protein hydrolyzate consisting of a mixture of as many as 20 amino acids is to be analyzed. Specific reagents for each of the 20 acids do not exist and quantitative isolation in the pure state is generally not possible.

By the isotopic dilution technique, this analysis may be achieved. The amino acid to be assayed is synthesized with its appropriate labeling isotope and a given quantity of this is added to the unknown mixture. The unlabeled amino acid will then serve as a dilutent for the "carrier" amino acid and the amount of dilution that occurs depends on the amount of unlabeled acid present. Thus, all the technique requires is that, after adequate time for mixing has been allowed, an amount of the added carrier (now diluted with unlabeled acid) be recovered in the pure state. It is important to note that only a portion of the amount of the amino acid has to be recovered pure. This is technically possible, whereas recovery of the total amount of any one acid in the pure state as required by a straightforward method of assay is often not possible.

The errors inherent in the method of isotopic dilution may be reduced below 1 per cent which is excellent accuracy for such determinations. The method may be used for the quantitative analysis of single elements and of molecules.

The mathematical relationship between the amount of added and recovered samples has been derived by Gest, Kamen, and Reiner for a general system.[9] The general relationship involving the analysis of a large number of components in a mixture is complex. However, a simple case dealing with the analysis of one component in a mixture will be discussed.

Consider the case of the mixing of two batches—batch 1 containing a labeled or "tagged" component and batch 2 containing the same component in the normal or "nontagged" state. The problem is the determination of the quantity of the component in batch 2. Let A equal the mass of nontagged component in batch 2, and A′ the mass of this component in batch 1 before the batches are mixed. If the specific radioactivity of batch 1 before mixing is A** and that of the mixture is A*, where the activity in counts per minute equal Y, then

$$A^{**} = \frac{Y}{A'}$$

$$A^{*} = \frac{Y}{A' + A}$$

and, therefore

$$\frac{A^{**}}{A^{*}} = \frac{A' + A}{A'}$$

or

$$A = \left(\frac{A^{**}}{A^{*}} - 1\right) A' \tag{6.1}$$

The method has two advantages: it is capable of giving higher accuracy especially in biochemical assay than available chemical techniques and quantitative chemical separation procedures are not necessary. For accuracy with radioactive labeling, the change in specific activity on dilution must be at least by a factor of $\frac{1}{2}$. It is evident that both the isotope added and the sample isolated from the mixture must be obtainable in a chemically pure condition.

REVERSE ISOTOPE DILUTION METHOD

A modification of the isotope dilution technique termed reverse isotope dilution may be used to advantage when the study involves a system which already contains a tagged component of interest. In this method, a measured quantity of unlabeled component is added to a mixture containing the unknown quantity of labeled component. Portions of the initial and the final mixtures are recovered and assayed. The relation between the initial and final specific activities is as follows:[8,9]

$$A = \frac{A'}{\frac{A^{**}}{A^{*}} - 1} \tag{6.2}$$

where A = amount of inert component originally present
A' = amount of inert component added
A^{**} = specific activity of extracted sample before dilution by the inert component
A^{*} = specific activity of extracted sample after dilution

If a large excess of the inert component is added, the ratio of specific activity before dilution to that after dilution (A^{**}/A^{*}) becomes a number much greater than unity and in this case Eq. 6.2 reduces approximately to the following:

$$A \simeq A' \frac{A^{*}}{A^{**}} \tag{6.3}$$

if $A' \gg A$

ISOTOPE DERIVATIVE METHOD

Both the isotope dilution and reverse isotope dilution techniques require the production of a specific labeled compound identical to that under study in the particular system being investigated. The production of this specific tagged compound may be difficult or inconvenient. In this case, the isotope derivative method may be used to advantage since in this procedure a simple radioactive reagent may be used rather than a specific labeled compound.[10] One procedure of the isotope derivative method involves the use of radioactive reagent of known specific activity which is used to convert the inactive component of interest into a radioactive derivative. Any excess of the radioactive reagent is then removed and the specific activity of the tagged derivative determined by means of a material balance and count of radioactivity. Then a measured weight of the inactive form of the derivative is added which dilutes the tagged derivative. The mathematical expression used for the analysis is the same as in the reverse isotope dilution method as given by Eq. 6.2 but the terms A and A′ are defined differently.

$$A = \frac{AA'}{\frac{A^{**}}{A^{*}} - 1} \tag{6.4}$$

where A = amount of radioactive derivative originally present
A′ = amount of inactive derivative added

RADIOMETRIC ANALYSIS METHOD

In radiometric analysis, the radioactive isotope is used to determine the presence of another nonradioactive element using analytical laboratory techniques such as precipitation, filtration, centrifugation, and titration, except that the radioisotope serves as a radioactive indicator. In one procedure a tagged element or compound is selected that will react with the element or compound under analysis so as to form a precipitate. An example would be the use of radiosilver to analyze for the chloride ion. In this case radioactive silver nitrate added to the test solution will be precipitated out until the chloride ion in the solution has been consumed. A plot of the radioactivity of the filtrate as successive additions are made to the parent solution will show a constant low activity if the radioactive silver has been removed by precipitation and filtration and will show a break in the curve at the end point where the chloride ion is exhausted. Thereafter, the activity of the filtrate will increase in proportion to the addition of the radioactive silver nitrate solution.

In this method of radiometric analysis, the tagged component reacts with the material to be measured as in the isotope derivative method, but the inactive derivative is not added and the quantitative determination depends on ordinary analytical procedures rather than on the count of the radioactivity. The count serves only as an indicator of the completion of a chemical reaction. (See Fig. 6.19 and Ref. 122.)

ACTIVATION ANALYSIS METHOD

Activation analysis is a method of analyzing for certain elements in a sample by inducing radioactivity into atoms present in the sample rather than by adding tagged atoms as described for the other tracer methods. The nuclei of the sample elements are activated by nuclear particles from some external source. In 1936 Hevesy and Levi[5] first suggested analysis by radioactivation and described procedures using neutrons from a 300-mc radium-beryllium source. Without chemical separation, these pioneers were able to analyze rare earths for concentrations as low as 0.1 per cent. Today more powerful neutron sources are available which increase the sensitivity of this method of analysis. A radium-beryllium source weighing 25 mg will produce a thermal-neutron flux of about 100 neutrons/cm^2-sec. High-velocity protons or deuterons in a particle accelerator may also be used for activation, but neutrons with thermal energies are most frequently used.

In addition to the means of [illegible]ion, this method of analysis requires the use of sensitive radiation-detection equipment a[illegible]owledge of nuclear properties of the isotopes of the element being studied. Since isotop[illegible] different elements may be activated in the sample, an exact knowledge of the nuclear properties of the different isotopes permits analysis for each on an individual basis. It is necessary to know the probability of neutron capture, termed cross section, for the isotopes in question as well as the half-lives of the daughter radioisotopes produced. Since the cross section varies with neutron energy, but is usually large in the thermal region, activation analysis data are collected at one definite energy in the thermal region. In some systems unwanted radioisotopes of the other elements may have short half-lives and may be eliminated by decay before analysis. For analysis by activation: Meinke[11] gives

$$A' = \frac{YA}{6.02 \times 10^{23}\, f\, (\sigma)\, (1 - e^{-0.693t/t^{*}})} \tag{6.5}$$

where A′ = weight of element in sample
A = atomic weight of element
Y = measured activity, disintegrations/sec

f = neutron flux in activator, neutrons/cm^2-sec
σ = atomic cross section, barns (isotope cross section times per cent abundance)
e = Napierian base, 2.718
t = duration of neutron bombardment, hr
t^* = half-life of radioisotope produced, hr

In addition to direct analysis by use of the technique described, neutron activation is also used to tag a component in a complex system after which the analytical determination may be made by the isotope dilution method or other tracer techniques. Additional information on activation analysis is given in Refs. 11 to 15.

AUTORADIOGRAPHIC METHOD

The problem of the determination of amounts of radioactivity is basic to all tracer studies. However, there may be interest not only in the amounts of radioactivity but also in its location within the specimen. This problem of location has resulted in the use of photographic emulsions for the measurement and location of radioactivity.

As early as 1904 photographic means were used to measure the distribution of activity in ore specimens,[16] and until fairly recently the use of photographic films was restricted to such macroscopic studies. With the advent of the artificially produced isotopes there has been increasing interest in extending the use of photographic films to microscopic studies. The term autoradiography is generally understood to mean the use of photographic films for the localization of radioactivity on a microscale. Theoretically it would be desirable to be able to locate and detect a single atom. The diameter of one atom is approximately 1 A, or 1×10^{-8} cm. At present, under ideal conditions, resolution of the order of 1 μ (10^{-4} cm) can be obtained. Thus there exists opportunity for great improvements over present techniques.

In principle, the technique consists of placing a photographic film in intimate contact with the specimen long enough to enable the radioactivity to activate the silver grains in the emulsion. The film is processed, using routine photographic procedures. For macro work, the film is clamped on the surface of the specimen and stored in a light-tight container for the duration of the exposure period. The film is then removed and processed in the usual manner. The areas of blackening are then correlated with the areas of radioactivity in the sample. The films usually used for this purpose are medium grain size lantern-slide plates, no-screen X-ray film, and NTB plates (plates with special emulsions for recording tracks produced by nuclear particles). An example of such an application using X-ray film is the use of paper chromatography for the detection of various chemical compounds. In such an analysis the compounds are separated into different areas on the paper and located by treatment with various dyes. In order to find which compounds contain the radioactive tracer, the paper can be clamped in an ordinary X-ray cassette with a sheet of X-ray film and left there for a period of time. After the film is processed, the areas of blackening will correspond to the areas on the paper that contain the radioactive tracer.[17] For high resolution detection and localization, the sample and autoradiograph may be greatly magnified with a microscope. Additional information on autoradiography is given in Refs. 17 to 20.

COUNTING NUCLEAR RADIATIONS IN TRACER EXPERIMENTS

Radiochemical analysis depends on accurately determinating the disintegration rate of a radioactive sample. This is not only true in tracer techniques previously described, but also in the determination of half-life and various other assay methods. For a given radioisotope, the rate of disintegration, on the average, is directly proportional to the rate of emission of nuclear radiations. The "counting" of the nuclear radiations, or of a known fraction of the nuclear radiations, over a given period of time, permits this determination. Today, electronic instruments known as counters are used in this procedure. However, early workers counted alpha particles by the tedious process of observing the scintillations on a plate coated with a phosphor. The equipment required for rapid and accurate counting consists of a detector such as a Geiger-Müller (G-M) tube, scintillation counter, etc., and an assortment of auxiliary equipment for shielding the sample and timing and recording the counts.

GEIGER-MÜLLER COUNTERS

The Geiger-Müller counter is the most widely used instrument in counting radioactivity. For the sake of brevity, this instrument is sometimes referred to as a Geiger counter or a G-M counter. The detecting portion of the instrument consists of a G-M tube.

The modern highly sensitive form of the G-M tube was designed by Geiger and Müller in 1928 in Germany. However, this sensitive radiation detection instrument could not be used to full advantage until modern vacuum tubes and circuits were developed. Figure 6.1 shows typical types of G-M tubes.

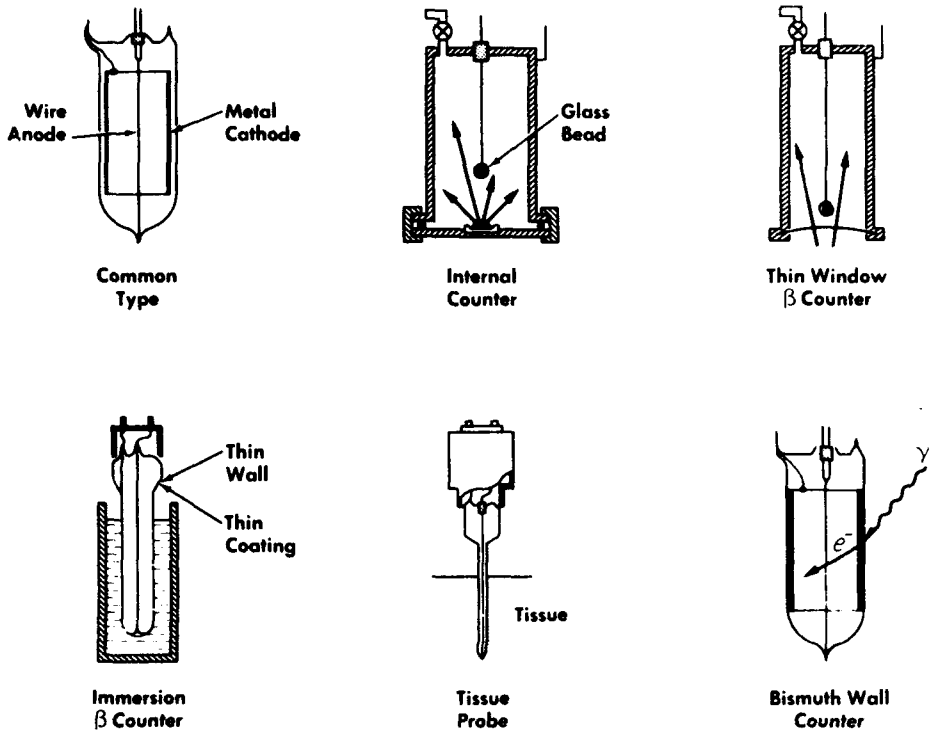


Fig. 6.1—Some types of G-M tubes.

The G-M counter, as the term implies, is a counting instrument in which a pulse is produced for each ionization event above a certain magnitude that occurs in the sensitive volume of the tube. This pulse may be indicated without record by a flashing light, an audible ticking, a ratemeter, or may be recorded on an instrument known as a scaler. Such signals are operated by the output current from the electronic amplification circuit of the counter. The high voltage used with a Geiger tube results in a large multiplication of an original ion pair produced by the interaction of radiation and a gas atom or molecule; the amplification may be as high as 10^8. An ion pair produced by radiation results in an avalanche of ionization, near the anode, in the region of the original ionization. The avalanche spreads along the entire anode. Once the avalanche travels the length of the anode, the electrons released are rapidly collected at the anode. The positive ion of each ion pair consists of a gas molecule less one or more electrons; it has much less mobility than the electrons and migrates toward the cathode much more slowly than the electrons move to the anode. This results in an accumulation of a positive space charge near the anode, which decreases the electric field near the cathode and terminates the ion avalanche.

The G-M tube produces a pulse, the magnitude of which is the same for all the basic types of ionizing radiations. Alpha particles, beta particles, and gamma photons all produce pulses of the same magnitude. Most sealed G-M tubes, however, are not suitable for counting alpha particles because the walls and windows stop most alpha particles. All alpha particles and beta particles can be stopped by the wall of a G-M tube with a thick window; therefore, thick-

windowed G-M tubes are used for gamma counting. Alpha particles and weak beta particles may be counted in a flow counter where the radioactive material is actually inside the tube volume.

In operating a G-M tube, the voltage must be increased to a certain minimum value known as the starting potential before any pulses are produced. The pulse rate then rises rapidly until a plateau is reached. As the voltage is increased, the pulse rate increases only slightly with voltage over a region known as the Geiger plateau, the voltage range in which a G-M tube is operated. Beyond this plateau the breakdown voltage is reached, above which continuous discharge occurs. Figure 6.2 is a curve of counts per minute vs. applied voltage for a G-M tube.

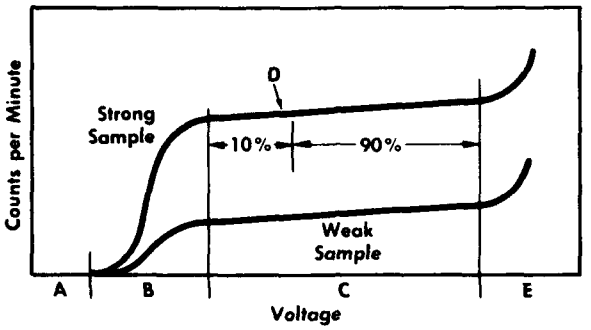


Fig. 6.2 — Counts per minute vs. applied voltage for typical G-M tube: A) No pulses large enough for recorder; B) Avalanches grow larger with voltage, and number of recorder pulses increase; C) All particles trigger maximum avalanche and all pulses recorded; D) Optimum setting for operating voltage (higher voltage gives shorter tube life); and E) Spurious discharges lead to breakdown.

The magnitudes of the starting potential, the Geiger threshold and the breakdown voltage depend on the gas used, the gas pressure, and the design of the tube. Starting potential varies from around 800 to 1800 volts. The length of the plateau may vary from 100 to 300 volts. The slope of the plateau in the proper operating range is usually about 2 to 5 per cent per 100 volts. It is important that the slope be small if the G-M tube is to give the same pulse rate for the same radiation field even when line voltage varies. The operating voltage is usually selected to be somewhat less than the midpoint of the plateau.

SCINTILLATION COUNTERS

The phenomenon of luminescence induced by radiation has been employed since soon after the discovery of radioactivity in the study and analysis of radiation. In 1899 Becquerel found that ionizing radiation produced luminescence in a variety of materials, including zinc sulfide, diamonds, and platinocyanide. In 1903 Crookes reported that the luminescence produced in zinc sulfide by alpha radiation was nonuniform in character and actually consisted of a number of individual flashes that could be observed under a microscope. In 1904 Rutherford commented that each scintillation was probably the result of the impact of individual alpha particles and that the number of particles could be determined by counting the individual flashes. This was later demonstrated by Rutherford and Geiger, who allowed alpha particles from RaC to impinge on some zinc sulfide. These counts gave good agreement with those obtained with an early Geiger tube. However, the visual method of counting flashes was very tedious and was discarded in favor of counting tubes such as the G-M tube. The recent development of the photomultiplier tube renewed the interest in the scintillation phenomenon and has stimulated the development of very sensitive instruments called scintillation counters. An advantage of scintillation counters is that rates of over a million counts per second are possible. Basically, the scintillation counter consists of two parts, a phosphor and a photomultiplier circuit. The

phosphor absorbs a portion of the energy of radiation and converts it to light. The photomultiplier tube transforms the light into an electrical pulse which is amplified by a vacuum tube circuit to operate a counter. The schematic diagram of a scintillation counter is shown in Fig. 6.3.

The photomultiplier tube can be placed in a probe which is very useful in medical applications such as localization of brain-tumor using radioactive diiodofluorescein (I^{131}). In such applications, the scintillation counter probe is several times as efficient as the G-M tube probe. Another important use of the scintillation counter is the field of gamma-ray spectroscopy for making accurate determinations of the energy of gamma photons. In such studies, and for research experiments requiring greater sensitivity than possible with ordinary G-M counters, the well type scintillation counter is used. A sectional diagram of such a counter is shown in Fig. 6.4.

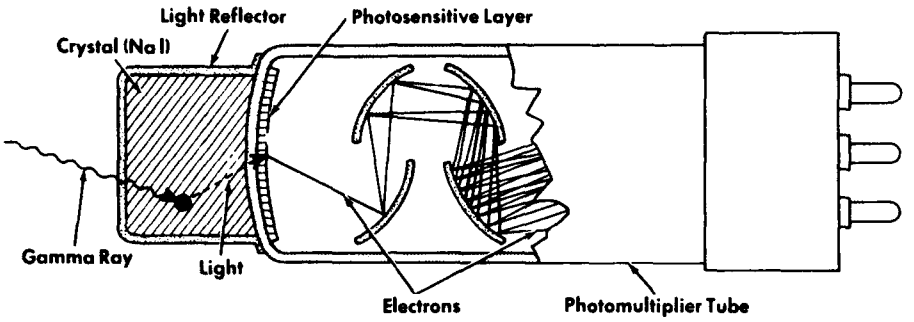


Fig. 6.3—Schematic diagram of a scintillation counter.

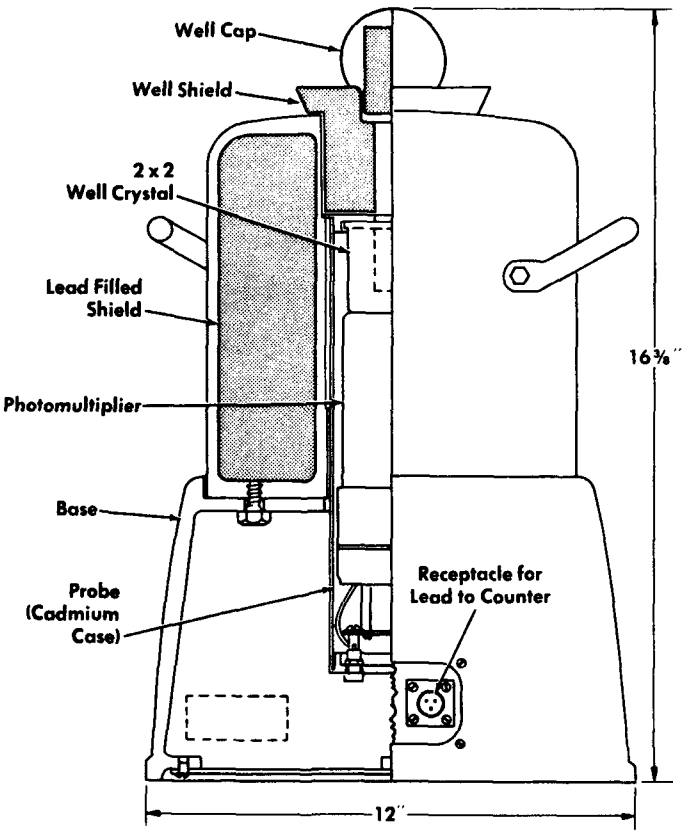


Fig. 6.4—Sectional view of well-type crystal scintillation counter. (Courtesy of RCL, Inc.)

With reference to Fig. 6.4 the sample to be counted is placed in the porthole within the crystal well. The lead shield cuts down background radiation. There are also shields available where the sample is introduced through a door in the side. The light produced in the well crys-

tal by scintillation passes through the well pipe into the photomultiplier tube. The signal pulses pass through the preamplifier at the base and then to an auxiliary scaler by means of the cable leads.

In addition to such special applications, the scintillation detector tube can be substituted in most instances for the ionization chamber, the proportional counter tube, or the G-M tube. Like the ionization chamber, it permits measurements linearly related to the radiation absorption over a wide range of quantum energies. It has the advantage of greater stability than the ionization chamber when operated at full sensitivity. As compared to the proportional counter, both are capable of high count rates and both give pulse amplitudes that depend on the type and energy of the radiation. The proportional counter probably has greater discrimination with regard to radiation energy. The scintillation counter is faster and has a greater life, greater counting rate, and greater efficiency for higher energy radiation than the G-M counter.

AUXILIARY AND COUNTING EQUIPMENT

A certain radiation level is always present because of the background produced by cosmic radiation, radon in the air, and minute amounts of radioactivity in structural materials and in the earth. For example, the uranium present in 1 lb of granite has a potential fission energy equal to the potential combustion energy of 1 lb of coal. This uranium decays slowly (half-life = 4.5×10^9 years) contributing to the background. The background level of radiation limits accurate counting if the count rate of the sample is small. Also the background level changes from day to day, depending on the weather (fresh rain dissolves the radon in the atmosphere). To provide a low, more uniform background level for accurate counting, a lead or iron shield is used around the sample and detection tube.

Manual counting of pulses is very tedious and can be performed only at low count rates. Therefore, most counting is performed by instruments known as counters or scalers. The counter contains a vacuum-tube amplification circuit similar to those described previously for the various types of detectors (ion chambers, G-M tubes, scintillation wells, etc.). The pulse is amplified to provide sufficient output to operate a mechanical register. However, the maximum rate at which a mechanical counter can register is about 30 to 50 counts/sec. To permit counting at a higher rate, binary circuits are included in the measuring system. Each binary circuit produces one pulse for each two pulses it receives. The use of a scaler in which the register records once for every 64 counts (thus, 6 binary circuits) is common. Scalers called decade scalers are also available, based on factors of 10 rather than factors of 2. In using a scaler, the number indicated on the register is multiplied by the scaling factor to give the total number of counts for the period that the scaler is operated.

Some scalers may be operated on a scale of 10, 40, 100, 400, or 1000 or on any binary scale of 16, 64, 256, 1024, and 4096 by setting the selector on the face of the scaler. The scaler contains a built-in register on the face of the instrument. Controls for the voltage regulation, pulse height, attenuation and automatic or manual operation are also on the face of the instrument. An external timer may be used with the scaler or a timer may be included in the scaler.

Effect of Alpha-Particle Radiation on Counting

The efficiency of scintillation detectors may be made very nearly 100 per cent for alpha- and beta-particle emitters with particles having energies of about 100 kev or more. Due to their low penetrating power, however, alpha particles are generally counted in a windowless flow counter operated either in the G-M region, or proportional region, or with some form of ionization chamber.

Alpha emitters are rarely used in tracer experiments. The only elements having alpha-emitting isotopes are polonium, thorium, uranium, radium, and other elements of atomic number 206 and higher.

Effect of Beta-Particle Radiation on Counting

Beta-particle emitters are used more often in tracer studies than either alpha-particle or gamma-photon emitters. However, the accurate counting of beta particles is complicated by

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consideration of the over-all efficiency, sometimes referred to as the geometry factor. This geometry factor depends not only on the geometry of the counting arrangement but also on the tube efficiency, process efficiency, side scattering, back scattering, window absorption, air absorption, cover absorption, self-absorption, dead time, sample thickness, sample area, and energy of the beta particles. Some of these factors are indicated in Fig. 6.5.

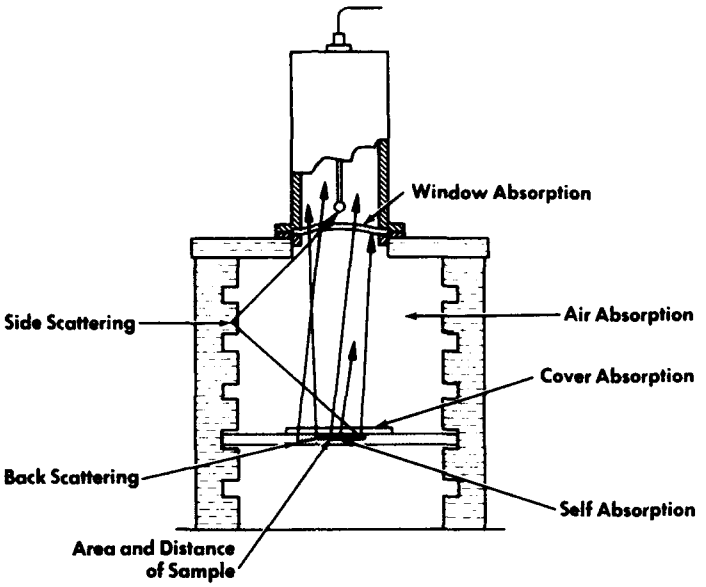


Fig. 6.5—Diagram showing some of the corrections involved in counting the activity of beta-particle emitters.

The geometry factor is always important, since it describes the maximum percentage of the total number of particles that can be counted. It varies from approximately 50 per cent for a windowless flow counter to values ranging from about 30 per cent to 1 per cent or less for end window counters.

The absorption factor may be 20 per cent lower for a strong beta-particle emitter like P^{32} than for a weak beta-particle emitter such as C^{14}, when using a thin-window tube. With thick windows or longer air paths, the absorption factor may be much higher for weak beta-particle emitters and the effects of self-absorption become appreciable with them. In each case the geometry factor will depend on the physical arrangement used. The determination of all the correction factors is not necessary if one can keep them constant. This indicates the advantage of using a fixed geometry and mounting samples in the same way. In the case of the weak beta-particle emitters, one should try to use samples that are of the same thickness (preferably thick enough to have the properties of infinite thickness) and in the same chemical form. The self-absorption factor also depends on the density of the material. Therefore it is advantageous to count samples having the same density. In C^{14} tracer experiments it is quite common to convert all samples to barium carbonate for counting.

Effect of Gamma Radiation on Counting

G-M counters can be used for counting the activity of gamma emitters, but the efficiency of these counters is very low (around 1 per cent) compared to their high efficiency when used for counting beta particles. When the activity levels are high enough, however, G-M tubes are used for gamma counting. Special tubes have been designed incorporating high density plates made of bismuth metal within the counting volume in order to increase the gamma interaction probability. Such tubes have efficiencies as high as 4 or 5 per cent.

For greater efficiency, one often uses a gamma scintillation counter. These counters usually have gamma counting efficiencies of 20 to 50 per cent. The high efficiency results both

from the high densities of some phosphors that are available and from the large possible detecting volumes that can be used. The latter point is particularly well illustrated by the plastic and liquid scintillators that have been devised. High counting rates are achieved by using phosphors, such as stilbene, which produce very short light pulses. The resolving time of the counter can be made quite low. The most commonly used counter is the well type scintillation counter described previously and shown in Fig. 6.4. It has the advantage of good gamma efficiency and excellent geometry. The gamma scintillation counter has the additional advantage of being energy sensitive; i.e., it can be used in a gamma spectrometer system to distinguish one isotope from another.

The sample preparation is much simpler for gamma emitters than for beta-particle emitters, since in most cases absorption and self-absorption are negligible. This means that usually there is no problem in regard to sample thickness, chemical form, or density.

Absolute gamma counting is usually done by comparison with a known standard of the same isotope or some long half-lived standard which has a similar gamma spectrum.

Figure 6.6 shows a research worker placing a radioactive sample in a lead shield for counting with a scintillation counter connected to a scaler and a recorder.



Fig. 6.6 — Placing radioactive sample in shield for counting. (Courtesy of Goodyear Tire and Rubber)

INDUSTRIAL APPLICATIONS

Tracer analysis has been used for wear studies in the tool and the machinery industries, for the study of mass transfer in the transistor field, for the detection of minute amounts of impurities or residues, for oil prospecting in the petroleum industry, for the determination of optimum mixing in the chemical industry, and for a multitude of other industrial applications.

WEAR STUDIES

Studies on wear have been carried out extensively in connection with automobile engines, tires, gears, dies, electrical contacts, turbine cavitation, cutting tools, and plastic molds. One of the first studies was made by Ferris[21] who investigated the wear of piston rings in an automobile engine by using radiophosphorus produced by activation in a cyclotron beam. In 1947 Pinotti[22] made piston rings radioactive by exposing them to the neutrons inside a reactor. Tool wear studies have been carried out using radiocobalt, tungsten, tantalum, and chromium. In 1949 a special cutting tool having a radioactive tip of W^{185} (half-life = 73 days, beta energy = 0.43 Mev, gamma energy = 0.13 Mev) was designed to study the rate of wear. The radioactivity of tool particles in the chips was measured with a scintillation counter. The activity was found to be about 100 times the background from 100 g of chips.[23,24] Tool wear data usually obtained in 1 year by conventional methods have been obtained in 2 to 4 weeks by tracer methods.

One study of the wear of gears reported by Schwartz and Eaton[25] will be described in some detail as an example of this technique. In this study the rate of gear wear was determined by measuring the radioactivity of oil circulating continuously over one gear, which was made radioactive for test purposes. The cast-iron gear that was to be tested was placed in a reactor to produce Fe^{59} (half-life = 2.9 years). For purposes of calibration, iron powder of the same composition as the gear was wrapped in aluminum foil and placed in an opening in the base of the gear before insertion in the reactor so that on irradiation, both received the same dose. A known weight of the radioactive iron powder was suspended in a known volume of oil and the oil and suspended powder were circulated through a closed system. Varying the ratio of iron powder to oil gave the calibration curve.

In testing gear wear, the radioactive gear was installed in the sun-gear position in a gear case with long-handled tongs. The oil system was then filled with one of the oils used in the study and the gears operated for 4 hr for "breaking-in." The gears were loaded using a "four-square" or locked torque test system. The test assembly is shown in Fig. 6.7

In determining the gear wear, the calibration system was modified to circulate the oil from the sump through the gear box as well as through the calibration system by means of a separate pump, as shown in Fig. 6.8. A number of variables were studied in these tests. Figure 6.9 shows the results of gear wear for two oils as a function of gear speed and with parameters of torque.

Radioisotopes also have been very useful in seeking means of preventing or reducing the actual amount of wear.[26,27] Ordinary lubricants do not provide satisfactory service under applications of high unit loading such as found in hypoid gears. Chemical agents are used as additives to the lubricants in order to increase their effectiveness. These additives affect the ability of the lubricant to form surface films, but the exact mechanism involved has yet to be determined. As an aid in studying the nature of the film formation several investigators have used radioisotopes. The procedure followed in many of these investigations was to tag the chemical additive with a radioactive tracer. Then, a known percentage of the additive was mixed with the lubricant. The metal specimens were immersed in the lubricant and after a fixed immersion period were cleaned in a benzene bath which removed the surface film from the metal. The quantity of lubricant that was removed from the surface was determined by standard tracer techniques. This method was used to determine the effect of temperature and surface treatment on the adherence of certain additives to the metallic surface.

Figure 6.10 shows the degree of binding that was obtained on the surface of parts of a V-8 engine when phosphorus was used as an additive to lubricating oil.[27]

Additional information on the dynamic and static testing of lubricating oil additives may be found in Refs. 28 to 40.

Radioactive phosphorus has been used as a labeling agent in studies on the wear of rubber tires by incorporating the radioisotope in triphenyl phosphate which is used as a plasticizer. A study was made of the wear of tread rubber of the auto tires on starts, stops, curves, and straightaways. In these studies the tread wear was determined by measuring the activity of P^{32} on the road surface.[41] Wear and erosion problems of dies and electrical contacts between two surfaces have been studied by radioactive tracers.[42,43] The effect of generator load on the rate of erosion of a turbine wheel as a result of cavitation was determined by painting the wheel with a paint containing a small amount[44,45] of As^{76}.

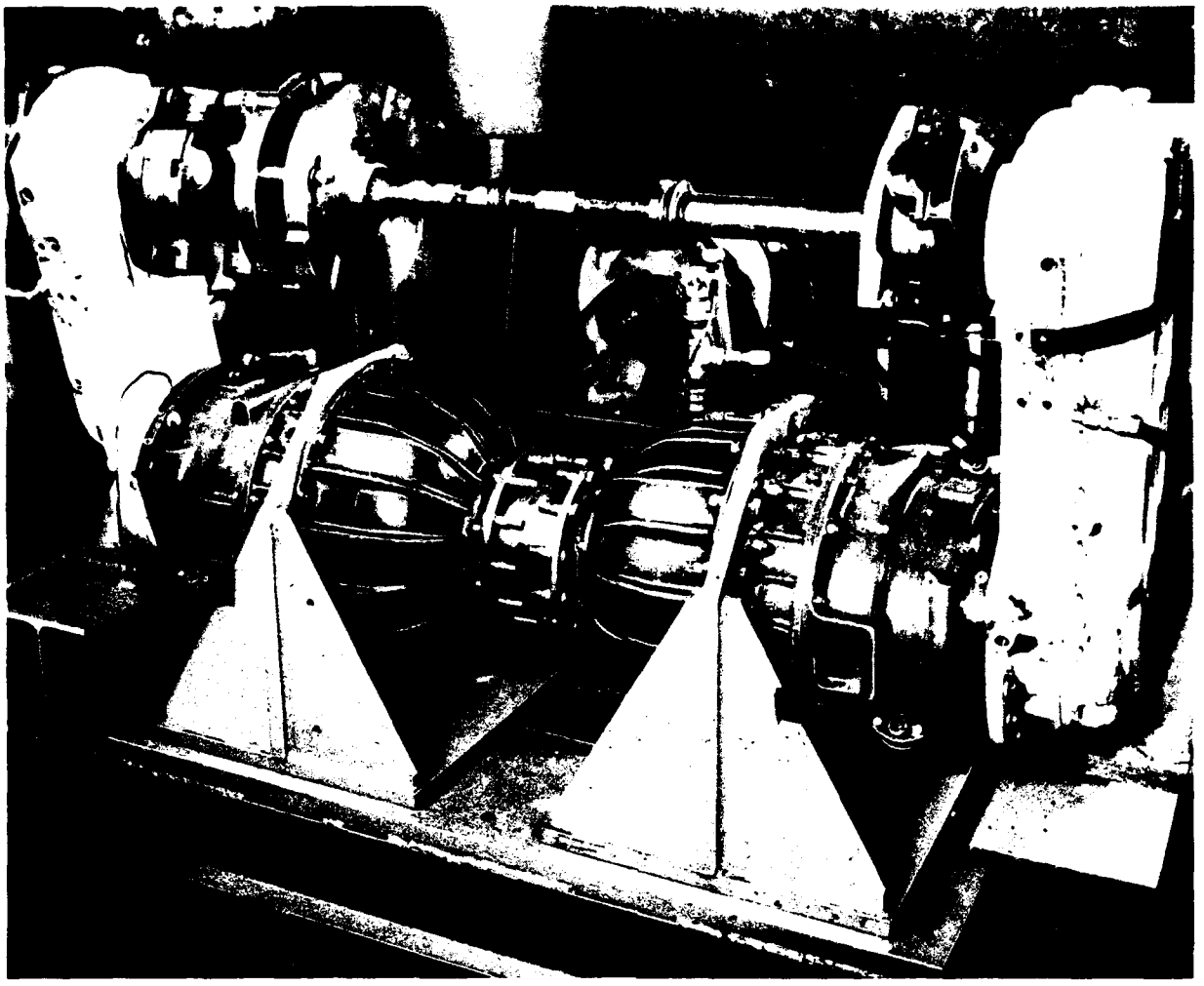


Fig. 6.7—"Four-square" gear-test arrangement.[25]

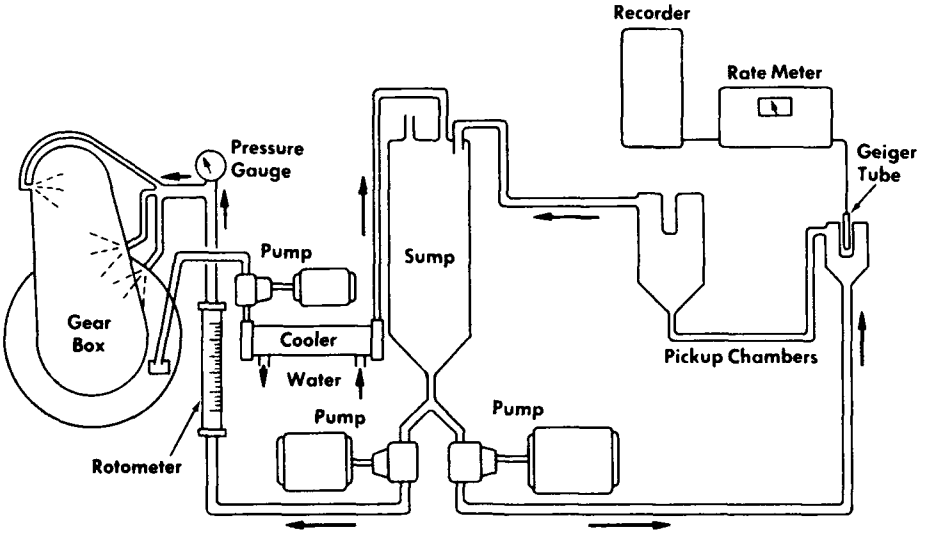


Fig. 6.8--Schematic diagram of gear-testing system.[25]

In the field of plastics, the effects of mold pressures, preheating, resin modification, lubricants, and fillers have been found on the mold erosion.[46] Using radioactive tracer methods, the mold erosion test can be made in 30 min, compared to 3 weeks required by previous methods. Additional information on the use of radioisotopes in wear studies is given in Refs. 47 to 52.

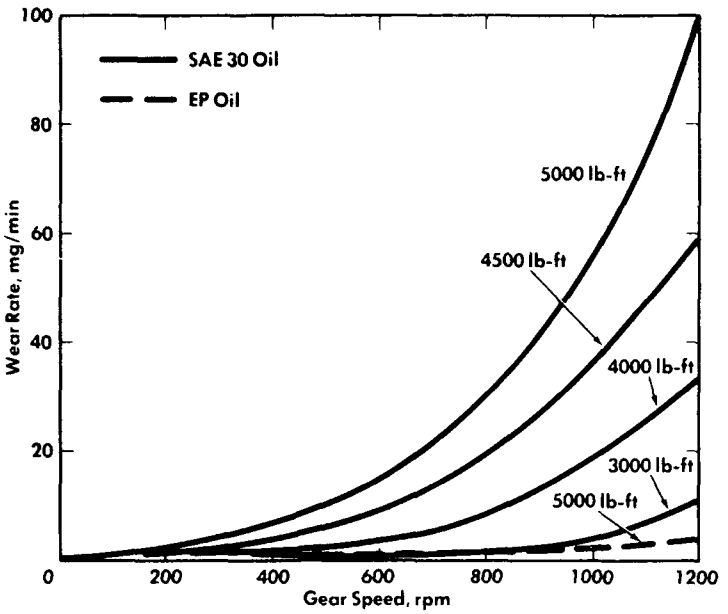


Fig. 6.9—Gear wear vs. gear speed under different loads.[25]

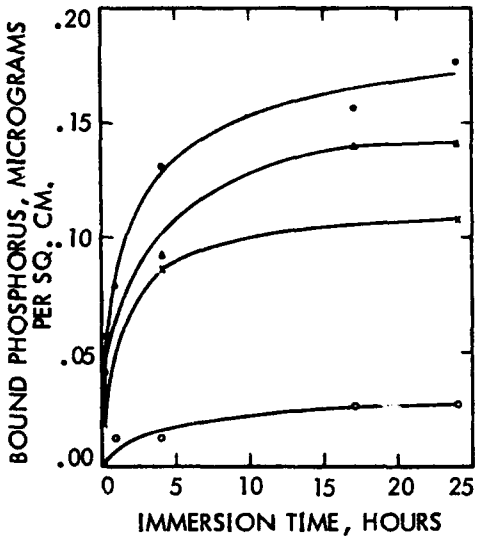


Fig. 6.10a—Rate of film formation as a function of surface treatment.[27]

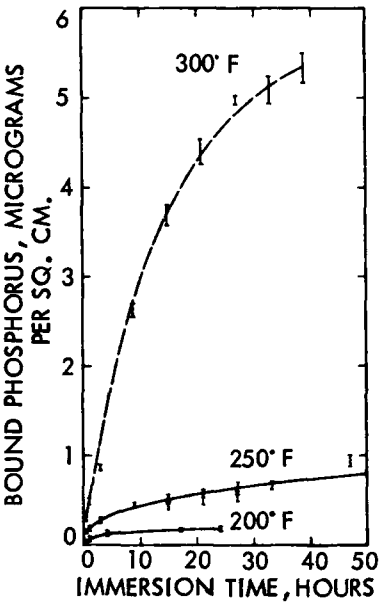


Fig. 6.10b—Effect of temperature on film formation with lubricated tappets.[27]

MASS TRANSFER

Tracer analysis has been successfully employed in the detection of the transfer of small quantities of material. Gold-199 has been used in the study of the mass transfer in transis-

tors.[53,54] The gold transferred from a gold-plated tungsten electrode to a germanium semiconductor in the presence of a voltage pulse was determined by using the technique of autoradiography. The amount of gold transferred was found to be about 10^9 atoms or 3.3×10^{-13} g in a volume of 10^{-7}cm^3 of the crystal. Autoradiographs showed that the gold was distributed in a conical shape with a base 100 μ in diameter and the apex 2.5 μ above the base.

Radioisotopes have been used in tracing material difficult to separate from other materials. In one application Ca^{45} has been employed in the detection of lime residue adhering to the steel wires drawn from the lime tank. It has been possible to detect a lime coating 2.5×10^{-6} in. thick on a steel wire.[53] Water labeled with tritium has been used in the study of the migration of the moisture into protective varnish coatings.[55]

In the multicolor printing of textiles "color soiling" may occur if one color is carried forward from one dye box to the next. Such color contamination reduces the value of the printed textiles and many methods of control have been tried. The most satisfactory solution has involved the use of radioactive tracers to label the "pirate" color so that its gradual invasion can be continuously monitored, as indicated in Fig. 6.11. In such an application a few millicuries of P^{32} as soluble phosphate is added to the dye bath of the pirate color. Migration of the pirate color to the next dye bath is continuously checked by dip counters.

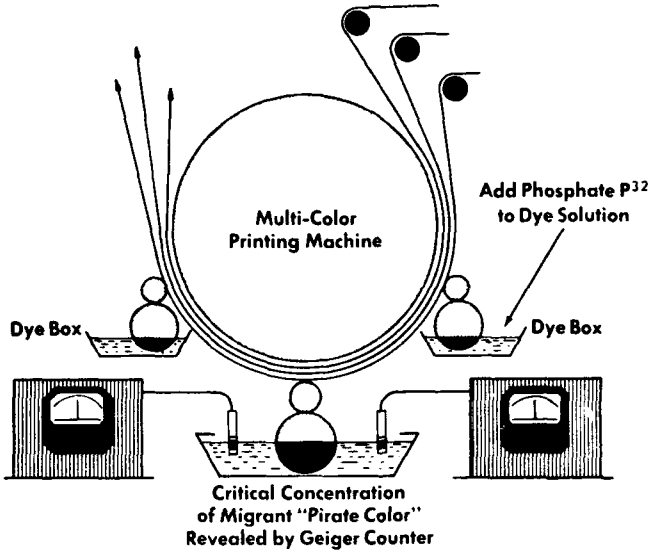


Fig. 6.11—Detection of dye migration with P^{32}.

The textile industry has used radiotracers extensively in other applications. A few of these involving mass transfer are:

1. To test the efficiency of soil removal techniques.
2. To study the wear of mechanical parts of textile machines.
3. To mark individual wool fibers in order to study their movement in drafting.
4. To determine the absorption of a detergent in a fabric.
5. To measure the diffusion coefficients in polymers.
6. To investigate the uniformity of oiling of nylon.

METAL INDUSTRY

Material from special steel melts must be followed through the various steps in fabrication and is often difficult to identify. Radioactive tracers may be added to such melts in quantities of millicuries per ton so that the tracer may be used to follow the melt from the mill to the finished product.[57] For such applications, gamma emitters are preferred to beta-particle emitters because the beta particles have low penetration and are easily absorbed. Scintillation counters can be used to detect the gamma rays in the tagged steel. In selecting a suitable

tracer, a radioisotope should be chosen that does not alter the chemical and physical properties of the melt. Preferably, the tracer should be an isotope of one of the constituents of the steel alloy. However, if this is not possible, the addition of a small quantity (such as 1 ppm) of an alien radioisotope is permissible without altering the desired properties of the alloy. It is important that the material added be mixed thoroughly with the metal and dispersed homogeneously. Tantalum-182 (half-life = 110 days, beta-particle energy = 0.53 Mev, and gamma energy = 1.1 and 1.2 Mev) has been found to be an ideal isotope for tagging steels. In tagging steel melts an isotope is required with a half-life of months, such as that of Ta^{185}. Moreover, Ta^{185} is available in high specific activities of about 50 curies/g.

The diffusion of elements through metals has a marked effect on the properties of metals, particularly at elevated temperatures where diffusion occurs more rapidly. Some examples are the case hardening of steels, hydrogen embrittlement, and failure at metal grain boundaries. The technique of autoradiography is very useful in studying these and other phenomena. Figure 6.12 shows an autoradiograph of the diffusion of Ni^{63} into polycrystalline gamma iron. This autoradiograph was prepared from Eastman permeable-base stripping film.[58] The diffusion period was 118 hr at 985°C and the magnification is 150.

In preparing the autoradiograph shown in Fig. 6.12, the metal surface was protected by dipping the metal sample in a solution of 2 per cent vinylite in methyl ethyl ketone and allowing the sample to dry in a vertical position. The coated sample was baked under an infrared heat lamp to harden the plastic coating. Similar studies have been reported of the diffusion of Ni^{63} into copper[59] at temperatures of 650 to 925°C. The extent of the grain-boundary diffusion was found to be a function of the grain-boundary angle and the diffusion temperature.[59]

Other uses of radioactive tracers in the heavy-metal industries have involved full-scale experiments on the rate of melting of ferrochrome in a 190-ton open hearth using Cr^{511} as a tracer, and on the rates of mass transfer in open hearth furnaces using Co^{60} and Cr^{51} as tracers. The mechanism of slag formation has been studied using Na^{24}, Ca^{45}, S^{35}, and P^{32}. The mechanism of crystallization and the distribution of impurities in 10-ton steel ingots and the speed of melting of steel scrap have been studied using radioactive tracers, and the nature and origin of slag inclusions in steel have been studied using Ca^{45}, S^{35}, and P^{32}. Radon has been used to study the flow of solids and gases in blast furnaces. Calcium-45 and P^{32} have been used for rapid analysis of steel, since, by using these radioisotopes, only one calibration analysis is required per melt. The dilution technique can also be used with a radioisotope to determine the weight of steel in a ladle. Radiotracers have been used in the steel industry to study the mechanisms involved in corrosion and in alloy formation to study the rate of hydrogen removal from steel samples.[56] Additional information on the use of radioisotopes in the metal industry is given in Refs. 60 to 72.

OIL INDUSTRY

Tracer techniques have been widely used in the petroleum industry in the study of mixing. Every blending operation requires proper proportioning and an optimum time must be given for the production of a uniform, homogeneous product. The mixing of grease has been studied[73] using I^{132}. This isotope is obtained from Te^{132}, which has a half-life of only 2.5 hr.

A knowledge of the degree of mixing is desired in scrubbing impurities out of products, in acid or alkaline treatment of petroleum stocks, in catalytic hydrogenation, in cracking and isomerization, and in determining catalysis loss in cracking units.[74,76] In the petroleum and other process industries, radiotracers are being used to study the effect of the various parameters, such as velocity, viscosity, and density of liquids, and the pipe size, on mixing rates.[75]

Mixing of two petroleum liquids (one of varying high viscosity and the other of low fixed viscosity) in a thermal cracking plant has been studied.[77] In this study Zr^{95} was used in a chelate compound and was injec ed in one of the lines feeding the tank. The outgoing stream was monitored to measure the activity of the mixed stock. A steady reading on the monitor indicated good mixing.

Barium-140 has been used in the study of the entrainment occurring in refinery vacuum flashers.[76] Barium-140 and Sb^{124} compounds have also been employed as tags for long slugs of different petroleum cuts which are pumped in series through pipelines.[75]

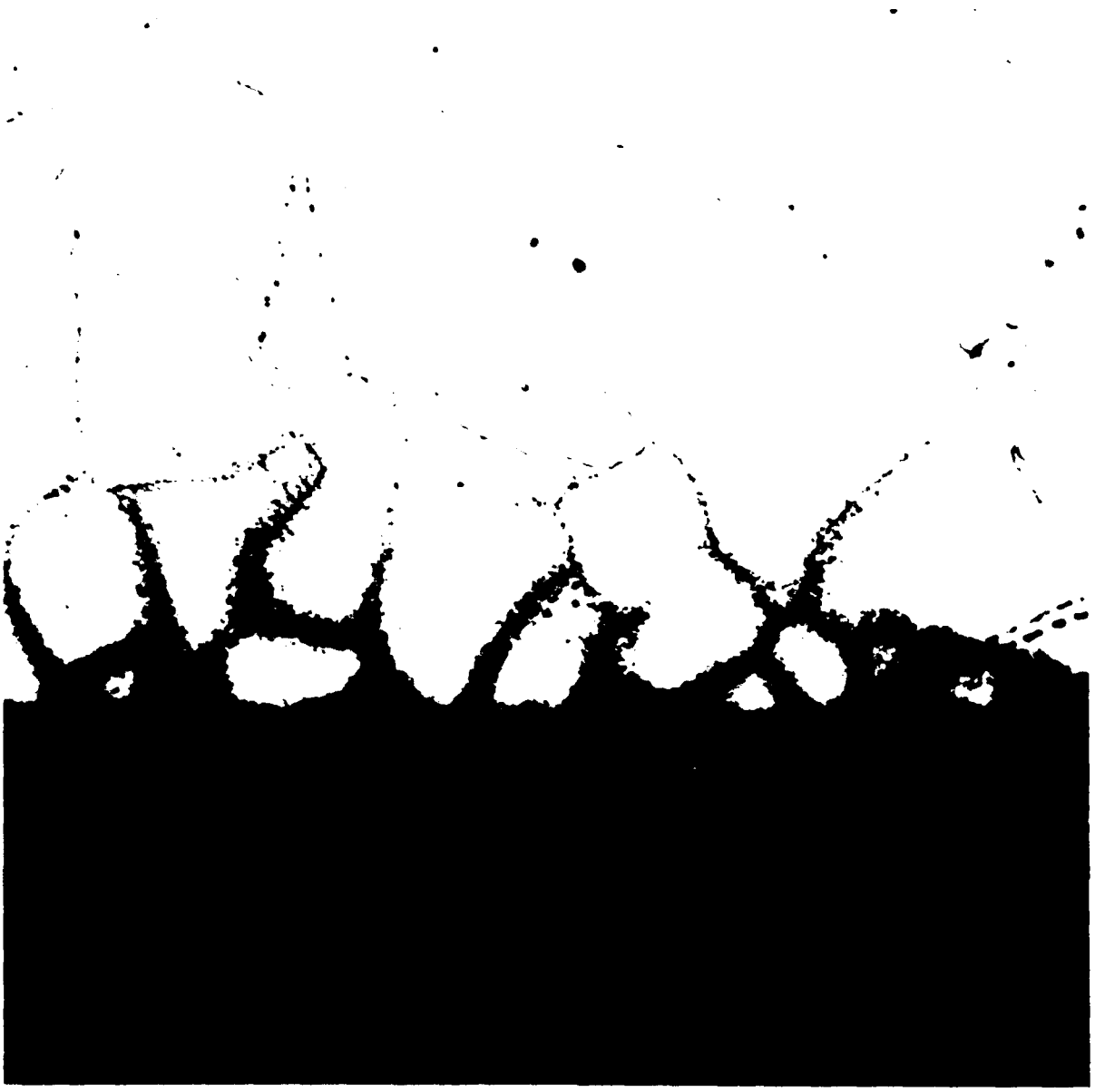


Fig. 6.12—Autoradiograph (150X) of the diffusion of Ni^{63} into gamma iron[42] after 118 hr at 950 C.[58]

This operation is important in pumping large amounts of petroleum crudes and various petroleum products, such as fuel oil, gasoline, kerosene, etc., in series for long distances through overland pipelines. The location of the interface between two products pumped in series must be known in order to route the different products to different take-off points and terminals along the line. It is important that the interface be located as exactly as possible to prevent mixing the two products, except for a small slug which can be pumped to storage and reseparated. By injecting a small amount of radioactive tracer into the line at the interface of crudes or products, the interface may be quickly located by counters placed along the line. Such counters may be used to initiate the operation of pumps and valves automatically so that the crudes or petroleum products may be properly routed and such undesirable events as the possible mixing of gasoline and fuel oil may be prevented.

The use of a radioactive tracer in a pipeline may also be used to measure the flow rate of fluids.

Three methods have been used with radioisotopes for flow measurements. The simplest is known as "peak" timing. Esso Standard Oil Company uses this method to determine the veloc-

ity of petroleum fractions in its pipelines.[78] Two radiation detectors are mounted on the pipe that is to carry the products. Since the internal diameter of the pipe is known as well as the distance between the detectors, the volume of the pipe between the detectors may be calculated. If a small amount of radioactive material is added to the hydrocarbon product, upstream from both detectors, it will eventually pass by each detector. As the radioactive material passes each detector it causes an increase in radiation or a "peak" on a recording device that is connected to the detector. Since the pipe volume is known, it is possible to determine the fluid flow rate from the relation

$$F = V/t \tag{6.6}$$

where F = flow rate
V = pipe volume
t = time interval between peaks

If a radioisotope is thoroughly mixed with a quantity of fluid so as to form a homogeneous mixture with respect to radiation emission the method of "integrated counts" may be used. An advantage of this method is that only one detector and one counter is required. However, a larger amount of radioactive material is required in this method as compared to the peak timing method. The flow rate is obtained by a calibration of integrated counts as related to the flow rate. Each time the concentration of the radioisotope is changed, it is necessary to recalibrate.

Another method of measuring flow rates is known as the "activation" method.[78] A system using this method is shown in Fig. 6.13.

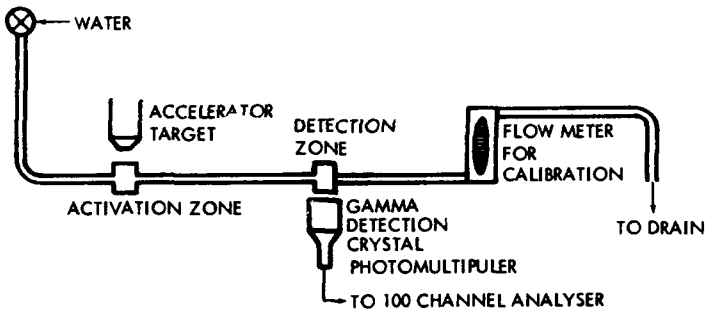


Fig. 6.13—Activation method flowmeter.[78]

This system has some distinct advantages over the two previously described. For example, it eliminates the need for adding of radioactive material to the system and the need for direct access to the flow system. Also, it allows the measurement to be independent of flow patterns if adequate mixing of active material can be assumed.

In the activation method, a neutron source is placed as close as possible to the fluid. The neutrons from the source interact with a small portion of the fluid causing some element present to become slightly radioactive. In other words, a radiotracer, is artificially formed in the fluid. A detector is placed downstream from the neutron source. The time for an amount of material to move from the neutron source to the detector is known as the transit time from which the fluid velocity can be calculated.

An important factor is the activation time which is the time that the fluid is near the neutron sources. If the transit and activation times are short when compared with the half-life of the radioisotope that is formed by activation, a plot of count rate vs. flow rate will have the shape shown in Fig. 6.14.

Assuming no radioactive decay Eq. 6.7 may be derived for the flow rate[78]

$$F = g\phi\lambda V/A \tag{6.7}$$

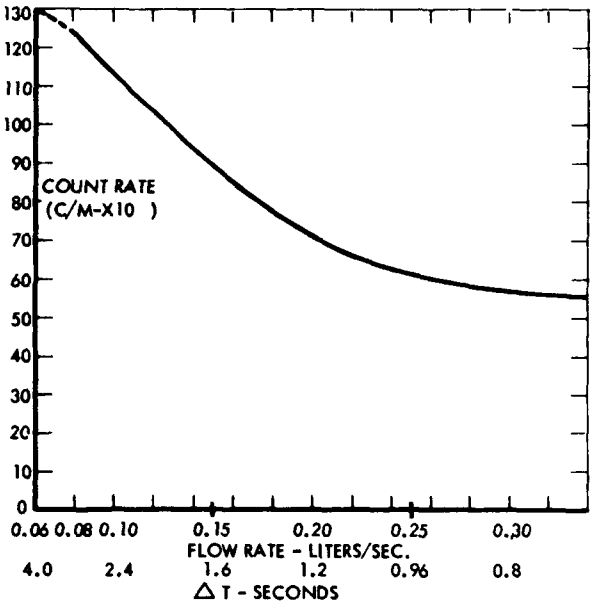


Fig. 6.14—Activity vs. flow rate for short transit times.[78]

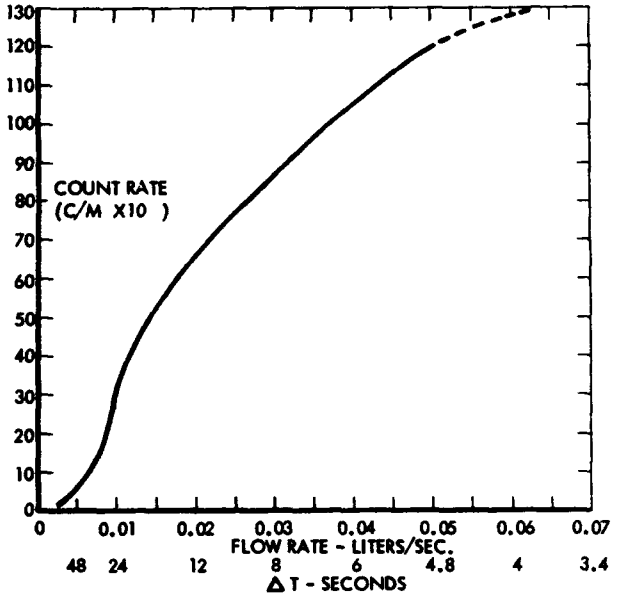


Fig. 6.15—Activity vs. flow rate for long transit times.[78]

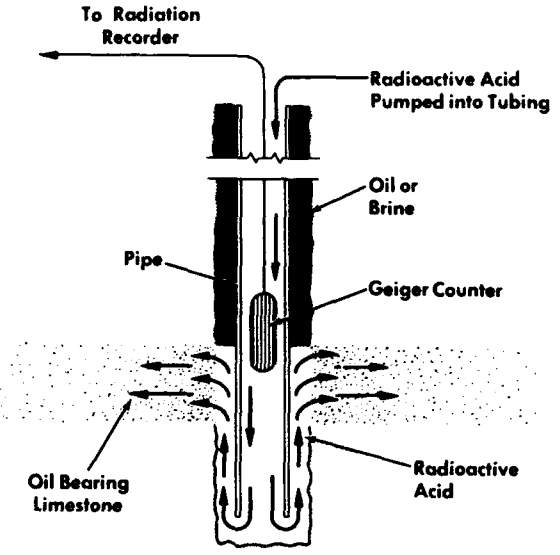


Fig. 6.16—Use of radioisotopes for control of oil-well acidizing.

where F = fluid flow rate
g = fraction of neutrons effective in creating active atoms
ϕ = neutron source emission rate
λ = decay constant of activated atoms
V = sample volume
A = activity in the sample volume

However, if the transit time or activation time is significant when compared with the half-life of the isotope, Eq. 6.7 is no longer valid. Under these conditions, a plot of count rate vs. flow rate is represented by Fig. 6.15.

The transit time for the case shown in Fig. 6.15 may be determined[78] from

$$T = (1/\lambda) \ln (A_0/A) \tag{6.8}$$

where T = transit time
A_0 = activity of the material leaving irradiation zone
A = activity of the material at the detector

Since the distance between the activation zone and the detector is known, the fluid velocity is[78]

$$V = D\lambda/\ln (A_0/A) \tag{6.9}$$

where V = fluid velocity
D = distance between activation zone and detector

It may be noted that A_0 is the saturation activity (or very nearly so) and thus can be calculated rather than measured.

In a cracking plant the catalyst, in the form of small porous beads, passes slowly down through the gasoline cracker where it assists in converting petroleum hydrocarbons to gasoline. The high temperature and dehydrogenation occurring in the cracker results in an accumulation of carbon on the catalyst beads, which must be removed to regenerate the catalyst. This is accomplished in a continuous operation by removing catalyst beads from the bottom of the cracker, burning off the carbon in a furnace, and returning the regenerated catalyst to the top of the cracker. The efficiency of the cracking operation depends upon the rate of catalyst circulation, which can be measured by tracer technique. In this method a few of the catalyst beads are made radioactive by impregnation with radiozirconium. Counters placed in the circulating system can be used to measure the time required for a radioactive bead to travel from one counter to the other, thus permitting calculation of the catalyst flow rate.

Another important application of radioactive tracers in the petroleum industry is in oil-well "acidizing." Acid has been found useful in increasing the porosity of certain oil-bearing strata so as to increase the output of oil. In previous techniques it was often necessary to remove the pipe used to introduce the acid. Sometimes, as a result of excessive acidification, several thousand feet of 2-in. pipe sections had to be removed; an expensive operation. The tracer technique is illustrated in Fig. 6.16 and is much more rapid, less hazardous, and less expensive than removing the pipe.

Referring to Fig. 6.16, the technique consists in suspending a radiation detector in the pipe at the desired depth for acid treatment. A radioisotope tracer is added to the acid being used. As the acid passes the counter a reading is obtained, but a much higher reading is produced when the labeled acid fills the well to the height of the detector, which is hung at the level to which the acid must rise in the strata. Application of pressure forces the acid into the strata, and the continual addition of acid maintains the acid level at the proper depth. This operation can be judged readily and controlled by the use of the detector.

Since the consumer demand for heating oils is heaviest during the winter months, the oil refineries are faced with the problem of storing large quantities of heating oils that are produced during the summer. It is not always economically feasible to build sufficient storage tanks for such quantities of oil. The Esso Standard Oil Company solved this problem by using an abandoned slate quarry as a reservoir.[56] However, before this was done it was necessary to determine whether the quarry would hold the oil without allowing significant leakage. An experimental batch of oil was tagged with radioactive hydrogen and placed in the quarry. After a period of time the area around the quarry was monitored for radioactivity and the survey indicated that the quarry could be used.

In other applications in the petroleum industry, Co^{60} and I^{131} have been used in studying water flooding in oilfield work.[79] Tritium (H^3) and Kr^{85} have been found of great use in oilfield gas injection.[80] Krypton-85 has also been used in studies of air mixing in fluid catalytic cracking unit regenerators.[81]

Additional information on the use of radioactive tracers in the petroleum industry is given in Refs. 81 to 90.

OTHER INDUSTRIES

Tracer techniques have been used for a variety of purposes in many industries. Some of these other applications will be discussed briefly.

In the road construction industry, asphalt has been used very successfully for paving. However, there are many types of asphalts with widely different properties. Often the resins in asphalt are removed to be used in producing plastics, but if such removal is excessive the asphalt may lose its binding strength and be unsatisfactory for road construction or repair. If the asphalt is not properly diluted with solvent or applied at the proper temperature or if mixing with the aggregate is incomplete, again poor bonding may result. The efficiency of bonding may be checked by using Ca^{45} in calcium chloride. A trace amount of radioactive calcium chloride is mixed with some plain calcium chloride and added to the aggregate; then the asphalt is applied, and if the mixing and bonding are adequate and impervious, the calcium chloride will be completely entrapped by the asphalt. If the bond is incomplete or broken, water applied to the pavement will wash out some of the radioactive calcium chloride, and this can be measured with a suitable detector.[56]

Another application of tracers is in determining the efficiency of a separation such as evaporation or distillation. In distilling sea water to produce boiler feed water for the steam boilers on a ship, it is important to minimize the entrainment carried over to the condensate by the vapor. The use of the dilution technique and a radioactive tracer, as indicated in Fig. 6.17, permits a rapid determination of the amount of entrainment.

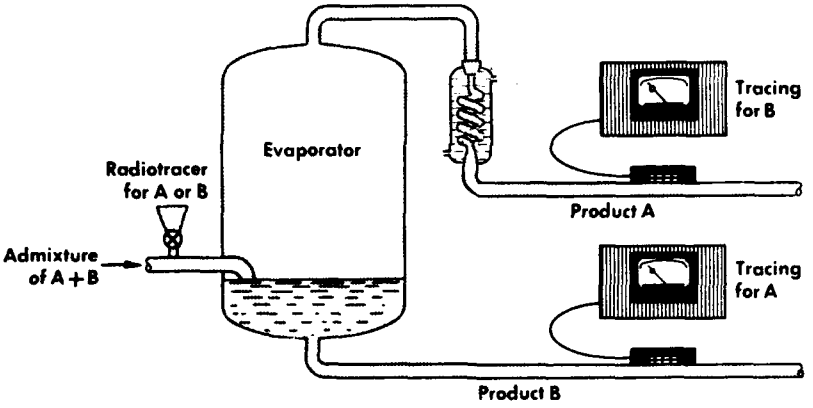


Fig. 6.17—Use of radioactive tracers to measure efficiency of separation in a conventional evaporator.

Another industrial application of radioactive tracers is the determination of the efficiency of household washers or detergents; P^{32} is sometimes used as the tracer. Swatches of cloth are contaminated with the labeled bacteria or dirt and then washed. The radioactivity of the swatches is determined and this gives a rapid indication of the amount of bacteria or dirt remaining. Other radioisotopes, such as Ca^{45}, may be used. Additional information on the use of tracers in washing and cleaning studies is given in Refs. 91 to 96.

Several important problems having to do with turbines have been solved using radiological techniques. In one case, it was found that certain minerals were collecting on the blades of a set of steam turbines. As the mineral deposit increased, the delicate turbine balance was upset. In order to correct this it was necessary to clean the turbine blades each year. The accumulation was caused by the carry-over of minerals in the droplets entrained in the steam from the boilers. To determine the amount of carry-over as a function of boiler load, several curies of Na^{24} were placed in the boiler. It was assumed that the amount of Na^{24} carried over was proportional to the amount of minerals. The results of these tests showed that by reducing

the boiler load to half, it would be necessary to clean the turbine blades only once every six years instead of once a year. Since a spare boiler was available this did not result in a decrease in turbine output, yet allowed a savings of about $80,000 per year.[78]

It is desirable to measure the clearance between the stationary nozzles and the rotor buckets of a turbine. Because of thermal expansion of the stator and the rotor the clearance is a function of the turbine temperature. It is difficult to make this measurement by conventional methods because of the high temperatures, inaccessability of the turbine interior during operation, and the physical restrictions of the rotor. However, it was possible to measure the clearance by the use of an instrument called the radiological position monitor.[78] A diagram of the technique is shown in Fig. 6.18. A 10 μc Co^{60} source was mounted on the stator casing perpendicular to the turbine axis. Another source was produced on the tip of one of the turbine buckets by neutron bombardment. This source also was located perpendicular to the turbine axis. A scintillation counter was installed adjacent to the turbine. The detector was connected to a mechanical drive that moved the detector along the outside of the turbine. The scanning distance included the locations of the radioactive sources. Each time the detector passed over a source a peak was indicated on a recorder. The recorder also showed the relative position of the detector. Thus, by measuring the distance between the peaks it was possible to determine the clearance between the nozzle and bucket. This system gave results that were accurate to within 0.015 in.

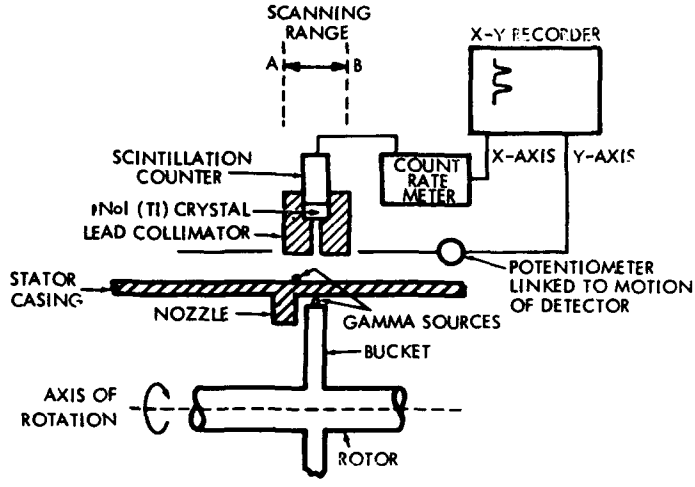


Fig. 6.18—Turbine axial position monitor.[78]

Another very useful industrial application of radioisotopes is to detect leaks in underground pipes. Probably the most satisfactory radioisotope that can be used in water mains is Na^{24}. The use of this radioisotope is completely safe as the concentration required for the detection of leaks is less than the drinking tolerance. Furthermore, the radioisotope has a short half-life (15 hr) and can be retained in the pipe lines until the activity has been greatly reduced as a result of decay. Both the short half-life and the rapid turnover of sodium in the body tend to place this radioisotope in the "more safe" category.

In one British method of leak detection, the section of the main under suspicion is isolated, filled with Na^{24} bicarbonate solution and pressure is applied. The line is drained and flushed with fresh water. A gamma-radiation detector can be used to determine the location of the leaks from presence of gamma radiation emitted from the radioactive sodium bicarbonate solution that has accumulated around leaks in the pipe.

A British modification of this technique involves the use of a "go-devil" which has a built-in gamma detector and wire recorder. In locating the leak, the go-devil is inserted in the line and moved through a section under suspicion for leakage. Leather pistons on the go-devil seal the walls of the pipe so that the go-devil moves with the fluid that is forced through the pipe. Small Co^{60} sources may be placed at regular intervals along the pipe to orient the recorder

readings. After the unit is recovered, the recording is played back to give a plot of leak positions. Such units have been used over distances as long as 12 miles, with the recording capacity as the only limitation on the length of travel.

Radioisotopes other than Na^{24}, such as I^{134} and C^{14}, also have been used to detect leaks in underground pipes. The use of either I^{131} or Na^{24} with surface detectors is limited to pipes only a short distance below the surface. If several feet of earth or other over-burden cover the pipes, most of the gamma radiation will be absorbed, making the method insensitive. In the case of deeply buried pipes samples of radioactive gas such as $C^{14}O$ may be collected, or a go-devil may be used.

Radiotracers have been used in the food industry to solve particular problems. In one application C^{14} was used to determine the location of grease in fried doughnuts.[56] Also the paper industry has found that radiotracers are useful in studying the movement of water vapor through paper and in determining the penetration of oil into porous paper used in high voltage applications.[58]

RESEARCH

Tracer analysis has been employed in a wide range of fields for solving research problems. In all the branches of pure and applied science, tagged atoms are being used to obtain quantitative and qualitative information. Studies of chemical reactions, rates of diffusion of metals, vulcanization of rubber, and water analysis are some selected examples of fields in which the radioactive isotopes have been used to advantage. Some of the most useful radioisotopes in such research are C^{14}, S^{35}, and H^3, an isotope of hydrogen. Selected research studies are mentioned in the following sections.

CHEMICAL REACTIONS

Tracers have been used in the study of chemical reactions in the fields of inorganic and organic chemistry and biochemistry. Before the availability of radioactive tracers, the mechanism of reactions often could not be determined. The use of tracers has provided a new tool for such studies. Calvin[97] studied the path of carbon in photosynthesis, starting with $C^{14}O_2$. Exchange reactions of halogens with completely halogenated methanes have been studied. A specific example of this is the exchange of bromine between Br_2 and CCl_3Br.[98,99] Radioiodine has been used by Gevantman and Williams[100] to identify the radicals produced in the radiolysis of four alkyl iodides and of six liquid hydrocarbons. The mechanism of chemical reactions produced by neutron irradiation has been studied. Sulfur-35 has been used by Smith[101] and M and Peterson[102] in the study of the polymerization.

Intermolecular exchange reactions between Br_2 and HBr (Ref. 103), $SnCl_4$ and HCl (Ref. 104), and HF and ClF_3 (Ref. 105) have been studied extensively. The chemical properties of S^{35}, produced by Cl^{35} absorbing a neutron and then giving off a proton, have been investigated by irradiating KCl crystals with neutrons. Studies of surfaces covered with only 10^{-7} g/cm^2 of a chemical substance have been undertaken to investigate the nature of surface forces and the kinetics of reactions involving surface atoms and exchange reactions.[106]

Bonner and Collins[107] have studied the molecular rearrangements of the derivatives of triphenyl ethyl using C^{14}. The cold-flame oxidation of butane at a temperature of 285°C has been studied[108] by the kinetic method employing C^{14}. A study of the thermal oxidation of methane, carried out with O^{18}, demonstrated that at high temperatures the principal products of this reaction are formaldehyde, carbon monoxide, and carbon dioxide.[108] Using Cl^{36}, several exchange reactions of the Friedel-Crafts type have been studied, and it has been found that gaseous HCl and a variety of chlorinated hydrocarbons exchange readily[109-112] with solid $AlCl_3$.

The mechanism of the Fischer-Tropsch synthesis[113] has been studied to determine whether a metallic carbide is an intermediate product in the synthesis of hydrocarbons from carbon monoxide and hydrogen over iron, nickel, and cobalt catalysts.[114] Using radioactive carbon dioxide, carbon monoxide, and water, the poisoning phenomenon on solid catalysts has been studied.[115]

In the study of the heterogeneous catalytic reactions, the radioisotopes H^3, C^{14}, S^{36}, and P^{32} have been widely used. A number of stable isotopes, C^{13}, N^{15}, and O^{18}, have also been employed

and analyzed by infrared absorption or mass-spectrometry methods.[116,117] Proton and deuteron resonance methods have also been used.[118] Turkevich and Smith[119] used tritium-containing materials to study the migration of the double bond in the butene-1–butene-2 system and the tritium exchange between the catalyst and hydrocarbons. Radiocarbon has been used by Turkevich and Bonner[120] to study the reduction of small amounts of radioactive carbon dioxide on a large surface area of charcoal. The stable isotope deuterium has been employed in the study of the addition of hydrogen to a carbon-carbon double bond.[121]

WATER ANALYSIS

Radiometric techniques have been employed to determine the concentration of chlorides and sulfates in drinking water. In the case of chloride detection, radiosilver has been added in silver nitrate solution. Silver chloride is precipitated out according to the reaction:

$$XCl + AgNO_3 \rightarrow XNO_3 + AgCl \qquad (6.10)$$

The end point is determined by radiometric analysis[122] as indicated in Fig. 6.19 which shows the activity of the filtrate vs. the amount of radiosilver added. The end point is the break in the curve.

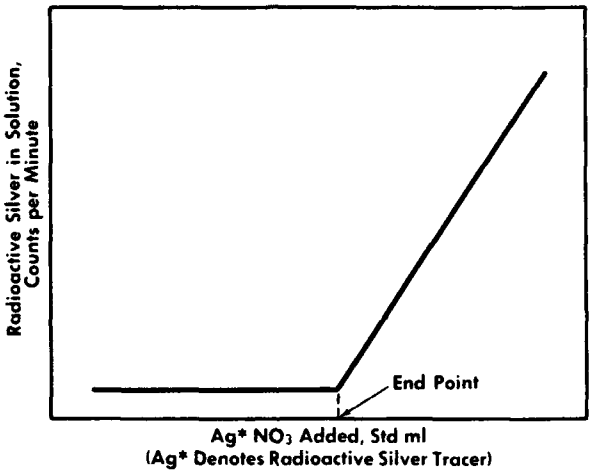


Fig. 6.19—Typical curve in chloride analysis using radiosilver.[122]

In sulfate analysis barium chloride is precipitated as barium sulfate. A small amount of radiosulfur may be added to determine this end point. The chemical reaction proceeds as follows:

$$XSO_4 + BaCl_2 \rightarrow XCl_2 + BaSO_4 \qquad (6.11)$$

Tests were made of the water of the Mohawk River, using P^{32} to assess the health hazards resulting from the accidental spillage of radioactive wastes[123] and to determine the amount of low-level waste that could be safely discharged into the river. In Germany, measurements involving the rate of water flow, hold-up time of sewage in settling tanks, and other sanitary problems have been carried out in the Ruhr basin using tracer techniques.[124]

DIFFUSION

Research studies on diffusion and diffusion rates are ideally suited for the use of tracer techniques. Diffusion involves the migration of atoms or molecules of one species into or through groups of atoms or molecules of another species. The rate of diffusion is a function of the "diffusivity" of the atoms in the system being considered and is an important consideration

in mass transfer, heat transfer, and related fields. In order to determine the rate of diffusion or the diffusivity, the atoms or molecules must be traced during their migration in the diffusion process. There is no better means for such tracing than the use of radioisotopes.

O'Hern and Martin reported on the diffusion of carbon dioxide at elevated pressures, using C^{14} as a tracer.[125] The literature contains little information on the diffusion of gases at high pressures. The range covered in this study included determination of diffusion coefficients in the $C^{14}O_2$–$C^{12}O_2$ system at temperatures from 0 to 100°C and at pressures up to 200 atm. The results indicated[125] that the diffusion coefficient varied approximately inversely with the gas density. A two-chambered diffusion cell was used in this study consisting of two ionization chambers connected by a diffusion opening, as indicated in Fig. 6.20. Such apparatus permits the use of both chambers as detectors. The presence of radiocarbon in either chamber will result in ionization of the gas present and a collection of electrons at the anode to produce an ionization current. The ionization current is proportional to the concentration of radioisotope and ranged from 10^{-14} to 10^{-11} amp. This small current was accurately measured by means of a sensitive instrument (Bechman Ultrohmeter) to determine the relative concentrations of C^{14} in either chamber.

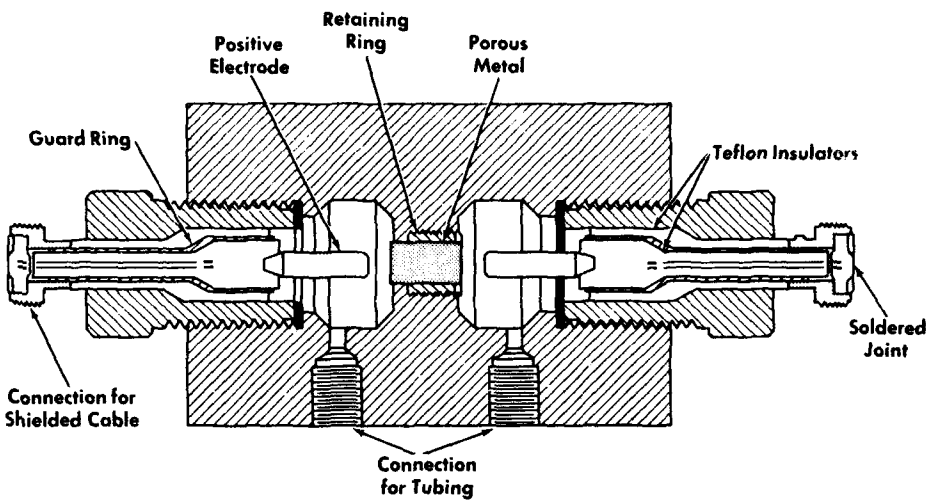


Fig. 6.20—Diffusion cell with two ionization chambers used to measure the diffusion of $C^{14}O_2$ at elevated pressures.[125]

In the rubber industry, the problems of solubility and diffusivity of sulfur in rubber have been studied by Auerbach and Gehman[126] using S^{35}. In these studies specimens of rubber in the form of small disks were prepared and a layer of sulfur powder containing S^{35} spread on the underside of the disk. Each specimen was then kept at a constant temperature and the activity of the top surface of the specimen as a function of time was recorded, as shown in Fig. 6.21. The equilibrium activity obtained at a particular temperature for a given concentration of sulfur is used to determine the solubility. A typical calibration curve of the concentration of sulfur in rubber (1 g sulfur per 100 g of GR-S) and the equilibrium activity (counts per minute) is given in Fig. 6.22. Such a curve was utilized to determine the solubility at a given temperature. The variation of solubility with temperature has also been studied by repeating the experiment at different temperatures.[126]

Diffusivity of sulfur in rubber was determined from the observations recorded in the solubility experiment. The diffusion equation with the necessary boundary conditions was applied to give the solution

$$\ln \frac{c_s - c}{c_s} \approx -\frac{D\pi^2 t}{4^2} + \ln \frac{4}{\pi} \qquad (6.12)$$

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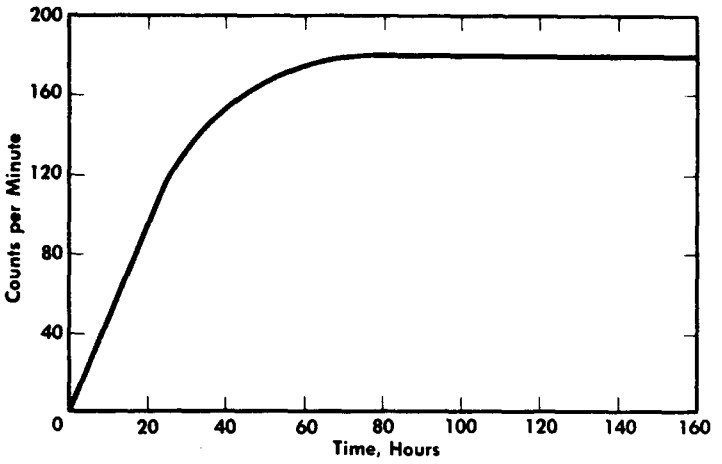


Fig. 6.21—Typical plot of activity of rubber samples vs. time in study of diffusion of S^{35} through rubber.[126]

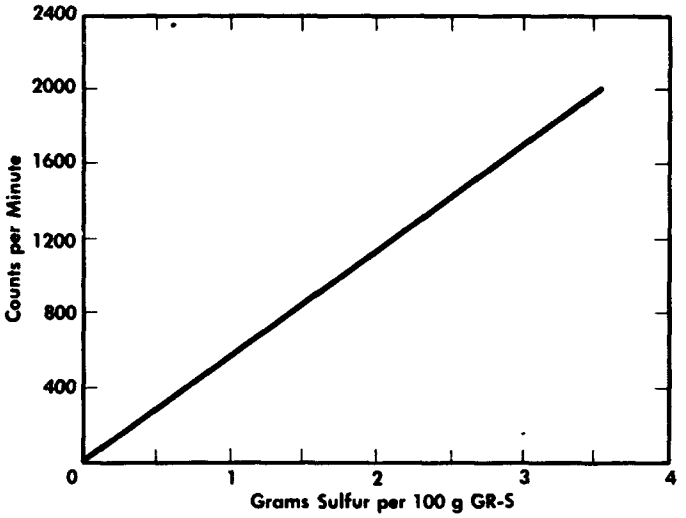


Fig. 6.22—Typical calibration curve for converting activity to sulfur solubility in study of diffusion of S^{35} through rubber.[126]

where c_s = concentration for saturation
c = concentration at distance, d, at time, t
d = thickness of the rubber specimen
D = diffusivity constant

Equation 6.12 is linear in time and the slope of the straight line gives diffusivity constant, D.

The diffusion of iron, silver, and cobalt has been studied in a number of alloys.[127,128] The self-diffusion coefficient D, was found to be related to the activation energy of diffusion according to the equation

$$D = D_0 e^{-Q/RT} \tag{6.13}$$

where Q = activation energy
T = absolute temperature

The influence of the addition of a substance of small atomic mass (like carbon) on the mobility of the atoms and energy of activation has been studied. The diffusion studies have confirmed earlier results on the nature of interatomic binding in the metallic phase which were obtained from X-ray studies.

Many diffusion studies have been made with materials in the solid state by the technique of autoradiography. Additional information on the use of radioisotopes in studying diffusion is given in Refs. 129 to 142.

VULCANIZATION

The function of mercaptobenzothiazole in the vulcanization process has been investigated[143] using radioactive S^{35} and mercaptobenzothiazole tagged with S^{35}. The effects of varying the proportions of mercaptobenzothiazole, sulfur, and zinc oxide on the combining rates of sulfur were determined. It was found that during the process of vulcanization, the ratio of sulfur combined to mercaptobenzothiazole consumed is constant.

AGRICULTURE

Tracer methods of analysis and research would appear to be made to order for studies in the biological fields. The use of tracer techniques has resulted in remarkable progress in the fields of medicine and agriculture. In agriculture alone, there has been a vast number of investigations performed with the aid of tracer techniques. Some of these could never have been performed using conventional methods. Selected investigations will be briefly described and a few applications will be discussed in some detail.

One useful application of radiotracers in the field of agriculture is the determination of the dispersal of certain insect populations. It was found that by tagging insects such as mosquitoes, locusts, grasshoppers, and blackflies with P^{32} or Co^{60} an excellent determination of dispersion rates was possible. Methods used in tagging insects depend upon the nature of the experiment and the type of insect to be tagged. The techniques developed for insect tagging have also been used successfully in measuring the dispersion of pollen grains by wind as well as by insect activity.

It is known that the root plays an important part in the life of higher plants. But because roots are beneath the soil it has been difficult to make studies of the functions of roots in obtaining and transmitting nutrients to other portions of the plant. However, by mixing radioactive elements with the soil it has been possible to determine the amounts of various elements that are absorbed by the roots.[144] It has also been possible to map the soil locations from which most of the plant food is obtained.

Radioisotopes also have been used to measure certain physical properties of soils such as moisture content, density, and gas diffusion rates. A spectrum of uses of radioisotopes in agriculture[145] are listed in Table 6.1.

Table 6.1 — APPLICATIONS OF TRACERS TO AGRICULTURE[145]

I. Isotopes in plant and soil research
   1. Mineral plant nutrition
      a. Ion intake
      b. Metabolism
      c. Minor elements
      d. Foliar nutrition
   2. Growth regulators, thinning hormones, and insecticides
      a. Growth regulators
      b. Thinning hormones
      c. Insecticides
   3. Nitrogen and organic matter in soil
      a. Nitrogen fixation
      b. Decomposition of organic matter
   4. Soil fertility studies
   5. Miscellaneous
      a. Root grafting
      b. Spore labeling

II. Isotopes in animal research
   1. Nutrition experiments
      a. Digestion
      b. Phosphorus utilization
      c. Phosphatides synthesis
      d. Iron, calcium, and phytate absorption
      e. Detection of estrogen residues
      f. Coprophogy studies
   2. Intermediary metabolism
   3. Basic physiology
      a. Endocrinological studies
      b. Body-water content determinations
      c. Ruminology
   4. Veterinary medicine
      a. Diagnosis and therapy
      b. Pathogenesis of diseases
      c. Pharmacology
      d. Medical microbiology
      e. Immunology
   5. Entemology
      a. Metabolic studies with insect nutrients
      b. Insecticide research
      c. Tagging insects
   6. Microbiology

PHOTOSYNTHESIS

Photosynthesis is the basic chemical process by which plants synthesize, from simple inorganic chemicals, the many and varied complex organic compounds used in their growth.

The study of photosynthesis began with the work of Stephan Hales (1677-1761) and still continues. Although it has been studied for over 200 years much is still unknown about photosynthesis, and most of what is presently known has been discovered within the last 25 years. The reason is the advent of new and better methods of investigation of the nature of the processes involved in photosynthesis. Two of the most useful methods to date have been tracer techniques and paper chromatography.

In general, the process of photosynthesis may be summarized by the chemical equation

$$6CO_2 + 12H_2O \xrightarrow[\text{Plants}]{\text{Sunlight}} C_6H_{12}O_6 + 6O_2 + 6H_2O \qquad (6.14)$$

The reverse of this chemical equation is referred to as respiration and for a long time respiration was thought to be the exact reverse of photosynthesis. Detailed investigations have shown that although the initial products of one process may be the final products of the other, the intermediate products may be quite different.[146]

According to the present thinking the photosynthetic process may be divided into three distinct parts.

1. The absorption of light energy by the pigments.
2. The photolysis of water to produce stable biochemical energy and oxygen.
3. The reduction of carbon dioxide to a carbohydrate.

The details of reactions in parts 1 and 2 have not been determined with certainty. In part 1, it is believed that light energy absorbed in the plant, raises the energy state of the chlorophyll. The activated chlorophyll then interacts with energy-carrying molecules so as to form a chemically stable complex.

The photolysis of water is believed to follow the scheme

$$B + H_2O + YX \rightarrow A + HX + YOH \tag{6.15}$$

In Eq. 6.15, B is the previously mentioned chemically-stable complex that was formed from the chemical A. The identities of the naturally-occurring groups X and Y remain unknown, but the photolysis products HX and YOH are believed to undergo decomposition reactions of the form

$$HX \rightarrow H + X \tag{6.16}$$

$$2YOH \rightarrow \tfrac{1}{2}O_2 + H_2O + Y \tag{6.17}$$

In reaction 6.16 the term H represents a biochemical reducing agent. It is not presently known what chemical intermediate possesses this property but H in some way subsequently causes the reduction of CO_2. This reaction is not believed to be accomplished directly because H apparently is involved in certain reactions which form the reducing enzymes, triphospyridine nucleotide (TPNH) and adenosine triphosphate (ATP).

The mechanisms involved in the reduction of CO_2 have been fairly well established but are rather complex and will not be discussed. The proposed cycle is shown in Fig. 6.23. Much of the information shown in Fig. 6.23 was obtained through the use of radiotracers.[148]

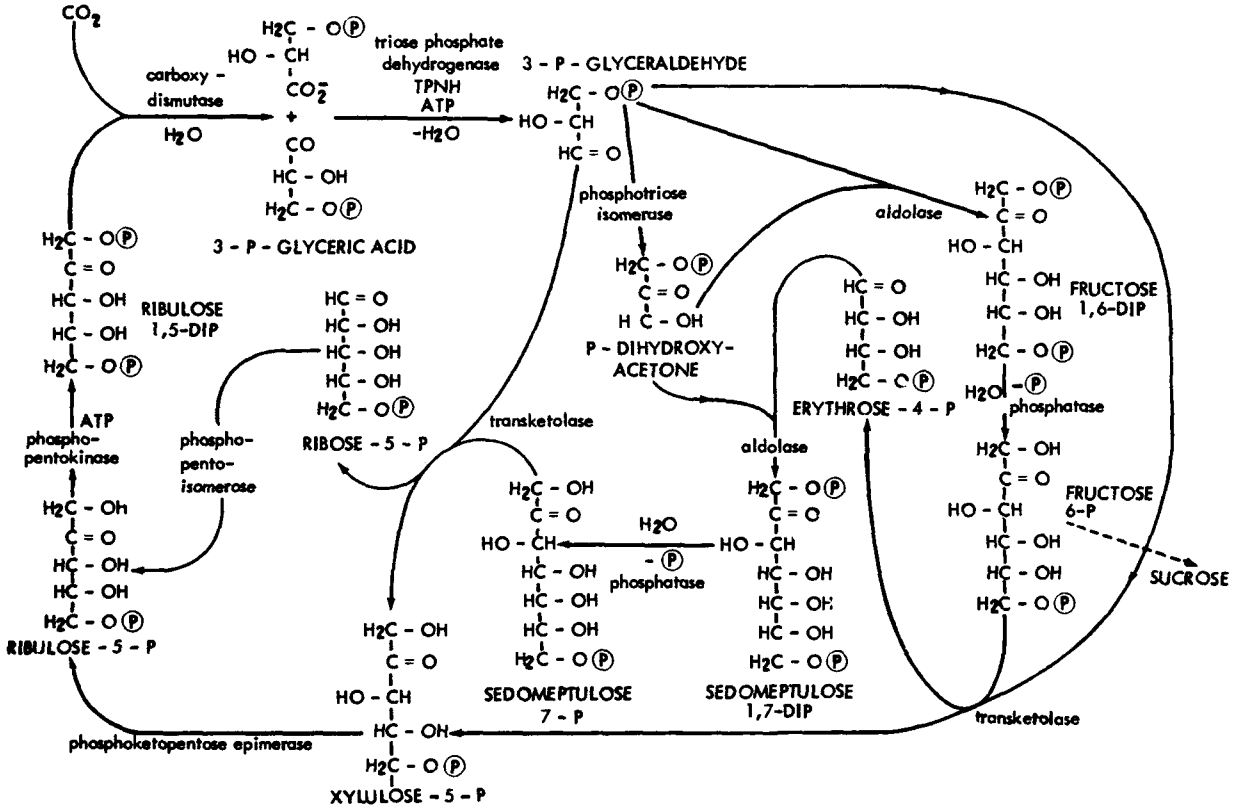


Fig. 6.23—Complete photosynthetic carbon cycle.[148]

Tracer techniques have answered many questions concerning the photosynthetic process. One such question was whether the photosynthetic process fixes both oxygen and carbon dioxide. In 1940, studies performed with O^{18} showed that the oxygen released from any plant was formed entirely from water rather than from carbon dioxide. Therefore, photosynthesis must

fix both carbon and oxygen.[146] Early research indicated that starch in the chloroplasts was formed according to the sequence

$$\text{Hexose sugars} \rightarrow \text{sucrose} \rightarrow \text{starch} \tag{6.18}$$

Equation 6.18 indicates that hexose sugars are the first products of photosynthesis. Use of tracer techniques with C^{14} showed conclusively that this was not the case. It is now believed that phosphoglyceric acid (PGA) is the primary product of photosynthesis.

For a long time, it had been assumed that all of the carbon dioxide involved in photosynthesis was assimilated through the plant leaves. Recent work has shown that as much as 12 per cent of the carbon dioxide may enter the plant through the roots. These experiments were performed using CO_2 tagged with C^{14}. Additional information concerning studies of photosynthesis may be found in Refs. 145 to 148.

PLANT NUTRIENTS

The role of mineral nutrients in the soil has been extensively studied by tracer techniques. It has long been known that plants require appreciable quantities of phosphorus, nitrogen, potassium, and trace amounts of other elements for normal growth. If these materials are not present in sufficient quantities in the soil, mineral or organic fertilizers must be added to assure good crop yields.

In the past the requirements for soil fertilization have been judged largely from crop yields. However, other variables such as rainfall, temperature, and disease complicate such procedures. By tracer methods the capacity of the soil to supply a plant nutrient can be determined by adding a labeled fertilizer to the soil as a partial source of the nutrient. The plant will draw upon both the natural supply and tagged additive and the uptake will be proportional to the availability of each. The method of dilution analysis may then be used to measure the amount and determine the percentage of the tagged fertilizer in the plant. From this information the availability of a nutrient such as phosphorus may be determined.

In the past it was believed that fertilizers were absorbed by the roots and that the leaves were used only for the release of water vapor and exchange of oxygen and carbon dioxide gases. However, tracer studies have demonstrated recently that fertilizers can be applied to the leaves, stems, flowers, and fruits of the plant where in many cases the absorption is more rapid with less loss of the fertilizer than if the same amount had been applied to the soil. Some fertilizers such as salts containing potassium and nitrogen are quite soluble. If applied to the soil, these materials may be dissolved and washed away by rainfall before a major portion is absorbed by the roots. The more rapid foliar absorption, plus absorption by the roots of fertilizer washed from the leaves, may result in much more efficient utilization of fertilizers applied to the foilage as compared to the soil. Furthermore, many crops such as fruits, potatoes, and truck crops are sprayed for control of insects and disease. Such applications of insecticides and/or herbicides may also be used for simultaneous fertilization. Figure 6.24 shows the rates of uptake of P^{32} in foliar applications and in root applications.[149] Additional information on such studies is given in Refs. 149 to 154.

In another study the uptake of calcium in the peanut plant was investigated. It was found that calcium was absorbed by both the fruit and the root but that absorption by the root alone was not sufficient for fruit development.[146] Other studies with Ca^{45} showed that although some highly mobile elements, such as nitrogen and phosphorus, are readily transported, both from the base to the apex (acropetally) and from the apex to the base (basipetally)[152,154] there is no basipetal movement of calcium.[149] This absence of basipetal transport of radiocalcium is depicted by the autoradiogram of the strawberry plant shown in Fig. 6.25. In the plants shown in this figure, Ca^{45} was applied to the roots of the mother plant and transport followed throughout the parent plant. Lateral movement of radiocalcium then followed through the stolens (runners) and aerial parts of the daughter plants. However, there was almost complete absence of basipetal transport into the roots of the daughter plant, indicating that roots of runner plants must depend upon their own absorption for a source of calcium.[149]

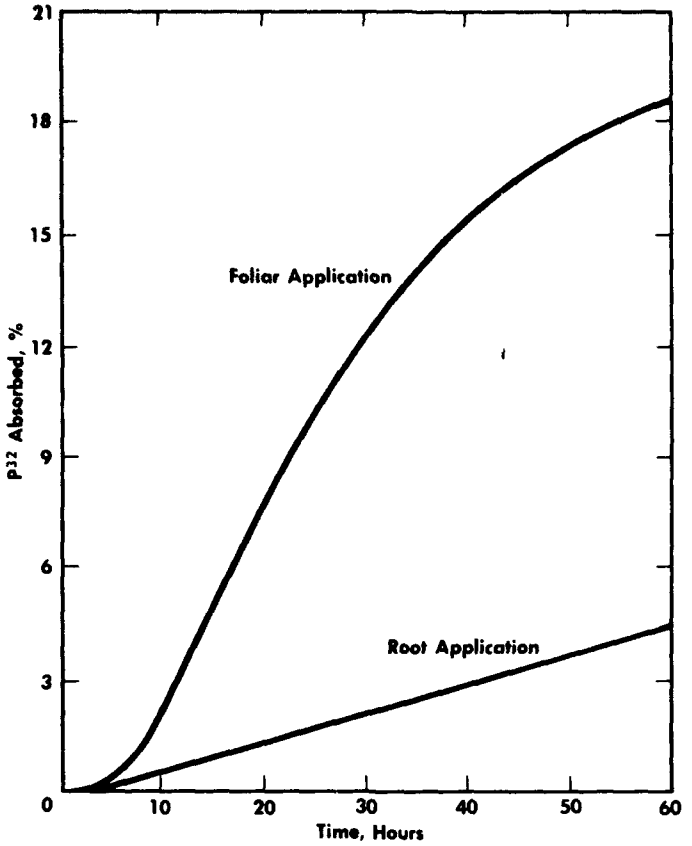


Fig. 6.24—Comparison of rate of P^{32} uptake from foliar and root applications.[149]

Rates of hydrolysis of urea from foliar applications have been determined using radiocarbon in various plants.[150] This study revealed the amount of nitrogen from urea used by the different plants. It was also found that urea (tagged with radiocarbon) reacts with water to give ammonia and carbon dioxide according to the reaction

$$\begin{matrix} NH_2 \\ NH_2 \end{matrix}\!\!>\!C{=}O + H_2O \rightarrow 2NH_3 + CO_2 \qquad (6.19)$$

(urea) + (water) → (ammonia) + carbon dioxide

The ammonia is absorbed by the plants, while the radioactive carbon dioxide is given off as a gas. The activity of carbon dioxide can be utilized to determine the absolute absorption of nitrogen from the urea.[150,155] A carbon dioxide absorber of porous material (ascarite tower) removes all carbon dioxide from the air returned to the plant.

ANIMAL HUSBANDRY

Radioactive tracers have been employed with great success in the field of animal husbandry. Calcium metabolism has been extensively studied using radiocalcium. The rates of digestion of feed calcium, rate of exchange of calcium between bone and blood, and the rate of formation of milk calcium from the calcium in blood were determined using the labeled calcium. In order to study the formation of milk substances within the udder, the artificial cow shown in Fig. 6.26, has been used in Minnesota, Maryland, and Wisconsin.[156] Formation of milk sugar by using the radioactive glucose and milk fat has been studied. Besides the study of the process of digestion, reproduction, and milk secretion, the tracers have been employed in the study of diseases of cattle such as ketosis.



Fig. 6.25—Autoradiograph showing distribution of Ca^{45} in a series of strawberry runner plants.[149]

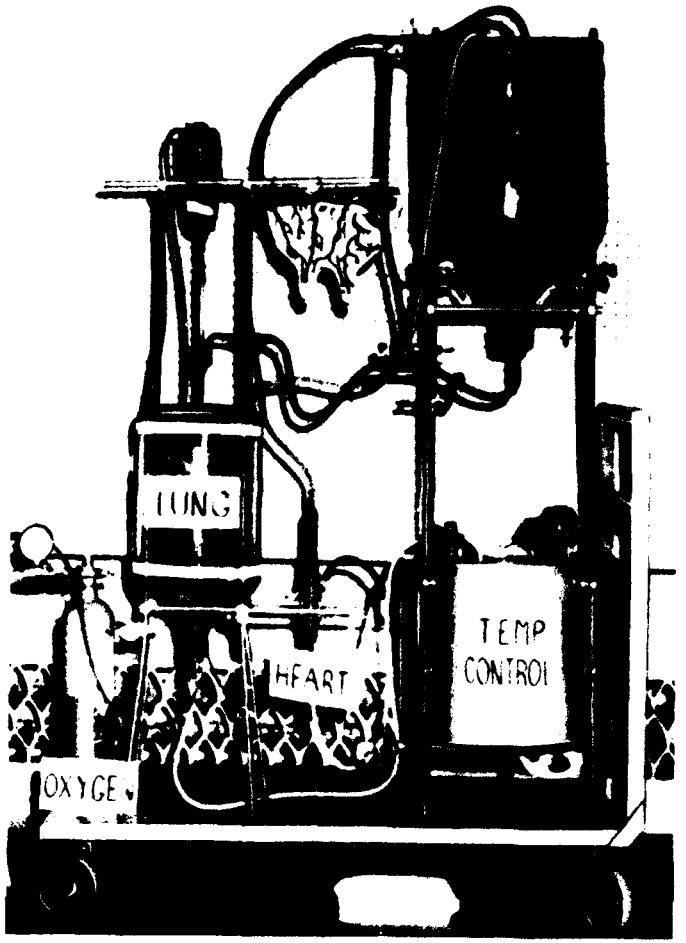


Fig. 6.26—Artificial cow for study of milk formation by use of tracers (a fresh udder in system forms milk).[156]

In other studies with cattle, radioiodine has been used to study the role of the thyroid in heat-resistance characteristics. Radiosulfur is being employed in studies of the synthesis of the amino acids, cystine, and methionine by sheep and cattle and also in the study of the problem of selenium poisoning, which has occurred in Wyoming. Radiocobalt is being used in the study of the synthesis of vitamin B_{12}. Radiophosphorous has been used in the study of the spread of honeybee diseases by carriers between colonies and in the study of the feeding habits of bees. The radioactive isotopes also are being employed as the tracers for insecticides and as markers in following hibernation and migration of insects.

Tracer techniques are ideally suited to investigating biological systems since there is nothing to alter the conditions of the experiments. As a result, more use has been made of tracer techniques in the fields of agriculture and medicine than in industry.

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Chapter 7

PROMOTION OF CHEMICAL REACTIONS BY RADIATION

Two major phenomena mark the atomic fission and fusion reactions. First there is release of tremendous radiation energy and heat. The radiation from fission and fusion can also promote significant chemical reactions. These chemical reactions, in turn, breaking as they do certain key molecules, are believed to cause the many and varied biological effects that follow exposure to radiation. Thus most of the biological effects discussed in Chaps. 8, 9, and 10 stem from the effects of radiation on chemical molecules.

This chapter begins by discussing the scope of radiation chemistry and comparing it with photochemistry. Effects of radiation chemistry are reviewed from the standpoint of chemical bonding types and physical states. Mechanisms of radiation chemistry, as well as the thermodynamics and kinetics involved, are considered.

Attention is given to chemical reactions that hold promise of possible industrial application. Such reactions include nitrogen fixation, ethylene polymerization, polyethylene cross-linking, halogenation of aromatics, sulfonation, rubber vulcanizing, ethylene oxidation, and many others. A discussion of radiation damage to chemical materials concludes the chapter.

SCOPE OF RADIATION CHEMISTRY

Interest in the chemical reactions that radiation can promote has been stimulated by the fact that this radiation has recently become available in large quantities as by-products of nuclear reactor operations. For more than a century, it has been known that solar radiation promotes some types of chemical reactions. Later it was recognized that the ultraviolet region of the solar spectrum is the most effective source of these reactions. Advent of ultraviolet lamps led to extensive research in photochemistry.

The discovery of radioactivity some 60 years ago made it possible to investigate how alpha, beta, and gamma radiation promote chemical reactions. These sources of "ionizing" radiation from natural radioisotopes have low specific activity, but more powerful sources came into use with the invention of X-ray machines, electron accelerators, and nuclear reactors. All this has made possible a wide range of studies in "radiation chemistry." A comparison of the fields of photochemistry and radiation chemistry follows.

PHOTOCHEMISTRY

Photochemistry is that branch of chemistry concerned with promoting chemical reactions by radiation in the wavelength range 1000 to 10,000 A, which corresponds to a quantum-energy range of 1.2 to 12 ev. In the first stage of a photochemical reaction an atom or molecule absorbs radiation and, thus raised to an excited state, the excited atom or molecule can undergo the following reactions:

1. Rupture of the molecule.
2. Reaction with another molecule or atom.
3. Internal rearrangement of the molecule.
4. Transfer of energy to another atom or molecule having a lower excitation potential.
5. Reemission of radiant energy by transitions to lower atomic or molecular energy states (fluorescence).

RADIATION CHEMISTRY

Radiation chemistry encompasses the interaction with matter of highly energetic radiations, including protons, deuterons, neutrons, alpha and beta particles, and X rays and gamma rays. In contrast with photochemical reactions, in which the relatively low energy radiation generally produces only excited atoms and molecules, the highly energetic atomic particles and radiation both excite and ionize many atoms and molecules. Also in photochemistry it is often possible to employ a specific wavelength radiation that will cause the reaction to go almost completely in the forward direction. This is not possible in radiation chemistry because the high energy of the atomic particles and radiation gives rise to reverse and side reactions.

TYPES OF CHEMICAL BONDS

In discussing the interaction of gamma radiation with matter it is important to know what types of chemical bonds are present in the molecular species being irradiated. Hence the primary bond types will be discussed.

Covalent Bonds

The chemical bonds between the atoms of most common liquids, gases, and organic compounds are covalent; the atoms are grouped as molecules and share valence electrons. Relatively weak Van der Waals forces hold the molecules together. The bonding energies are of the order of a few electron volts, as shown in Table 7.1. As these energies are less than the energy of ionization of an orbital electron, all types of ionizing radiation can break these covalent

Table 7.1 — CHEMICAL BOND ENERGIES[1]

Bond	Energy, kcal/mol	Energy, ev	Bond	Energy, kcal/mol	Energy, ev
C–C	80.5	3.5	C–Cl	78	3.38
C=C	145	6.5	C–Br	54	2.34
C≡C	198	8.6	O–O	34	1.47
C–O	79	3.38	H–H	103.2	4.48
C=O	173	7.5	N–N	37	1.6
C–N	66	2.36	Cl–Cl	57.1	2.48
C–H	98.2	4.35	Br–Br	46	2.0

bonds, producing free radicals and/or atoms by decomposing the original molecules. Also, the free radicals or atoms may combine to form new molecules, in which case the physical properties of organic materials may be considerably modified by ionizing radiation. This effect of ionizing radiation upon covalent bonds is the primary explanation for such phenomena as the degradation of methyl methacrylate, rubber, and various other high polymers, the cross-linking of polyethylene, and chemical reactions such as halogenation and polymerization of monomers.

Anderson and Martin[2] have discussed the effect of ionizing radiation on covalent bonds, using models of organic molecules as shown in Figs. 7.1 and 7.2 to demonstrate some possible types of interactions.

Figure 7.1 shows schematically some possible effects of ionizing radiation on hexane. Carbon-carbon bonds may be broken to give a two-carbon radical and a four-carbon radical or

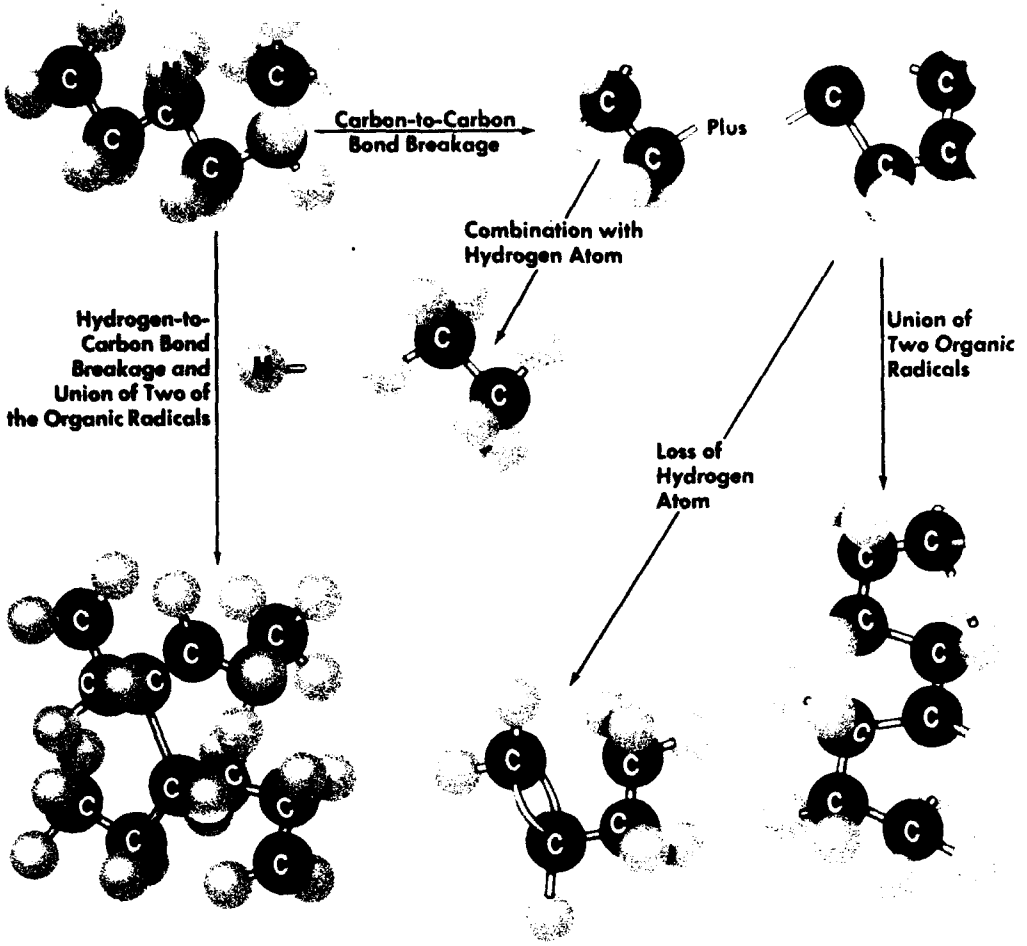


Fig. 7.1—Schematic diagram of some possible effects of ionizing radiation on hexane.[2]

there may be a three-three, or a one-five split of carbon-carbon bonds. One possible split of a carbon-hydrogen bond and subsequent combination is shown; two other types of split of the carbon-hydrogen bond (depending on the hydrogen atom removed) are equally feasible.

The figure illustrates that the first effect of ionizing radiation on such a hydrocarbon is the fragmentation of the original molecule into organic free radicals. These free-radical intermediates have a very short life of the order of thousandths of a second and recombine with other free radicals to produce new molecules. Products of higher molecular weight may be produced at the same time that products of lower molecular weight are produced. These reactions can be either constructive or destructive to the properties of plastics. Some, such as the acrylates, have been observed to disintegrate after irradiation. In other polymers, such as polyethylene, cross-bridging may occur, producing a desirable increase in rigidity and melting point.

If the organic molecule contains a double bond (as in the case of ethylene, Fig. 7.2) the first effect of the ionizing radiation may be to open the double bond. Such biradicals can combine end to end in a chain of reactions to form polymers of high molecular weight. As more ethylene molecules combine, the polymer may be broken by other gamma photons. The recombination of broken polymer molecules plus the continued addition of ethylene molecules leads

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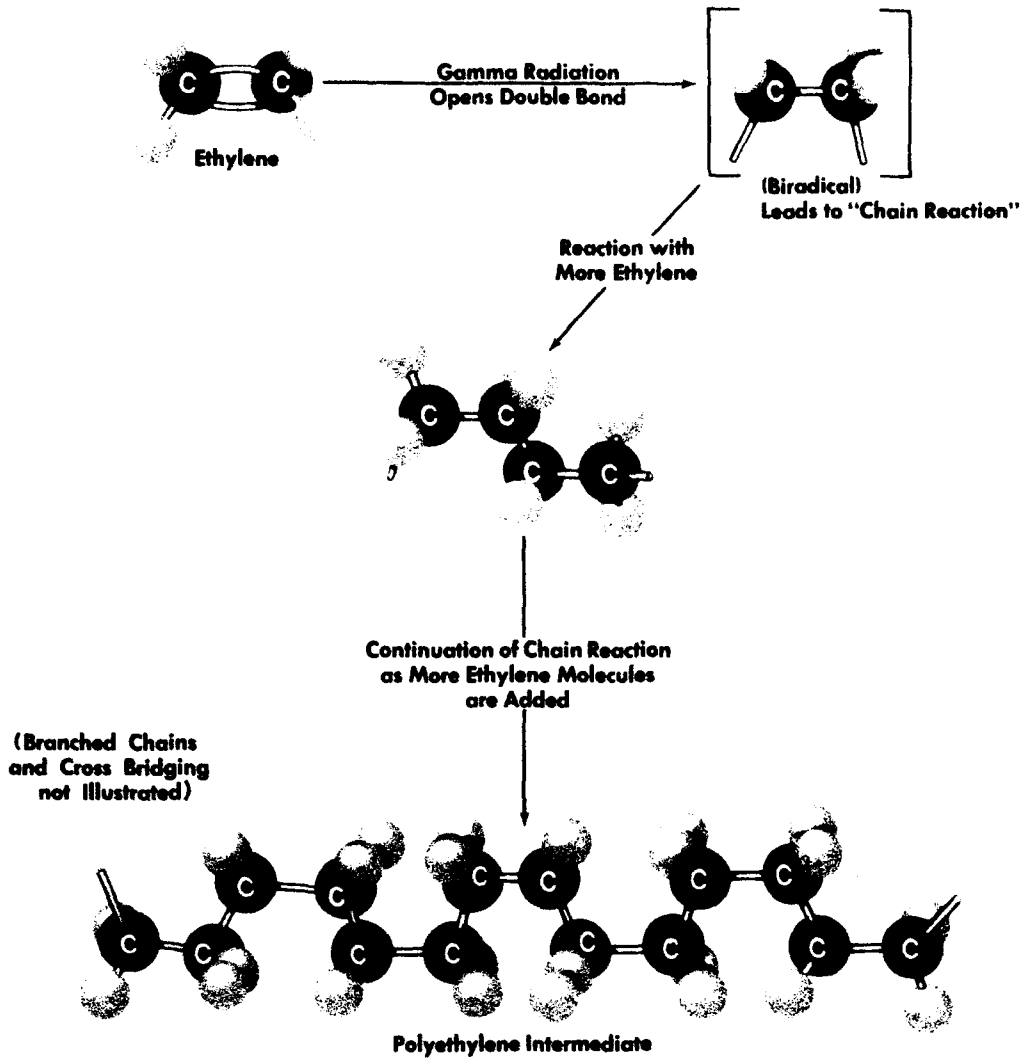


Fig. 7.2—Schematic diagram of some possible effects of ionizing radiation on ethylene.[2]

to the formation of branched chains and cross-linking. Thus, polyethylene produced by the use of ionizing radiation can be expected to have properties different from polyethylene produced by other methods.

Ionic Bonds

Solid metallic salts, such as KCl and NaBr, are examples of pure ionic bonding, in which a positively charged atom combines with a negatively charged atom. The molecules in this case consist of an ionic lattice with electrostatic charges alternately positive and negative. These lattice energies are greater than those covalent bonds (see Table 7.2), but energetic photons can eject electrons from ions to form free atoms, such as Cl and Br.

Unusual ions such as Na^{++} may also be produced, but there will be no change in the space lattices of the solid. The ejected electrons may migrate back to their original positions or to similar positions in neighboring atoms and recombine with the atoms. In such cases there will be little or no degradation of substances held together with ionic bonds. However, sometimes the ejected electrons become trapped in one of the lattice centers. Although this has little effect on chemical and physical properties, it can produce a color change such as is observed in gamma-irradiated glass.

Table 7.2—LATTICE ENERGIES OF METALLIC HALIDES[1]

Metal halide	Lattice energy, ev	Metal halide	Lattice energy, ev
NaCl	8.0	NaF	9.4
KCl	7.3	KF	8.4
RbCl	7.0	RbF	7.9
AgCl	9.3		

Semipolar or Dative Bonds

In semipolar bonding both of the electrons forming the shared duplet are supplied by one atom. Examples of this are the N–O bond in R_3NO and the B–P bond in BCl_3PCl_3. Semipolar bond energies are of the same order of magnitude as covalent bond energies.

Metallic Bonds

The metallic structure can be pictured as a free electron "gas" enclosed within the geometrical limits of the solid. There is no individual bonding of any given electron to any given metal atom; most metals are monovalent in the solid state and each contributes one electron to the free electron gas. Elements with one or two electrons in the outer shell are more likely to be metallic in properties than elements with nearly complete outer shells of electrons. At the next stage of complexity, the Sommerfeld theory of metals regards the electron gas as being subject to a potential which varies around each atom.

Alpha particles, protons, and deuterons rarely collide directly with the nucleus; instead, they strip away orbital electrons. X radiations and gamma radiations interact with atoms and molecules to produce ions and high-energy electrons, as well as excited atoms and molecules. The recoil electrons play an important role in the reactions to be discussed in the following sections.

The electrons are easily moved by passing an electric current through the metal. The only effect of the absorption of low-energy gamma radiation in metals is to produce heat.

INTERACTION OF IONIZING RADIATION WITH MATTER

Ionizing radiation actually produces two general classes of effects: atomic displacement (the Wigner Effect) and electronic excitation and ionization of the atom or molecule. By bombarding hydrogen, carbon, nitrogen, and oxygen with protons and alpha particles of approximately 5-Mev energy, Platzman[3] found that less than 0.1 per cent of the absorbed energy caused atomic displacements. With gamma radiation this effect is even smaller and hence it will not be further considered.

The three major processes by which gamma photons interact with matter are photoelectric effect, Compton scattering, and pair production. In the range of gamma energies most commonly used in radiation chemistry almost all interactions are by photoelectric and Compton effects. Burton[4] points out that for incident gamma photons of 100-kev energy about 3 per cent of the recoil electrons are photoelectrons; the remainder are produced by Compton scattering. The mean energy of the recoil electrons is slightly greater than 15 kev. For incident gamma photons of 1-Mev energy, essentially all the recoil electrons are produced by Compton scattering and their mean energy is about 440 kev.

EFFECT OF IONIZING RADIATION UPON MOLECULES

The effect ionizing radiation has upon molecules depends on the physical state of the molecules. The effects on molecules in gases, liquids, and solids are covered in the following paragraphs.

Gases

Gamma radiation ionizes gases by the photoelectric effect, Compton scattering, and pair production. The energy absorbed per ion pair varies from about 28 ev for helium to about 36 ev for nitrogen, whereas the corresponding ionization potentials for these gases are 24.5 and 15.7 ev, respectively. The difference between these energies is accounted for by interactions between molecules and photons that do not produce ionization. These interactions cause transfer of energy to gas molecules, raising them to a more excited state. The net result is to form ions, unstable atoms, free radicals, and reactive molecules. Recent concepts stress the importance of free radicals and short-lived transient species.

Reactions with gaseous molecules, ionized or excited by gamma radiation, are generally similar to those promoted by ultraviolet radiation and electric discharge. For example, the additive chlorination product of benzene, hexachlorocyclohexane (commonly known as benzene hexachloride) is produced commercially using ultraviolet radiation. It is also found to be one of the products of the interaction of chlorine gas with benzene in the presence of gamma radiation. Gases may react chemically with other molecules present, decompose, or interact by exchanging electrons. In the latter case interaction between ions may form new compounds; for example, ozone is formed by the ionization of oxygen. On the other hand, interaction between ions may also result in further decomposition. The products of these reactions will themselves be subject to interaction with gamma photons and secondary ionizing radiation; this may result in reconstitution of the original molecule. Thus, with a limited volume and a simple system, an equilibrium is reached in which the various possible compounds, ions, atoms, etc., are found in fixed proportions. In the case of water vapor and air, equilibrium is attained very rapidly. If one of the products is removed from the system by change of state or interaction with another material, equilibrium will not be reached. Irradiation of complex organic vapors forms so many radicals and intermediate products that they cannot be considered to reach equilibrium.

Liquids

Molecules in the liquid phase are much more closely packed than molecules in the gaseous phase and there is a more complex distribution of energy states. Free radicals and atoms form at a lower rate per unit mass than in a gaseous phase. Reactions in the liquid phase are believed to be initiated primarily by activated molecules. Some attempt has been made to analyze impact processes, radical reactions, neutralizations, and reactions of vibrationally excited ions in irradiated liquids. Collision recombination may occur before the products can diffuse to a reacting zone. This is known as the Frank-Rabinovitch "cage effect." Little is known about the distribution in space of ionized or excited molecules, but it is believed to influence the reaction rate. If X and Y are the product molecules and A and B are free radicals reacting as follows: $A + A \rightarrow X$ and $X + B \rightarrow Y$; the formation of X is a second-order reaction whereas the destruction of X is a first-order reaction. An increase in ion density will increase the concentration of free radicals A. The rate of reaction will depend then upon the velocity constants and the distribution of the free radicals. The concentration of free radicals is influenced by the rate of ionization.

The effect of gamma radiation upon liquids may conveniently be divided into the effects upon water and aqueous solutions and the effects upon organic liquids.

Water and aqueous solutions. Very little decomposition occurs when pure water is exposed to either gamma- or beta-particle radiation. However, if the water contains dissolved impurities or if the water is used as a solvent, appreciable decomposition of the water occurs. Probably the first step in this decomposition is the ejection of an electron to produce the ion pair e^- and H_2O^+.

$$H_2O \rightsquigarrow e^- + H_2O^+ \qquad (7.1)$$

This ion pair can then interact with other water molecules

$$e^- + H_2O \rightarrow OH^- + H \qquad (7.2)$$

$$H_2O^+ + H_2O \rightarrow H_3O^+ + OH \quad (7.3)$$

By Eqs. 7.2 and 7.3 the free radicals H and OH may be produced. These free radicals may interact with each other or may participate in other reactions

$$H + OH \rightarrow H_2O \quad (7.4)$$

$$H + H \rightarrow H_2 \quad (7.5)$$

$$OH + OH \rightarrow H_2O_2 \quad (7.6)$$

$$H_2 + OH \rightarrow H_2O + H \quad (7.7)$$

$$H_2O_2 + H \rightarrow H_2O + OH \quad (7.8)$$

$$H_2 + H_2O_2 \rightarrow 2H_2O \quad (7.9)$$

The reactions indicated by Eqs. 7.4, 7.7, 7.8, and 7.9 can be considered to be reverse reactions that tend to remove the products of the reactions indicated by Eqs. 7.2, 7.3, 7.5, and 7.6. An equilibrium will be reached which depends upon the nature of the ionizing radiation and the purity of the water. Pure water under gamma- or beta-particle irradiation can be considered stable, as the concentration of H and H_2O_2 at equilibrium is so low. In the case of heavy-particle irradiation, such as by neutrons, alpha particles, fission fragments, etc., a greater degree of decomposition occurs. Water containing either an oxidizing or reducing agent is further dissociated, as one of the products of the forward reaction can be removed. When aqueous solutions of organic compounds are irradiated, the OH will oxidize the organic compounds to produce CO_2, thereby removing the OH free radical and preventing its reaction with H. In this case, the concentration of the H free radical will increase until it competes with the organic material for OH. In complex organic systems capable of both reduction and oxidation, both H and OH are continuously removed and no equilibrium will exist. Dissolved inorganic reducing or oxidizing agents can also remove free radicals and prevent equilibrium. Inorganic solutes that are neither reducing nor oxidizing increase the equilibrium concentrations of hydrogen and hydrogen peroxide in the water above those for pure water.

Most of the characteristics of the radiation chemistry of aqueous systems can be accounted for without consideration of the primary events by which radicals are formed. A simplified equation giving a summation of the events is

$$H_2O \rightsquigarrow \text{Intermediates} \rightarrow aH + bOH + cH_2 + dH_2O_2 \quad (7.10)$$

A large number of experimental investigations have been conducted to determine the values of the coefficients a, b, c, and d for different types and dose rates of radiation, for different solutes and solute concentrations, and for other experimental variables. These studies have provided information on the formation of H and OH radicals and the spatial distribution of the primary products. Allen[5] and Dewhurst, Samuel, and Magee[6] have summarized these studies.

Organic liquids. Organic materials under continued irradiation will eventually decompose. With most organic systems there are so many intermediate products and so many reactions possible that there is little or no opportunity for reverse reactions to establish an equilibrium. Irradiation of pure hydrocarbon liquids will lead to one or more of the following reactions: degradation of the molecule (cracking); polymerization or condensation; hydrogenation of unsaturates; and dehydrogenation. Since these reactions both break down and build up the size of molecules, the final product will contain a mixture of gases such as hydrogen, methane, and ethane; a mixture of high-molecular-weight sludges and tars, including some carbon; and a mixture of intermediate products. When either oxygen or a halogen is available, oxidation or halogenation may occur. A number of these reactions are of commercial importance and are discussed in greater detail in a following section.

Solids

Metals and ionically bonded materials, such as most inorganic solids, strongly resist degradation or change, except in color, as a result of low-energy gamma irradiation from radioisotopes. These materials, however, suffer damage when bombarded by neutrons, other heavy particles, and high-energy gamma radiation.

Organic solids like cellulose and plastics undergo changes similar to those in organic liquids. Cellulose darkens and loses its tensile strength. Evolution of gas in methyl methacrylate results first in crazing and then disintegration to a dust. Cross-linking in polyethylene raises its melting point. Other organic solids undergo various changes as a result of irradiation.

REACTION PROCESSES IN RADIATION CHEMISTRY

Specific inorganic substances under irradiation form more than one radical per ion pair. Early attempts to explain polymerization and large yields per ion pair postulated a "cluster theory" in which it was assumed that molecules clustered around a central positive ion. It was thought that in polymerization induced by ionizing radiation, this cluster was discharged by an electron and left in an excited state in which polymerization occurred. Later studies showed that clustering probably is not the only mechanism in the promotion of chemical reactions by radiation. A more recent theory, based on the early work of Eyring, Hirschfelder and Taylor[7] postulates a mechanism of ionization involving the formation of excited molecules, ions, and radicals.

An excellent pictorial summary of the various primary processes that can occur has been given by Burton.[8] Harmer et al.[9] prepared a modification of this schematic drawing as shown in Fig. 7.3. The most important processes have been indicated by heavy lines in the drawing. Examination of the complete figure shows the many and complex processes that can occur in any chemical system subjected to gamma radiation. The several steps in the series of processes which form the reactive species following the encounter of a molecule with the high-energy particle or photon are considered in this section.

IMPACT PROCESSES

In an impact process a gamma photon interacts with atoms or molecules. Since most gamma-photon energies are of the order of 1 Mev, the interaction will be primarily by Compton scattering, giving rise to the reactions

$$A \rightarrow A^+ + e \text{ (ionization of atom)} \tag{7.11}$$

$$M \rightarrow M^+ + e \text{ (ionization of molecule)} \tag{7.12}$$

$$M \rightarrow AB^{+*} + e \rightarrow A + B^+ + e \text{ (ionization and excitation followed by decomposition)} \tag{7.13}$$

Also, although the theoretical ionization energy is less than 15 ev, in practice over 30 ev must be expended to produce an ion pair. Thus, more than 50 per cent of the gamma-photon energy goes to excite atoms and molecules. This reaction is represented as

$$A \rightarrow A^* \tag{7.14}$$

$$M \rightarrow M^* \tag{7.15}$$

Similarly, the ions produced in the previous three reactions may also be in excited states if the incident gamma-photon energy is large enough. The theory of Eyring, Hirschfelder, and Taylor states that the electron rather than the positive ion plays the major role in the subsequent reactions.

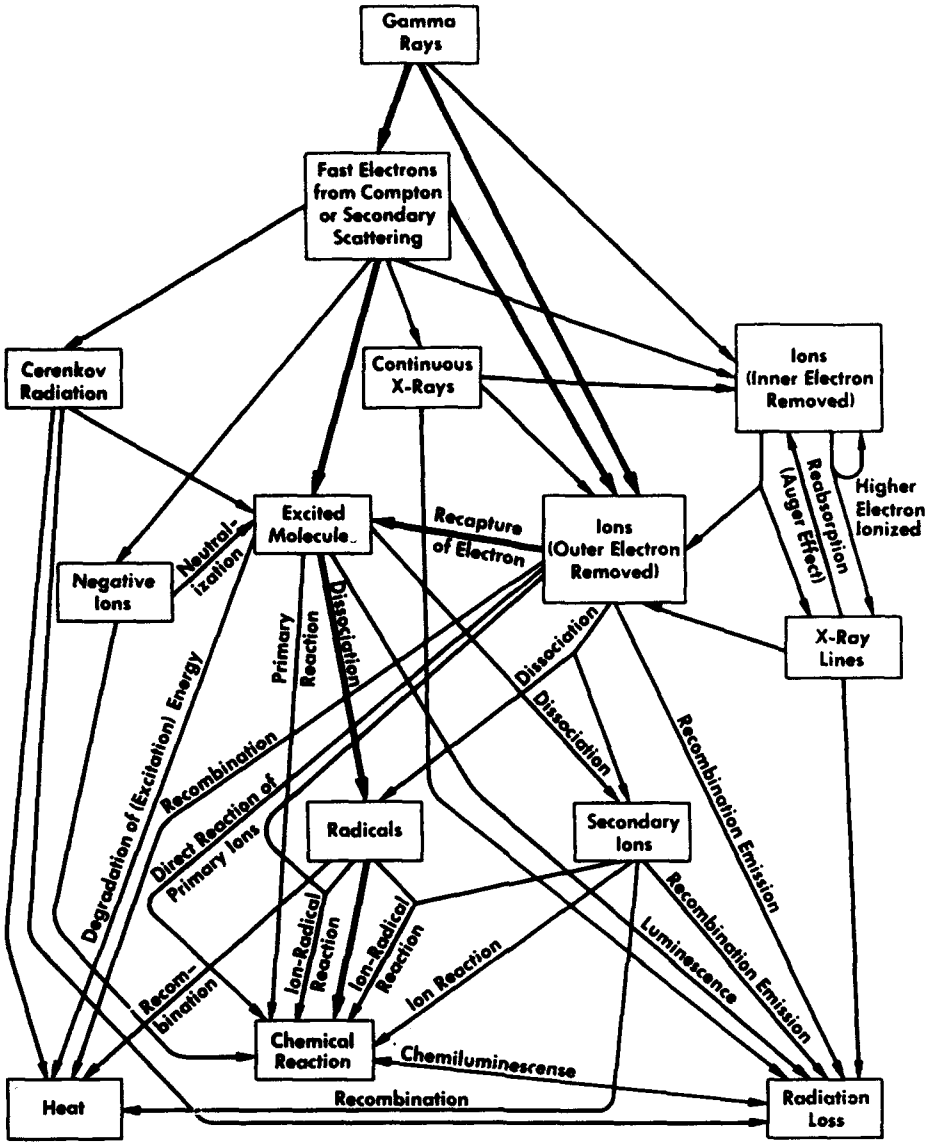


Fig. 7.3 — Possible processes of energy transfer and intermediate stages resulting from the action of gamma radiation on a chemical system.[9]

ELECTRON CAPTURE

The electron produced in an impact process may undergo the reactions

$$A^+ + e \rightarrow A^* \text{ (capture of electron by atomic ion)} \tag{7.16}$$

$$M^+ + e \rightarrow M^* \text{ (capture of electron by molecular ion)} \tag{7.17}$$

$$M + e \rightarrow M^- \text{ (capture of electron by molecule)} \tag{7.18}$$

$$AB + e \rightarrow AB^{-*} \rightarrow A + B^- \text{ (electron capture and dissociation)} \tag{7.19}$$

$$AB + e \rightarrow AB^* + e \rightarrow A^+ + B^- + e \text{ (ion-pair production by high-energy electron)} \tag{7.20}$$

It has been observed that negative-ion formation, Eq. 7.18, occurs only when the reacting molecule contains a functional group with a high electron affinity. Where molecules have low

electron affinities, the electron must slow down considerably before its capture is likely. Equations 7.18 and 7.20 show processes by which negative ions may be produced by high-energy electrons.

IONIZATION AND EXCITATION TRANSFER

In certain cases the product ions of the impact processes may transfer their ionization or excitation energy. If there is an un-ionized molecule B having a lower ionization potential than the ion A^+, ionization transfer may occur

$$A^+ + B \rightarrow A + B^+ \tag{7.21}$$

Similarly, if B is unexcited, but has a lower excitation potential than the excited molecule A*, excitation transfer may occur

$$A^* + B \rightarrow A + B^* \tag{7.22}$$

If the molecule B is large, the excitation energy may distribute itself uniformly among the various bonds so that no one bond will possess enough energy to rupture. This explains the stability of organic aromatic systems to irradiation.

NEUTRALIZATION OF POSITIVE IONS

Positive ions that do not capture electrons may interact with negative ions

$$A^+ + B^- \rightarrow AB^{**} \rightarrow A^* + B^* \tag{7.23}$$

where either or both of the product radicals may be in excited states.

DISSOCIATION OF EXCITED MOLECULES

Unless the molecule is large and the excitation energy can distribute itself among the various bonds without causing one to rupture, the excited molecules produced by electron capture or by the interaction of unlike ions will dissociate rapidly. Decomposition reactions may occur

$$M^* \rightarrow A^+ + B^- \text{ (secondary ion formation)} \tag{7.24}$$

$$M^* \rightarrow A + B \text{ (molecule formation)} \tag{7.25}$$

$$M^* \rightarrow A^{\cdot} + B^{\cdot} \text{ (free-radical formation)} \tag{7.26}$$

According to Burton[4] one of the free radicals formed in the last reaction is probably in an excited state.

The decomposition of hydrogen bromide in the presence of gamma radiation is a classical study[10,11] of a reaction mechanism. It has been determined experimentally that four atoms of hydrogen bromide decompose for each ion that is formed. The mechanism proposed to explain this was

$$HBr \rightarrow HBr^+ + e \text{ (impact process)} \tag{7.27}$$

$$HBr + e \rightarrow H + Br^- \text{ (electron capture)} \tag{7.28}$$

$$Br^- + HBr^+ \rightarrow H + Br_2 \text{ (charge neutralization)} \tag{7.29}$$

$$2H + 2HBr \rightarrow 2H_2 + 2Br \text{ (atom reaction)} \tag{7.30}$$

$$Br + \dot{B}r \rightarrow Br_2 \text{ (atom recombination)} \tag{7.31}$$

The mechanism for this series of reactions follows the theory just considered.

Detailed studies of carbon dioxide under ionizing radiation[12–14] show that in the absence of catalysts, carbon dioxide irradiated at room temperature undergoes only about 0.1 per cent of the expected degree of decomposition. Early workers[13] ascribed this in part to the occurrence of the back reaction

$$3CO + O_3 \rightarrow 3CO_2 + 103 \text{ kcal} \tag{7.32}$$

But recent experimental evidence[14] indicates a heat of activation of greater than 28 kcal for this reaction at room temperature. Therefore the reaction in Eq. 7.32 proceeds very slowly and cannot account for the low degree of decomposition of the carbon dioxide.

Experimental studies of carbon monoxide under ionizing radiation indicate that about three molecules of carbon monoxide are decomposed per ionizing event. The mechanism postulated to explain this reaction is

$$CO \rightarrow C + O \tag{7.33}$$

$$C + CO \rightarrow C_2O \tag{7.34}$$

$$C_2O + CO \rightarrow C_3O_2 \tag{7.35}$$

$$C_3O_2 + O \rightarrow C_2O + CO_2 \tag{7.36}$$

The heats of activation of the last three reactions are about 3 kcal/mole. This back-reaction mechanism explains the low yield of carbon monoxide when carbon dioxide is irradiated in the absence of catalysts.

Ionizing radiation does decompose carbon dioxide if nitrogen dioxide is present in the system. It has been found[14] that as little as 0.5 per cent nitrogen dioxide stops the carbon monoxide back reaction, and that using larger percentages of nitrogen dioxide does not appreciably increase the decomposition. The nitrogen dioxide is thought to inhibit the back reaction by interacting with the intermediate atoms and radicals by the mechanisms

$$NO_2 + O \rightarrow NO + O_2 \tag{7.37}$$

$$NO_2 + C \rightarrow NO + CO \tag{7.38}$$

$$2NO + O_2 \rightarrow 2NO_2 \tag{7.39}$$

In this decomposition approximately three molecules of CO_2 are decomposed per ionizing event.

Additional information on the mechanism of chemical reactions promoted by ionizing radiation are given in Refs. 15 to 30.

THERMODYNAMIC AND KINETIC CONSIDERATIONS FOR REACTION PROMOTED BY IONIZING RADIATION

Martin and Anderson[31] have discussed the thermodynamic and kinetic considerations for chemical reactions promoted by ionizing radiation. Thermodynamically a chemical reaction can be caused if the products are at a lower energy level than the reactants. Reactions such as the oxidation of carbon or hydrogen proceed readily, once started, because the products of combustion, CO_2 or H_2O, represent a lower energy state than the reactants, C or H_2 and O_2. However, the noncatalyzed reaction does not occur at room temperature except at very slow rates. A portion of the reactants must be heated to an ignition temperature providing sufficient energy for a significant portion of the reactants to combine. The energy released by the reaction then provides new energy to cause an additional portion of the reactants to combine, re-

sulting in a chain of reactions which can be explosive in a system such as a mixture of hydrogen and oxygen.

Consider the reversible reaction

$$A + B = C + D \tag{7.40}$$

The law of mass action states that for a homogeneous system the rate of a chemical reaction is proportional to the active masses of the reacting substances. The rates of the direct and reverse reactions are

$$r_1 = k_1 C_A C_B \tag{7.41}$$

$$r_2 = k_2 C_C C_D \tag{7.42}$$

The constants of proportionality, k_1 and k_2, are called the reaction velocities for the direct and reverse reactions. At equilibrium

$$r_1 = r_2, \text{ or } k_1 C_A C_B = k_2 C_C C_D \tag{7.43}$$

or

$$\frac{C_C C_D}{C_A C_B} = \frac{k_1}{k_2} = K \tag{7.44}$$

where K is the equilibrium constant for the reaction. For the reaction to go predominantly in the forward direction, the equilibrium constant K must be greater than unity.

Thermodynamic considerations show that

$$K = e^{(-\Delta F^\circ/RT)} \tag{7.45}$$

In Eq. 7.45 the term ΔF° is the standard free energy change, R is the gas constant, and T is the absolute temperature. For products to predominate in a system at equilibrium, ΔF° must be negative.

For a specific reaction velocity constant, the Arrhenius equation may be assumed to hold

$$k_t = Ae^{(-E_t/RT)} \tag{7.46}$$

In Eq. 7.46 the term E_t is the energy of activation and A is the frequency factor for the reaction.

For a reaction occurring in a field of radiation Eq. 7.46 may be modified as

$$k_{tr} = A[e^{(-E_t/RT)} + g] \tag{7.47}$$

In Eq. 7.47 the term g is the fraction of molecules activated by irradiation. A study of this equation with regard to the temperature and radiation intensity factors, shows the following: The rate of a reaction induced by gamma radiation is dependent on the dose rate and may be nearly independent of temperature over a wide range of temperatures less than a certain temperature at which or above which, the reaction due to thermal activation may be controlling.

CHEMICAL REACTIONS OF POSSIBLE INDUSTRIAL INTEREST

Ionizing radiation offers several advantages as a catalyst for certain inorganic and organic chemical reactions. By employing radiation a reaction may be carried out at lower temperatures, which is an advantage both economically and when a reactant is temperature sensitive. The use of radiation also obviates the introduction of foreign substances as catalysts and, be-

cause of the highly penetrating nature of the radiation, there is great flexibility in the design of chemical reactors.

INORGANIC REACTIONS

Henley[32] divided the chemical reactions considered of possible industrial interest into inorganic reactions as shown in Table 7.3, and organic reactions as shown in Table 7.4. In general, because of the higher yield and higher value of the products, the organic reactions (Table 7.4)

Table 7.3—INORGANIC SYSTEMS OF POSSIBLE COMMERCIAL INTEREST IN RADIATION CHEMISTRY AFTER HENLEY[32]

Reactants	Products	Radiation	G, moles/ 100 ev	Tons of reactants converted per year per 10^6 Mev-curie*	Refs.
Oxidation					
Oxygen	Ozone	α, e^-	6.3	4	27
$O_2 + N_2$	NO, O_3, N_2O, NO_2	α	3	2	33
$CO + H_2O$†	CO, CO_2, H_2, HCHO	X	12	10	28
$NaCN + O_2$	$N_2 + Na_2CO_3 + H_2O$	γ	3	1.7	34
$Na_2S + H_2O$	$Na_2SO_4 + 4H_2$	γ	6	4.3	34
Reduction					
$CO_2 + H_2$	$H_2O + (H_2CO)x$	α	5	4.5	29
$CO + H_2$	Polymer	α	10.5	6	29
Decomposition					
HBr	H + Br	α	13	17	9
H_2S	$H_2 + S_2$	α	8	5	13
H_2O	$H_2 + H_2O_2$	α, e^-	2	1	14
Synthesis					
$H_2 + N_2$	NH_3	α	0.6‡	1.3	29
NH_3	$N_2H_4 + H_2$	α	0.6	1.15	15
$CO + Cl_2$	COCl	e^-	26×10^4	21×10^4	16

*Mev-curie is the product of source energy in Mev and source strength in curies.
†Conditions, pH 1 to 7.
‡No yield with gamma radiation.

are probably of greater interest than the inorganic reactions (Table 7.3). However, Harteck and Dondes[29] performed experiments on an inorganic system of great commercial interest, the fixation of nitrogen in a nuclear reactor. These authors used the resulting data to make an economic study of the process. The calculations were based on a reactor plant that each year consumes 1000 moles of U^{235} to produce heat at the rate of 550 Mw. If the plant were used to fix nitrogen, it could theoretically produce 250,000 tons of nitrogen dioxide. Besides the fixed nitrogen this plant could furnish 110,000 tons of by-product nitrous oxide and heat at 200°C, equivalent to 500,000 tons of coal.

The reduction of CO_2 to CO and O_2 is another inorganic reaction studied by Harteck and Dondes[14]. Their experiments indicated that the tremendous kinetic energies of the fission fragments might be converted directly into chemical energy in the form of CO and O_2. Such direct use of fission energy might be very useful in process industries where great quantities of CO and O_2 can be used to advantage. A more extensive discussion of the work of Harteck and Dondes is given in Chap. 4.

The radiolysis of water to produce H_2 and O_2 has been discussed in Chap. 6 as a source of hydrogen for the Bacon cell, which very efficiently converts chemical energy to electrical energy through a process the reverse of electrolysis of water. Appreciable amounts of hydro-

Table 7.4—ORGANIC SYSTEMS OF POSSIBLE COMMERCIAL INTEREST IN RADIATION CHEMISTRY AFTER HENLEY[32]

Reactants	Products	Radiation	G, moles/ 100 ev	Tons of reactants converted per year per 10^6 Mev-curie*	Refs.
Oxidations					
Benzene + H_2O	Phenol	X	16.5	23	17
Nitrobenzene + H_2O	Hydroxylated nitrobenzene	e^-	22	67	19
Benzoic acids	Salicylic acids	X	5	45	18
C_6H_6 + air	Phenol	β, γ	180	186	15
Steroids	Trans triols in 5-6 positions			655	20
Decompositions					
Palmitic acid	n-heptadecene	β	1.8	7.6	21
Wood	Edible matter	e^-	1.8	6×10^3	35
Condensation and polymerization					
CH_4	$C_2H_6 + H_2$ etc.	α	6.6	1.8	10
C_4H_{10}	$C_8H_6 + H_2$ etc.	α	5.4	5.4	10
C_2H_2	C_6H_6	α	60	26.6	36
C_2H_2	Polymer	γ, α	300	133	37
C_2H_4	Polymer	190°C, γ		2×10^3	38
C_2H_4	Polymer	1600 psi, γ	2000	870	39
Perfluoropropylene	Polymer	γ		60	38
Acrylonitrile†	Polymer	X	3×10^7	2.1×10^7	40
Chlorination					
$C_6H_6 + Cl_2$	$C_6H_6Cl_6$	γ	3×10^5	1×10^6	41

*Mev-curie is the product of source energy in Mev and source strength in curies.
†Condition, dilute H_2O solution.

gen and oxygen are produced in an aqueous homogeneous reactor. This reactor shows promise (because of its simplicity) as an industrial reactor. The recovery of energy from the products of radiolysis would aid in increasing the over-all efficiency.

ORGANIC REACTIONS

Ionizing radiation probably offers its greatest promise in chemistry as a catalyst for organic reactions.[33–41] The general types of organic reaction which are caused by irradiation are first discussed; then a few selected systems are considered in detail.

The effects of radiation on organic systems have been discussed by Collinson and Swallow[42] under synthesis, degradation, oxidation, halogenation, and reduction. These processes are briefly reviewed.

Ballantine[49] reported that liquid-phase radiation-initiated polymerizations generally have kinetic features very similar to the same reaction initiated by light or thermal catalysts. Thus the "Trommsdorff effect" has been observed in the gamma-ray bulk polymerization of styrene and methyl methacrylate,[47] and the gamma-ray emulsion polymerization of styrene is faster than the bulk polymerization.[43] Figure 7.4 is a plot according to Ballantine of the percentage conversion as a function of radiation dose for styrene.[49]

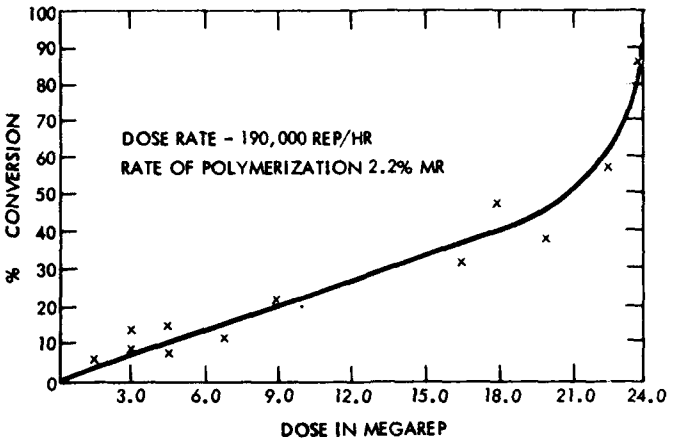


Fig. 7.4 — Polymerization of styrene at room temperature.[47]

Synthesis

In studies of polymerization by addition many liquid vinyl compounds have been irradiated both in the pure state and in solution. Polymerization by a free-radical chain mechanism is believed to occur.[42] The free-radical process is evidenced by the following factors: oxygen has an inhibiting action on this process, the molecular weights of the polymers produced can be very high and the products have all the usual properties of free-radical-produced polymers. Free-radical addition polymerization usually terminates as a result of the interaction of the chains. The rate of polymerization by this process depends on the square root of the dose rate in the cases of styrene, acrylonitrile, methyl methacrylate, and aqueous acrylamide.[43–45] In other cases, this dependence is not followed; the nonuniform distribution of the initiating radicals along the paths of the ionizing radiation may be responsible for these deviations.[46]

Radiation induced polymerization may occur in the solid state, as for acrylamide, or at low temperatures, as for tetraethylene glycol dimethacrylate which may be polymerized at −55°C.[48,49] Some halogen compounds, perfluoropropene, perfluorobutene and perfluoroacrylonitrile can only be polymerized by radiation.[50,51]

The polymerization of various unsaturated hydrocarbons, copolymerization of hydrocarbons, and copolymerization of hydrocarbons with sulfur dioxide are reactions of importance. The promotion of some of these reactions with gamma radiation will be discussed in the following sections.

A wide variety of graft copolymers has been prepared by gamma or X irradiation of a polymer in the presence of the monomer. Ballantine et al.[52,53] describe the formation of radiation-produced, trapped free radicals in thin films of polyethylene and their subsequent use in initiating the graft polymerization with styrene. Other similar studies have been made with polyethylene-acrylonitrile, 4-vinylpyridine, vinylcarbazole, and styrene and n-vinylprollidone with polytetrafluoroethylene (Teflon). Figure 7.5 shows data obtained on Teflon and styrene. A partial evaluation of the physical properties of several of these graft polymers is also given.[52,53]

Many polymers can be cross-linked by radiation, but apparently this will happen between $(-CH_2 \cdot CHR-)_n$ type polymers only, where R may be an organic group or hydrogen. Polymers of the form $(-CH_2 \cdot CR_1R_2-)_n$ will degrade and not cross-link under radiation. Thus polymethylmethacrylate is degraded and polyethylene is cross-linked by irradiation.

Gaseous polymerization is important in the case of acetylene, which yields 15 to 20 per cent benzene and a yellow solid product similar to cuprene. Also gaseous irradiated ethylene will polymerize at lower pressures and temperatures than are required without radiation. Hydrocarbons like methane will produce the dimer, ethane, and hydrogen. With higher hydrocarbons, some breakage of carbon-carbon bonds occurs along with the carbon-hydrogen bond fracture. The C−H bond has a higher bond strength than the C−C bond and various theories

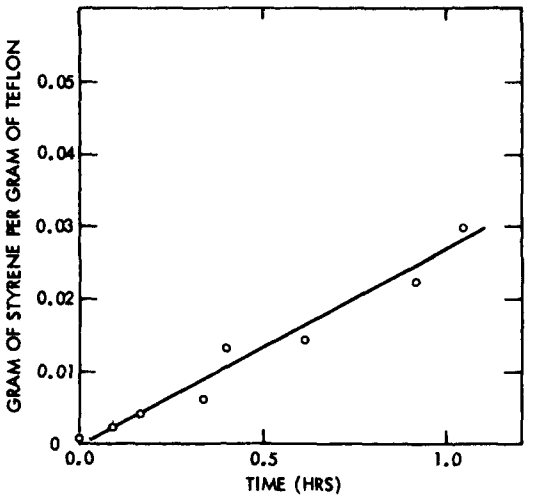


Fig. 7.5—Grafting of styrene to teflon (1-mil film at 183,000 r/hr).[52]

such as the "cage effect" have been developed in an attempt to explain the high degree of C–H bond breakage as a result of irradiation. Some specific attack also occurs on doubly bonded carbon atoms. Alcohols dimerize on irradiation; glycol, for example, being formed from ethyl alcohols. Alpha particles cause further oxidation in the case of ketones formed from secondary alcohols, but beta-particle radiation does not.[50,51] Dimers are frequently formed in aqueous solution and besides the formation of glycols from primary alcohols, formic acid dimerizes to oxalic acid, acetic acid to succinic acid, and benzene to diphenyl in the presence of dissolved hydrogen. If dissolved oxygen is present, phenol rather than diphenyl is more likely to form. Influencing the products by a hydrogenating or oxidizing environment is a method of fairly wide possible application.

Additional reactions of olefins with various sulfur compounds have also been investigated. The addition of n-butyl mercaptan to 1-pentene was studied by Fontijn and Spinks.[54] A study of reactions of hydrogen sulfide with various olefins was reported by Kothary and Yemin.[55] Nevitt has reported radiation-induced reactions of hydrocarbons with n-butyl mercaptan. The effectiveness of radiation for destruction of mercaptans for "sweetening" of petroleum is discussed.[56]

Oxidation

The radiation oxidation of aliphatic halides, notably of chloroform, has been studied. In the case of chloroform, the intermediate stage appears to be the formation, probably by chain reaction, of a peroxide CCl_3-O-OH with a high yield. The next stage is the decomposition of the peroxide with the formation of carbonyl chloride, hydrochloric acid, etc. Fats also proceed through an intermediate peroxide when subject to irradiative oxidation and this appears to be a general principle.[30] Aqueous solutions give hydroxyl radicals by irradiation and these hydroxyl radicals generally give some oxidation of any free radical formed. Aqueous alcohols appear always to be oxidized to carbonyl compounds, the amount of oxidation being increased by the presence of dissolved oxygen. Irradiation of thiols leads to oxidation, aqueous cysteine being converted to cystine. This latter remains unoxidized until most of the cysteine has gone, but is then oxidized itself to disulfoxide. Very high yields result in concentrated alkaline solutions indicating that a chain reaction is occurring.

Halogenation

Halogenation (discussed in more detail later) occurs when organic compounds are irradiated in the presence of a halogen such as chlorine, fluorine, iodine, or bromine. Some investigation of reaction mechanism[42] has been conducted with radioactive iodine. The radiation-

induced reaction between carbon monoxide and chlorine to give carbonyl chloride has been known as a chain reaction for many years. Aromatic hydrocarbons also chlorinate easily in the presence of gamma radiation, benzene giving good yields of hexachlorocyclohexane.

The effect of gamma radiation on the direct sulfochlorination of hydrocarbons and nitrobenzene was studied by Schneider.[57] The attempts to sulfochlorinate nitrobenzene and benzene were unsuccessful.

Liquid cyclohexane was found to react with sulfur dioxide and chlorine in the presence of gamma radiation to give very high yields of cyclohexanesulfonyl chloride and to a smaller extent chlorocyclohexane and cyclohexane disulfonyl chloride. A study of selectivity of reaction and yield of cyclohexanesulfonyl with respect to the variables of temperature, radiation intensity, and sulfur dioxide to chlorine ratio was made. The reaction rate was found to vary directly as the half power of the radiation intensity. The length of the chemical chain reaction was estimated to be over 2×10^5 and the radiochemical yield G (molecules of chlorine reacted per 100 ev of radiation absorbed) was greater than 10^6.

A basic plant design was developed for continuous sulfochlorination of cyclohexane initiated by gamma radiation. The economics of this process may become attractive with availability of cheap radiation sources.

Reduction

Irradiative reduction in the presence of hydrogen will occur for some molecules such as CO_2, which first yields formaldehyde and then a polymer, but this reduction does not occur with unsaturated hydrocarbon and hydrogen mixtures.

Of lesser large-scale commercial importance, but of some chemical interest, is the degradation of aliphatic compounds containing a double bond or alcoholic hydroxyl group. Carboxylic acids will undergo either dehydrogenation or decarboxylation on irradiation, but both effects will not occur in one molecule.

$$CH_3 \cdot (CH_2)_x \cdot CH_2 \cdot COOH \xrightarrow{\alpha} CH_3 \cdot (CH_2)_x \cdot CH_3 + CO_2 \qquad (7.48)$$

and

$$CH_3(CH_2)_x \cdot CH_2 \cdot CH_2(CH_2)_y \cdot COOH \xrightarrow{\alpha} CH_3(CH_2)_x CH{=}CH(CH_2)_y COOH \qquad (7.49)$$

Amino acids and other compounds containing amino groups are deaminated in air-free solutions and ammonia is produced. A few of these reactions are now considered in more detail.

Polymerization of Ethylene

Ethylene was first polymerized at 77°F and about 1 atm pressure by Lind, Bardwell, and Perry[58] using alpha-particle radiation from radon to initiate the reaction. The products were liquids reported to be olefins of higher molecular weight, and also gases, reported to be hydrogen and methane. Ethylene has also been polymerized by irradiation with ultraviolet light. LeRoy and Steacie[59] reported that the metal sensitizers cadmium, zinc, and mercury accelerated the polymerization of ethylene irradiated with ultraviolet light and simultaneously caused some dehydrogenation to acetylene. Their study covered temperatures from 77 to 572°F and pressures from 15 mm to 400 mm Hg absolute. Danby and Hinshelwood[60] investigated the polymerization of ethylene by ultraviolet light at 572°F and pressures to 400 mm Hg absolute, as sensitized by acetone, by diethyl ketone, and by acetaldehyde.

Ethylene has been polymerized with gamma radiation by Hayward and Bretton et al.[61,62] at pressures from 0.5 to 21 atm and at temperatures ranging from room temperature to 460°F. Both liquid and solid products were obtained, usually yellow or brown in color.

The effect of gamma radiation on the polymerization of ethylene was also studied by Lewis et al. (Refs. 31, 41, and 63 to 65) at room temperature and at varying pressures and radiation dosages. Bray et al.[63] reported the effects of varying the temperature of the polymerization reaction on the rate of the reaction and the nature of the polyethylene product. Lewis[65] irra-

diated ethylene with Co^{60} gamma radiation at room temperature and at pressures of 250 to 1600 psi in a stainless-steel pressure vessel used as the reaction vessel.

A white, solid polyethylene product was obtained in quite small yields until the system had received a dose of about 0.5 Mrep, after which the yield increased rapidly to a G value of about 2500 moles reacted per 100 ev for a dose of about 3 Mrep. In this system the yield also is approximately equal to 2500 moles reacted per metric ton of reactants. The yield remained nearly constant up to doses of 7 Mrep, the highest dose given.

Various types of polymer were obtained as a result of differences in treatment and were evaluated with respect to physical properties. Molecular weights of the polymers were estimated from viscosity measurements on solutions and melts. Ultimate tensile stresses and melting points were investigated as a function of radiation yield.

Anderson, Bray and Martin[66] reported a continuation of the work of Lewis which covered a temperature range of 6 to 220°C and a pressure range of 50 to 100 atm. White, curdy, solid polymers were formed at reaction temperatures from 6 to 130°C and a white wax formed at about 160°C. The solid and waxy products were measured on a melting point bar; because each product is a composite of polymers of varying molecular weights, a range of melting points was found. The points at which melting could first be detected have been plotted as a function of the reaction temperature in Fig. 7.6.

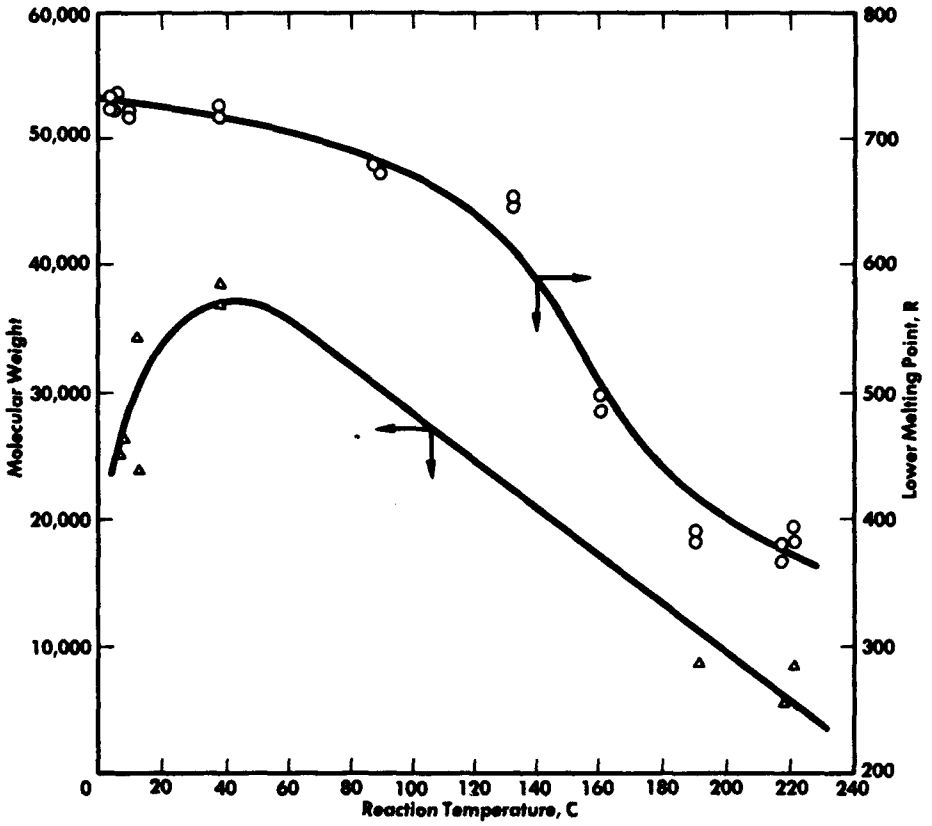


Fig. 7.6—Melting point and molecular weight of polyethylene as a function of reaction temperature.[66]

Melt viscosities of solid products were obtained in a parallel-plate plastometer at about 137°C. Viscosities of liquid products were determined in a modified Ostwald pipette. Approximate molecular weights were estimated from the viscosities and are shown in Fig. 7.6 as a function of reaction temperature.

Solid products produced in appreciable quantity were pressed in a one-compartment mold at 1500 psi and 150°C for 5 min. Figure 7.7 shows the densities as a function of reaction tem-

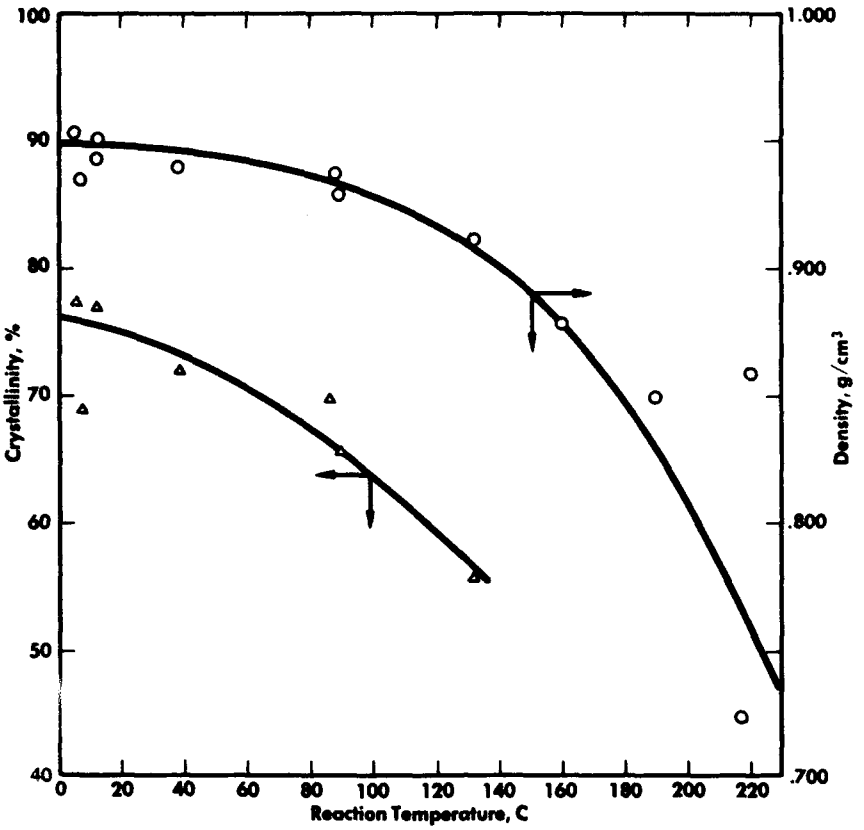


Fig. 7.7—Density and crystallinity of polyethylene as a function of reaction temperature.[66]

perature. Crystallinities or the degree to which the molecules are arranged in parallel positions were determined from the densities, and these are also shown in Fig. 7.7.

Anderson et al.[66] concluded that a wide variety of polymers of ethylene may be produced in the radiation reaction and that yields are sufficiently high for the process to be considered for commercial application.

Cross-linking Polyethylene

One important commercial application of irradiation techniques lies in improving the physical qualities of polymers. Physical changes are likely to occur for moderate dosages only if the polymer is linear or branched. These changes are caused by the rearrangement of the bond structure. Before irradiation, the polymers are bound by weak Van der Waals forces and owe their solid properties either to the stiffness of the entangled chains which prevents their slipping past one another or to regular side chains which allow the molecule to crystallize. When heat is applied, the rigidity of the chains falls, allowing them to slip, and the crystalline regions melt. In both cases melting and flow occur quite readily. It has been stated before that polymers of the formula $(-CH_2 \cdot CHR-)_n$ will cross-link on irradiation, whereas polymers with a tertiary carbon atom degrade. This is thought to be caused by steric hindrances between the side chains. Polyethylene shows some degradation, although the cross-linking reaction predominates.

Polyethylene, as usually prepared with high temperature and pressure, consists of a highly branched polymer so that the crystals present are very limited in size. Melting occurs at temperatures up to 115°C. If polyethylene is prepared at lower temperatures, the crystalline regions are larger and melting takes place up to 135°C. After melting the polymer flows, in both cases, as a plastic fluid.

Charlesby,[67] in extensive studies on irradiated polyethylene, has shown that after a dose of 50 to 100 Mrad, the extent of cross-linking is still low—only about 1 per cent of the mono-

597082 O-61—15

mer units are cross-linked. The physical behavior is the same when cold, but on heating the irradiated polymer remains cross-linked and has rubber-like properties instead of melting and flowing. The tensile strength and elastic modulus are increased at these higher temperatures by increasing the radiation dose responsible for the cross-linking.

Charlesby[67] gives an expression which permits the calculation of the radiation dose required to produce a given value of the elasticity. Higher radiation dosages eliminate the crystallinity (even at room temperatures) producing rubber-like properties, but making the material somewhat brittle. At doses of 1000 Mrad and over, irradiated polyethylene becomes a glass-like substance.[67]

The improved mechanical properties of polyethylene at higher temperatures allow it to be used for many new purposes. Wire covered with irradiated polyethylene can be soldered without melting and shrinking the wire insulation which occurred with nonirradiated polyethylene.[68] The hot polymer softens but keeps its shape and does not flow. Figure 7.8 shows soldering of the irradiated-polyethylene-covered wire.

Another important application is the insulation of transformer windings. Besides the fact that the polyethylene tape will not melt, the dielectric strength of polyethylene at 100 to 200°C is very much improved by irradiation[67] as shown in Fig. 7.9.

Several new commercial applications of irradiated polyethylene are now possible. Heat sealing of packets which previously had to be carefully controlled to prevent melting is now possible.

The rubberlike property of irradiated polyethylene is used to produce the so-called "memory" effect. Irradiated tubing of small diameter can be heated, dilated to a larger diameter, and held at the larger diameter by chilling. However, reapplication of heat permits the formed polymer to return to its original size. Thus a cylinder of dilated irradiated polymer slipped

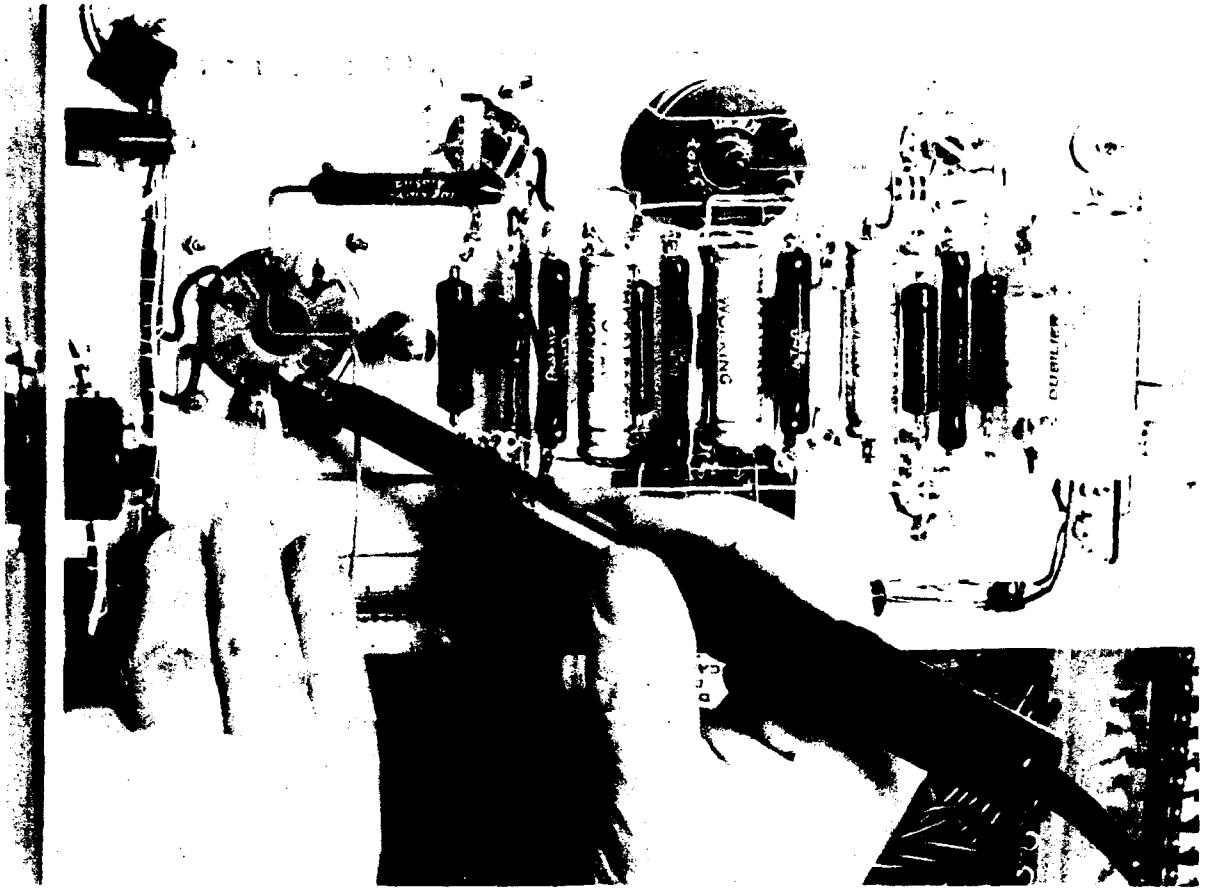


Fig. 7.8—Soldering of irradiated polyethylene-covered wire. (Courtesy of A. Charlesby, Tube Investments Ltd.)[68]

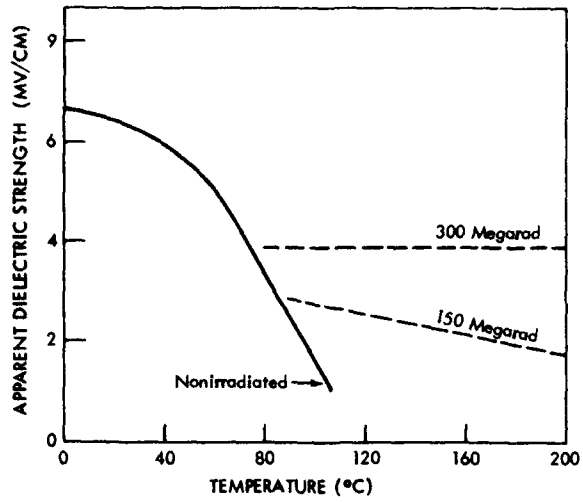


Fig. 7.9 — Dielectric strength of polyethylene irradiated with 2-Mev electrons.[67]

over an object such as the egg will shrink and tightly surround the object if the temperature is raised slightly. This technique holds much promise for packaging in the food industry.

Vulcanization of Rubber

Free-radical reactions are quite common among organic molecules with unsaturated bonds — for example, ethylene. The unsaturated bonds in these compounds are generally quite reactive unless the molecular structure is such that the bond is shielded by bulky side chains, making it difficult for another molecule to attach itself. However, the double bonds present in the long-chain rubber molecules are capable of further interaction if energized.

This interaction between chains is known as vulcanization and results in the cross-linking of the large rubber molecules. The energy necessary for activation can be supplied in several forms. Steam produces good results when used with activators and catalysts. The number of cross-links required for good vulcanization is relatively small, of the order of one link per hundred monomer units. Too many cross-links destroy the property of high elasticity; too few give excessive plastic flow and permanent set.

Preliminary investigations[69,70] indicate that rubber, when exposed to gamma radiation, is activated enough to cross-link at the unsaturated bonds. The exact mechanism by which radiation activates the molecule is not well understood. It has been hypothesized that locally produced high temperatures activate the unsaturated bonds, leading to polymerization via a free-radical type of reaction. Side reactions such as cyclization and chain breakage will occur to a small extent in all kinds of vulcanizing.

Vulcanizing by gamma radiation appears to be unaffected by the presence of sulfur and activators, both of which are required when heat alone is used. The bonding produced by radiation does not need the sulfur link commonly present in thermally vulcanized rubber. Carbon-to-carbon linkages can result, producing a material with different properties which are yet to be thoroughly evaluated. Radiation also permits grafting other desirable side chains to the rubber molecule, favorably modifying the properties. Copolymers of rubber and polyethylene may be possible. Radiation-induced reactions have been observed to take place at single-bonded carbon atoms as well as at the more reactive double bonds. The numerous products which such a treatment could conceivably produce have yet to be fully investigated.

One disadvantage of irradiation vulcanization is that the degree of cure for maximum tensile strength may require a radiation dose[67] of about 40 Mrep. The cost for such a large dose is too high to be justified except for the production of valuable new properties. Rubber with different elastic properties in two directions can be produced by irradiation of the rubber while it is held in a stretched condition. This property might be of interest for special ap-

plications. For silicone rubbers where the additional cost of radiation treatment can be recovered more readily, the absence of vulcanizing agents may be an advantage.[67]

The variation in physical properties of uncompounded natural rubber with gamma-radiation dose is shown in Fig. 7.10. Tensile strength, elongation, and modulus are plotted against radiation dose. With a radiation dose of 10 Mrep, tensile strengths of about 1900 psi have been

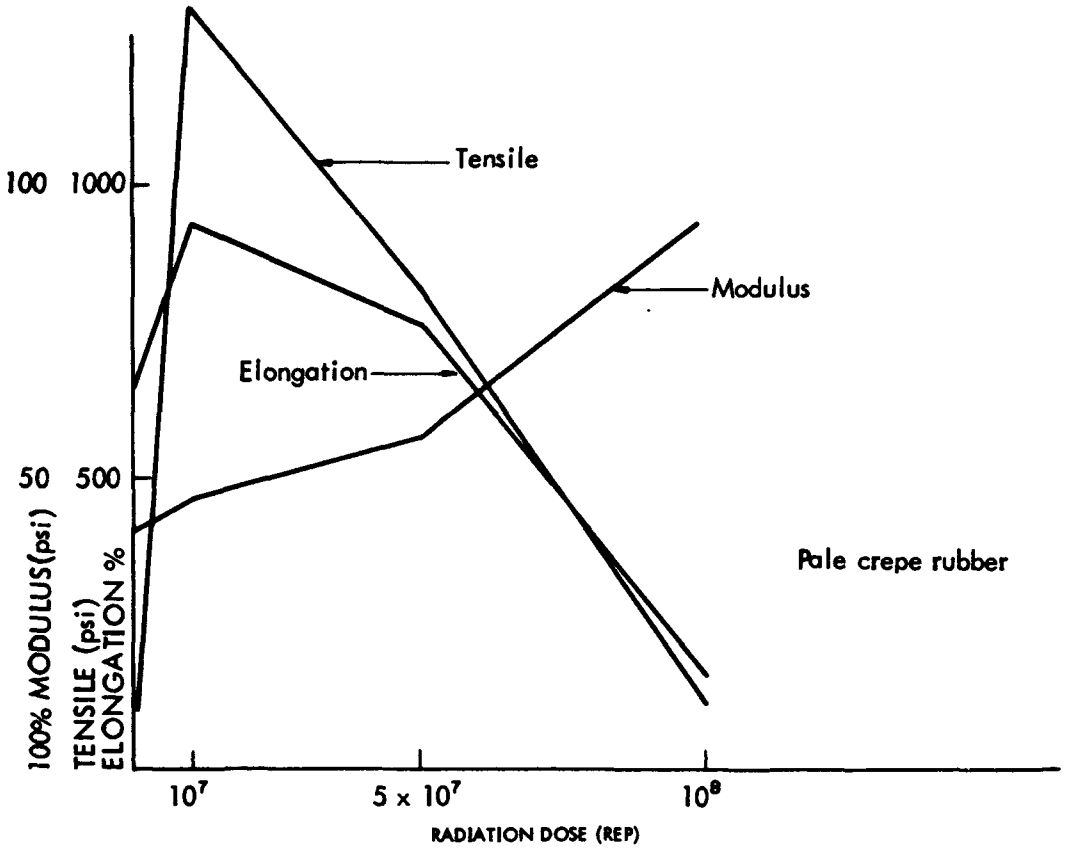


Fig. 7.10—Physical properties of raw rubber vulcanized with gamma radiation.[69]

reached. This is considerably lower than 3500 to 3900 psi tensile strengths resulting from the conventional methods of vulcanization. However, the absence of sulfur and other chemicals in the gamma-radiation vulcanization may compensate for the low tensile strength in some applications. It would be expected that carbon-carbon cross-links would be inherently more stable than the sulfur linkage since the energy of the C–C bond is 58.6 kcal/mole and that of a S–S bond is only 27.5 kcal/mole.

Figure 7.11 illustrates the crystallinity induced by irradiating pale crepe rubber with a dose of 10 Mrep. On the left is the X-ray-diffraction pattern of the nonirradiated amorphous rubber and on the right is the diffraction pattern of the irradiated control sample.[69]

Additional data on the tensile strength and percentage elongation of natural rubber, GRS, Buna N, and neoprene as a function of radiation dose are given by Jackson and Hale.[70] These investigators concluded that irradiated elastomers appear to be modified in a manner closely related to those chemically vulcanized in many respects, except that the modifications in most cases provide elastomers having improved aging characteristics. One exception is the case of vulcanizing polyisobutylene elastomers; this has proved impossible with radiation since, as discussed previously, polymers of this type readily depolymerize at relatively low dosages of gamma radiation.

Jackson and Hale[70] point out that many experimental polymers which resist vulcanizing by chemical means may be readily vulcanized by this process. A specific example of a polymer

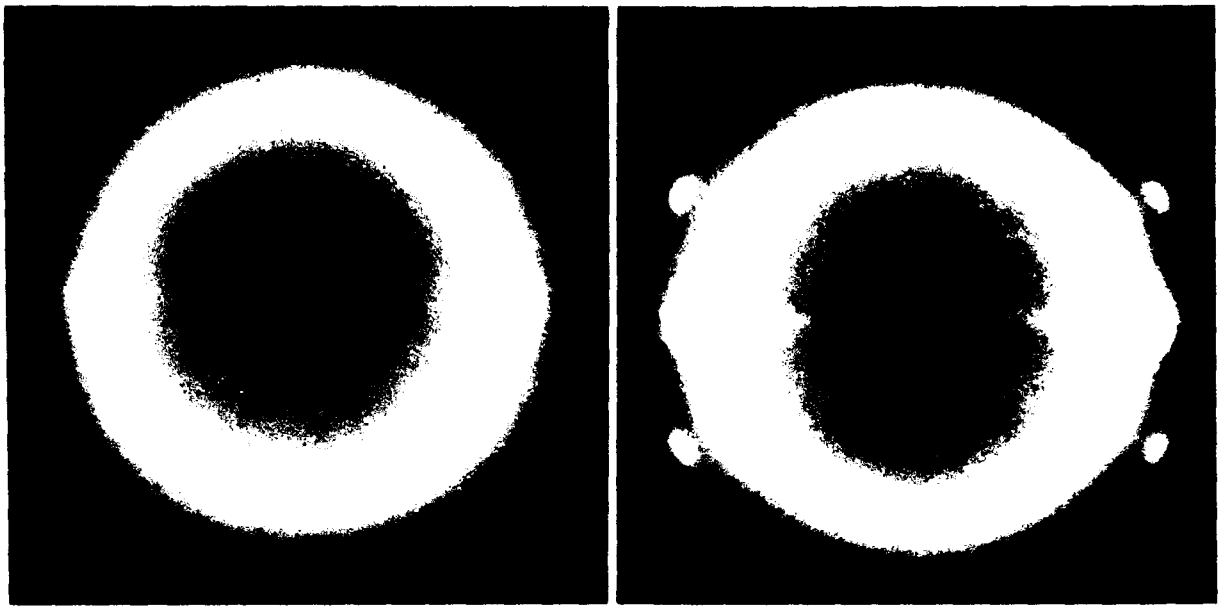


Fig. 7.11—Left: x-ray diffraction pattern of nonirradiated crepe rubber stretched to 500 per cent elongation. Right: x-ray diffraction pattern of irradiated (10^7 rep) crepe rubber equally elongated. (Courtesy of S. Gehman, Goodyear Tire and Rubber Co.)[69]

Table 7.5 — COMPARATIVE HEAT-AGING PROPERTIES OF GAMMA-RAY VULCANIZED AND CHEMICALLY VULCANIZED N5400 ELASTOMERS[70]

Gamma-ray vulcanized		Chemically vulcanized	
N5400	100 parts	N5400	100 parts
HAF	35 parts	Stearic acid	3 parts
Gamma-ray dosage	1×10^7 r	Magnesium oxide	2 parts
		Ethyl zimate	0.75 parts
		Cure 20 min at 400°F	
	Original Properties		
Tensile	1330 psi		1140 psi
Elongation	170%		200%
Shore A hardness	60		60
	Dry Air Aging at 350°F		
Embrittlement after 36 hr		Embrittlement after 8 hr	
	Properties After Aging 72 Hr in MIL-L-7808 Engine Oil at 350°F		
Tensile	575 psi		
Elongation	60%		
Shore A hardness	85	Embrittlement	

of this type is shown in Table 7.5. The response of N5400 elastomer (experimental acrylate developed by Monsanto Chemical Company) to irradiation is shown to be effective after relatively low dosage levels despite the fact that it strongly resists chemical vulcanizing. A chemical vulcanizate was prepared after considerable difficulty for comparison with the irradiated product.

Referring to Table 7.5, the comparative dry-heat aging data at 350°F between the two types of vulcanizates indicate a complete loss of rubberlike properties (embrittlement) of the chemically vulcanized N5400 elastomer in 8 hr while the gamma-ray vulcanized product reached a similar condition in 36 hr. The irradiated product showed better resistance to aging in oil at 350°F; the oil used was a synthetic-base diester oil. The chemical vulcanizate exhibited a complete loss of rubberlike properties in 24 hr.

Table 7.6 lists data on a buna N (Hycar 1001) compound irradiated to a dosage of 10^7 r. Comparative dry-heat aging data with a chemically vulcanized compound indicated a complete

Table 7.6 — COMPARATIVE HEAT-AGING PROPERTIES OF GAMMA-RAY VULCANIZED AND CHEMICALLY VULCANIZED HYCAR 1001 ELASTOMERS[70]

Gamma-ray vulcanized		Chemically vulcanized	
Hycar	100 parts	Hycar 1001	100 parts
Philblack A	35 parts	Zinc oxide	5 parts
Gamma-ray dosage	1×10^7 r	Stearic acid	1.5 parts
		Sulfur	1.5 parts
		Altax	1.5 parts
		Philblack A	35 parts
		Cure 20 min at 310°F	
Original Properties			
Tensile	2640 psi		2900 psi
Elongation	200%		350%
Shore A hardness	75		84
Properties After Aging 24 Hr in MIL-L-7808 Engine Oil at 350°F			
Both samples brittle; however, degree of brittleness substantially less in the irradiated vulcanizate on the basis of 180-degree flat-bend test.			

loss of rubberlike properties in both compounds after air aging 24 hr at 350°F. However, the irradiated vulcanizate was substantially less brittle on the basis of a 180° bend test.

Brownell et al.[71] have reported studies on the feasibility of the use of a multipurpose reactor in a rubber tire plant (see Chap. 4).

Copolymerization of Various Unsaturated Hydrocarbons with Sulfur Dioxide

Bray et al. made a study of copolymerization reactions of sulfur dioxide with ethylene, propylene, butene-1, butene-2, isobutylene, hexene-1, descene-1, dodescene-1, 1,3-butadiene, and cyclopropane.[72,73] This work was further extended[55] by Yemin and Kothary to include allyl bromide, allyl chloride, hexane-2 and 2-methyl pentene-1. Figure 7.12 shows data on the yield of the copolymer of propylene and sulfur dioxide at room temperature.[72]

The reaction between sulfur dioxide and the olefins proceeds under additive catalysts such as light of given wavelength, peroxides, nitrates, and other oxidizing agents.[74] The reaction for copolymerization of sulfur dioxide with olefins may be represented as

$$nR_2C{=}CR_2 + nSO_2 \rightarrow \left[-\overset{\overset{O}{\|}}{\underset{\underset{O}{\|}}{S}}-\overset{R}{\underset{R}{C}}-\overset{R}{\underset{R}{C}}-\right]_n \qquad (7.50)$$

Bray conducted the majority of reactions at a temperature of 0°C, but in the case of hexene-1, temperature was varied from −77.3 to 46.3°C. The radiation intensity was varied

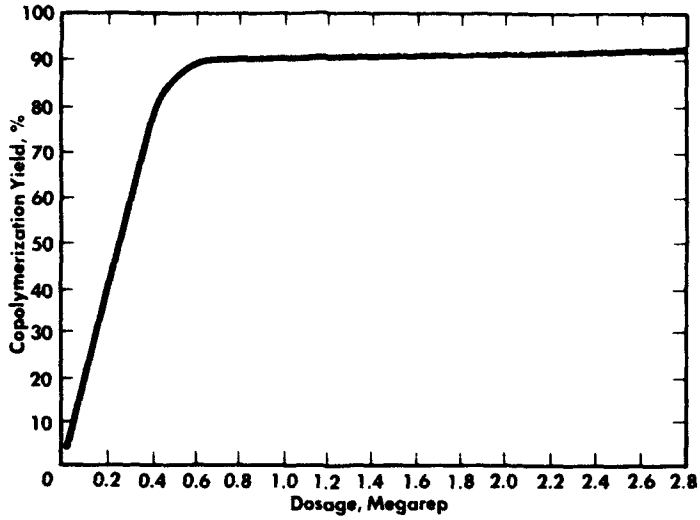


Fig. 7.12—Propylene–sulfur dioxide copolymerization at room temperature.[72]

from 3.58 to 987 krep per hour. The reaction was usually carried out with equimolar proportions of hydrocarbon to sulfur dioxide but concentration effects were studied in some runs.

It was found that melting or decomposition points, densities, and tensile strengths of the copolymers were somewhat higher than that of copolymers produced by other means. The values of these properties decreased with increasing molecular weight of the hydrocarbon reactant. Sulfur analysis showed that equimolar amounts of hydrocarbon and sulfur dioxide combined in the reactions. Molecular weights estimated from solution viscosity measurements were larger than previously reported.

Correlations of kinetic data were also reported with a proposed mechanism of the reaction. In the case of hexene-1–sulfur dioxide reaction, the rate was found to be proportional to the radiation intensity to the 0.76 power, and to the concentration of each reactant to the first power at high sulfur dioxide concentrations. At the lower temperatures used the logarithm of the reaction-rate constant varied linearly with the reciprocal of the absolute temperature. Near the "ceiling temperature" of 57°C (above which the reaction does not occur[74]) and at high hydrocarbon concentrations the rate varied more irregularly with intensity, concentration, and temperature.

Polymerization of Acetylene

The polymerization of acetylene in the presence of gamma radiation has been investigated by Bretton et al.[37] It was found that the rate of reaction is independent of pressure and temperature in the ranges 650 to 1400 mm Hg and 80 to 250°F. These results indicate an ion-clustering reaction mechanism. At a given total pressure, oxygen and nitrogen act as desensitizers for the reaction, while acetone acts as a sensitizer. The product is a solid.

Numerous other systems have been investigated such as the synthesis of butadiene-styrene elastomers.[72]

Chlorination of Benzene

Chlorine may react with an aromatic hydrocarbon in three ways: add directly on to the benzine ring, replace hydrogen on the benzene ring, or replace hydrogen in alkyl groups attached to the benzene ring.[75] The gamma isomer of hexachlorocyclohexane has only recently become commercially important as an insecticide.

The first use of radiation to promote the addition of chlorine to benzene was claimed by Loiseau[76] in a patent dated 1924. He used X rays to induce the reaction between benzene and chlorine. By regulating conditions, the substitution reaction could also be made to take place.

In 1930 Alyea[77] carried out the reaction with a mixture of liquid benzene, chlorine, and radon gas, the last of which supplied a source of alpha particles and produced $C_6H_6Cl_6$. Additional work was carried out in 1934 on the use of active chlorine in chemical reactions by Willey and Foord.[78] These investigators passed a chlorine gas stream through a silent electrical discharge tube, then into the benzene solution.

Present-day commercial methods are based principally on the use of ultraviolet radiation[79,80] to promote the reaction; however, gamma radiation might be used to advantage in place of ultraviolet light.[81,82]

The reactions of chlorine with benzene, toluene, xylene, mesitylene, and naphthalene in the presence of gamma radiation were investigated by Harmer et al. (Refs. 31, 63, 64, 66, and 75). It was found that gamma radiation was an outstanding promoter for adding chlorine to benzene. The reaction proceeded with such speed in the 200-curie source that it was necessary to dilute the reactant benzene with carbon tetrachloride to control the heat of the reaction. Radiation yields were of the order of 19,000 to 114,000 molecules reacted per 100 ev absorbed by the system. The product of the radiochlorination of benzene was found to be the several isomers of hexachlorocyclohexane. The commercially important gamma isomer occurred to the extent of about 11 to 13 per cent.

Figure 7.13 shows a schematic diagram of the apparatus used in these experiments. The reaction vessel consisted of a 25-mm glass tube long enough to extend completely out of the source. The assembled reaction vessel was inserted in a steel jacket placed in the 200-curie Co^{60} source and the gas-feed lines connected.

The glass reaction vessel was filled with benzene and carbon tetrachloride, after which it was lowered into the Co^{60} gamma source and purged of air with nitrogen. Coolant was passed through an outer jacket of the vessel. Chlorine was bubbled into the reaction mixture until the first traces of HCl or Cl_2 appeared in the off-gas from the reaction vessel and then the chlorine was regulated at a rate so as to eliminate HCl or Cl_2 in the off-gas. The reaction was usually conducted until all the benzene was consumed.[64] The reaction velocity is due to the combined temperature and radiation effects.

Chlorination of Toluene, Xylene, and Mesitylene

In 1867, Pieper[83] treated toluene with excess chlorine. Analysis of the crystals which separated from the solution upon standing revealed the best empirical formula to be $C_7H_6Cl_8$. It appeared that addition and substitution took place on the same molecules.

Smith[84] in 1951 reported that a mixture of methyl hexachlorocyclohexane and chloromethyl hexachlorocyclohexane at a concentration of 0.5 per cent in gypsum dust killed cat and rat fleas more effectively than the insecticide DDT.

The reaction of the radiochlorination of toluene as reported by Harmer et al.[75] proceeded rapidly but more slowly than the benzene ring, although substitution also occurred. The addition product was separated from other reaction products in good yields, and appeared to be similar to material previously reported to be formed in minor amounts during the photochlorination of toluene. The addition product was converted to known compounds. A sketch of the equipment used by Harmer is shown in Fig. 7.14.

A study of the initial reaction rates of the radiochlorination of toluene revealed that the addition and substitution reactions were both dependent upon approximately the 0.5 power of the radiation intensity, within the range of 7.9 to 28.4 krep per hour. The initial rate of addition was dependent upon the square of the chlorine concentration, while the initial rate of substitution was dependent upon the square root of the chlorine concentration, between 1.8 and 3.0 moles/liter of chlorine. The effect of temperature could not be determined with accuracy, but both the addition and the substitution reaction rates were increased by increasing temperature, after corrections for the changes in chlorine solubility had been applied. Long reaction chains were indicated in the toluene reaction, and the rapid addition produced by the gamma irradiation appeared to be unique compared to other reported reactions of toluene. The partial kinetics of the reaction can be summarized by

$$R = \frac{dc}{dt} = k_a I^{0.5} (Cl_2)^2 = k_s I^{0.5} (Cl_2)^{0.5} \qquad (7.51)$$

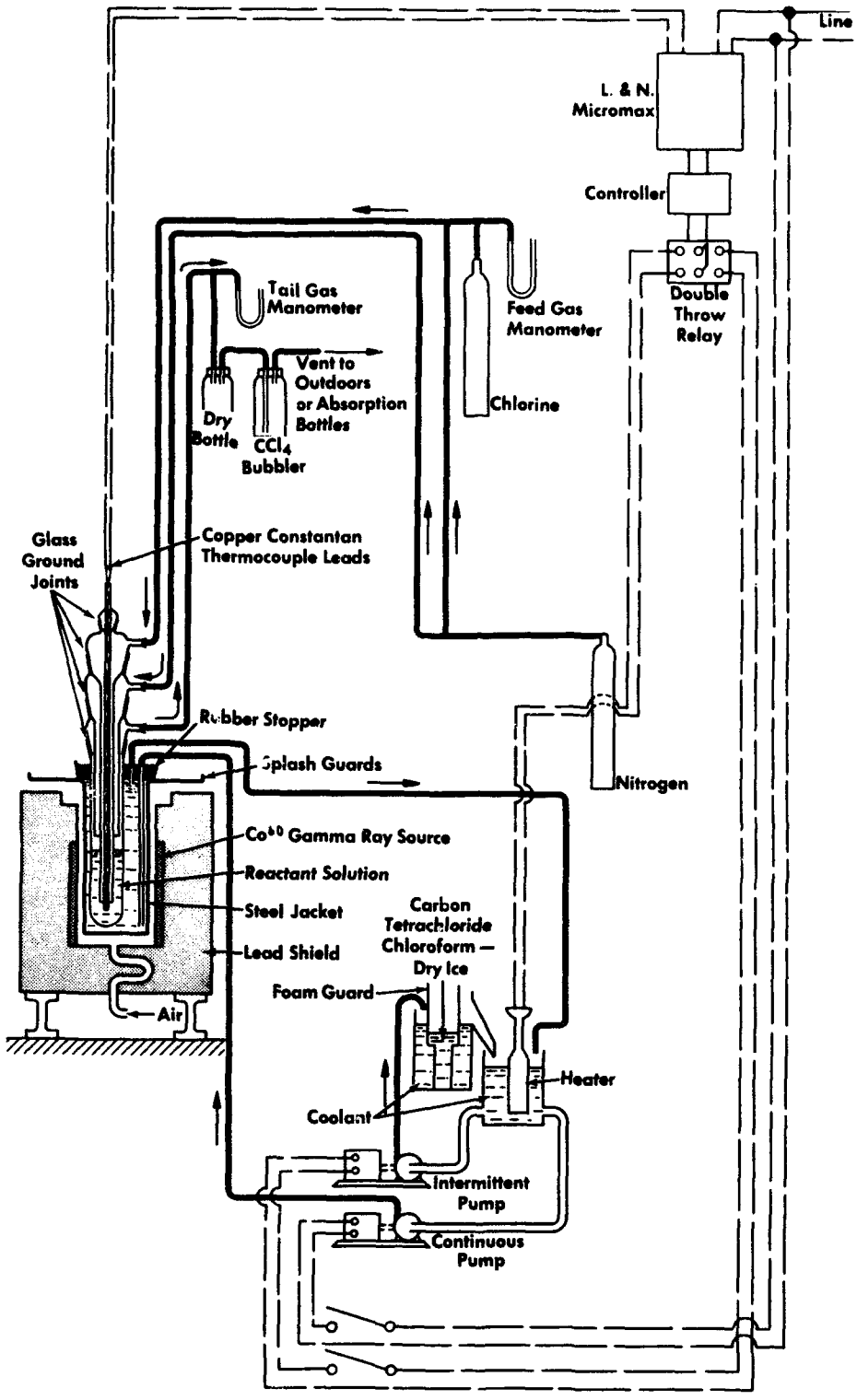


Fig. 7.13—Schematic diagram of apparatus for chlorination of aromatic compounds in 200-curie source.[75]

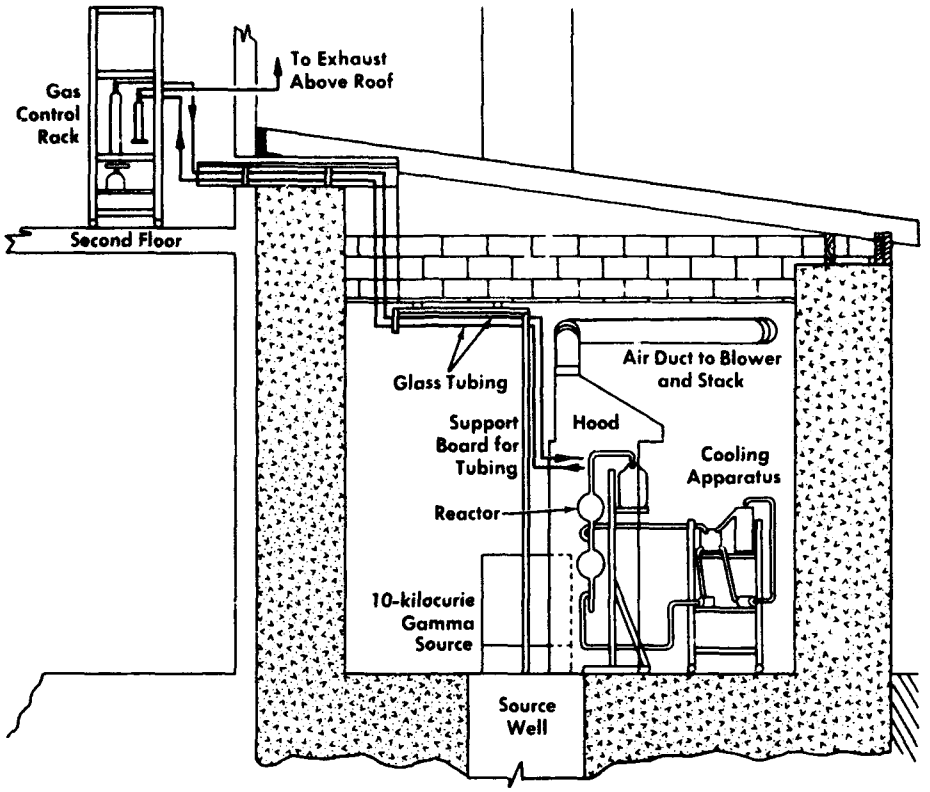


Fig. 7.14—Sectional view of radiation cave with chlorination apparatus.[75]

The equation is expressed in terms of consumption of chlorine, and k_a and k_s represent the reaction velocity constants for addition and substitution reactions.

Chlorobenzene was found to undergo a rapid addition of chlorine under gamma irradiation. Benzyl chloride, benzal chloride, and benzotrichloride, however, showed no reaction under the same conditions. Furthermore, the addition of small amounts of benzyl chloride to benzene or toluene produced a marked inhibition of the reaction when the mixtures were treated with chlorine under irradiation. Since a similar effect is not observed in the analogous photochemical reaction, the inhibition of the radiochlorination is very unusual. Oxygen was also found to inhibit both the addition and the substitution reactions. Possible explanations of the differences between the photochlorination and radiochlorination of the aromatic compounds are discussed by Harmer.[75]

Harmer et al.[66] also reported that xylene and mesitylene reacted vigorously with chlorine in the presence of gamma radiation. In each case the addition of six chlorine atoms to the ring and alkyl substitution appeared to be the primary reactions. The ratio of addition to substitution was different from that obtained in chlorination reactions without radiation. In general this ratio appeared to be increased by radiation and decreased by increasing the temperature. The radiation yield reported as G value is given in Table 7.7. Radiation had little effect on the reaction with ethyl benzene. Whether this is due to inherent inactivity or to inhibiting impurities is not known.[66]

A summary of some more results on radiation yields for various aromatic hydrocarbons in the presence of gamma radiation is given in Fig. 7.15.

In cases of chlorination of some of the compounds described above, where reaction is fast enough, and the addition product is desired, use of radiation may be attractive.

Oxidation of Ethylene

Henley et al.[85] reported a study of the oxidation of ethylene in the presence of water employing ionizing radiation as a catalyst. The products included acetaldehyde, ethanol, acetic

Table 7.7 — CHLORINATION OF AROMATIC COMPOUNDS IN GAMMA RADIATION FIELD[66]

Run	Aromatic chlorinated	Temp. reaction, °C	Dose rate, krep/hr	Chlorine addition, moles/liter	Chlorine substitution, moles/liter	Av. G* addition	Av. G* substitution
1	30% benzene	20	61.0	35.20		234,000	
2	20% benzene	20	61.0	32.60		252,000	
3	10% benzene	20	61.0	33.30		90,000	
4	30% benzene	−10	61.0	13.70		90,000	
5	10% benzene	−10	61.0	32.70		57,000	
6	Toluene	20	14.0	17.42	7.54	523,000	226,000
7	Toluene	−5	14.0	14.12	6.59	900,000	420,000
8	Toluene	35	14.0	10.35	6.12	812,000	480,000
9	Toluene	20	None	3.23	0.85		
10	Xylene	20	13.7	5.90	3.48	462,000	272,000
11	Xylene	−5	13.7	7.45	2.94	448,000	177,000
12	Xylene	20	None	3.20	2.63		
13	Mesitylene	20	13.7	4.20	15.10	210,000	827,000
14	Ethyl benzene	20	13.7	1.81	0.47	137,000	35,000
15	Ethyl benzene	20	None	1.61	0.13		

*G, (molecules reacted)/(100 ev absorbed) based on chlorine.

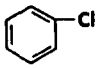
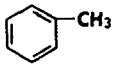
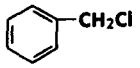
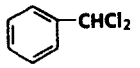
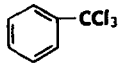
Reactant	Structural Formula	G, Molecules reacted per 100 ev of gamma irradiation	Molecules affected per original molecule affected
Benzene		85,000	28,000
Chlorobenzene		43,000	14,000
Toluene		17,000	5,000
Benzyl chloride		Small	——
Benzal chloride		Small	——
Benzo trichloride		Small	——

Fig. 7.15 — Radiation yields for the addition of chlorine to some aromatic compounds.[75]

acid and hydrogen peroxide, with the acetaldehyde predominating. At total radiation doses of less than 1 Mrep the over-all process was represented by

$$CH_2CH_2 + O_2 + H_2O \xrightarrow{\gamma} CH_3CHO + H_2O_2 \qquad (7.52)$$

At higher doses ethanol and acetic acid began to form.

The reaction was carried out by bubbling a 1 : 1 M mixture of ethylene to oxygen into water at an average pressure of about 100 psi. The chemical reactor was located in the 180,000 r/hr field of a Co^{60} source and the temperature range was 25 to 61°C.

Upon exposure to radiation, acetaldehyde began to form, continuing until a maximum concentration of about 0.24 per cent was obtained after a total dose of about 1.6 Mrep. After this point the acetaldehyde concentration fell off, and the acetic acid and ethanol concentrations began to rise. At a total dose of about 9 Mrep the ethanol concentration was about 0.004 per cent, the acetaldehyde concentration was about 0.0045 per cent, and the acetic acid concentration was up to 0.0055 per cent. The maximum G value for the acetaldehyde formation was about 60 and corresponded to a total dose of 0.5 Mrep.

Cracking of Hydrocarbons

Radiation-induced cracking of pure hydrocarbons and mixtures was investigated by Lucchesi, et al.[86] at temperatures of 320 to 510°C. Product distributions were similar to those obtained with thermal cracking. The yields of product increased with increasing temperature and increasing radiation intensity. The mechanism of reaction is discussed. Hardwick[87] reported on the effects of radiation on petroleum.

RADIATION DAMAGE TO MATERIALS OF CONSTRUCTION

The continued irradiation of organic compounds may change their physical properties and ultimately will decompose them. In considering materials for use in reactors, processing equipment, etc., an understanding of the degree of stability of these materials when exposed to radiation is very important. Much research effort has been expended on studies of radiation damage.[88–96] A discussion of some selected results is presented.

ORGANIC MATERIALS

In general, plastics containing the most plasticizer, filler, and other impurities are the least stable under radiation. Harder plastics, such as the phenolics, are the most resistant to radiation. Rubbers tend to become harder owing to increased cross-linking and thus lose their elasticity. Table 7.8 from Sisman and Bopp[91] gives the radiation resistance of some common plastics to reactor radiation. The unit of radiation exposure is thermal-neutron flux times exposure time, or ϕt thermal neutrons per square centimeter. However, the total flux (fast neutron, slow neutron, plus gamma) is directly proportional to the slow-neutron flux. It is seen that mineral-filled phenolics have the greatest resistance to radiation damage and the cellulose, casein, metacrylate, and halogenated polythenes such as Teflon have the least.

Bennett[92] gives a similar listing for plastics exposed to gamma radiation from a Co^{60} source. The data closely parallel those of Table 7.8. The reader is referred to the report by Collins and Calkins[93] for an extensive compilation of radiation damage to elastomers, plastics, and organic liquids.

INORGANIC MATERIALS

The primary way that metals suffer radiation damage is by displacement of atoms in the metal lattice. Displacement may be caused by neutron, proton, or very high energy gammas, such as from a reactor core, but not by gamma radiation from radioisotopes because the gamma energy is too low. Cold-working a metal, which also leads to dislocations in the lattice, may produce hardening of the metal. This same hardening has been found to occur in metals

Table 7.8 — REACTOR RADIATION DAMAGE TO PLASTICS[91]

Plastic	Exposure, 10^{18} neutrons	Radiation effects
Mineral-filled furan and mineral-filled phenolics: Duralon, Haveg 41, asbestos-fiber Bakelite, asbestos-fabric Bakelite, and Karbate	10	Little change except darkening in color
Styrene polymers: Amphenol and Styron 411C	10	Little change except darkening in color
Modified styrene polymer: Styron 475	10	Impact strength and elongation decrease until the same as unmodified styrene polymers
Aniline formaldehyde (Cibonite) and polyvinyl carbozole (Polectron)	10	Tensile strength decreases somewhat
Polyethylene and nylon	10	Impact strength decreases but tensile strength increases; these plastics become so brittle that the corners of the specimen chip off
Mineral-filled polyester Plaskon alkyd	10	Tensile strength and impact strength are decreased about 50%
Unfilled polyesters: Selectron 5038 and CR[39]	5	Develop small cracks; tensile strength decreases
Phenolics with cellulosic fillers: paper-base Bakelite, linen-fabric, Bakelite and Micarta	3	Become brittle, swell, and decrease in impact and tensile strength
Melanine and urea: Melmac, Bettle, Plaskon urea and Plaskon Melanine	2	Tensile strength and impact strength are decreased about 50%
Unfilled phenolic: Catalin	1	Tensile strength and impact strength are decreased about 50%
Vinylidene chloride (Saran B-115) and vinyl chloride acetate (Vinylite)	0.5	Soften, blacken, evolve HCl, and decrease in tensile strength
Casein (Ameroid), methyl methacrylate (Lucite), Teflon, Fluorothene, and the cellulosics: cellulose nitrate (Pyralin), cellulose acetate (Plastocele), cellulose acetate butyrate (Tenite II), cellulose propionate (Forticel), and ethylcellulose (Ethocel R-2)	0.1	Tensile strength and impact strength are decreased about 50%

as a result of irradiation.[94] However, with the irradiation the hardening effect is not as great, but the lattice undergoes a gradual change from an ordered to a disordered structure. All distortions in the lattice tend to reduce thermal and electrical conductivity.

SAE plain carbon steels increased hardness upon irradiation. Stainless steels became harder upon irradiation if they were soft or annealed before it, but did not get significantly harder if they were hard initially. Irradiation may increase the steel yield stress threefold and the ultimate stress by 25 per cent.[96] Nickel-base alloys such as Hastelloy-C, Monel, and Inconel increased in hardness when irradiated. Titanium increased slightly in hardness upon irradiation.

The effect of radiation on corrosion of metals is of great importance because of radiation damage to fuel elements as well as structural members of the reactor. It appears that neutron irradiation of the fuel element materials is more damaging than gamma irradiation; after very high neutron dosages the fuel element components may increase in size and fail. Aluminum corrodes in water and radiation increases the corrosion rate. Radiation increases the corrosion of copper and mild steel samples in an acid bath.

Additional information on radiation chemistry is given in Refs. 95 to 116 and in references for Chap. 4, and on radiation damage to structural materials in Refs. 117 to 132.

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Chapter 8

EFFECTS OF RADIATION ON BIOLOGICAL MATERIALS AND SIMPLE ORGANISMS

This chapter describes some effects observed when biological materials and simple organisms are exposed to ionizing radiations. The discussion covers changes produced in proteins and then the effects of radiation on protein-containing systems such as meat, vegetables, and milk; on carbohydrates, lipids, vitamins, nutrilites, enzymes; and on microorganisms such as bacterial molds and yeasts. Radiation sensitivity of other systems such as viruses and bacteriophages is described, along with the effects of environmental conditions on this sensitivity. Finally, the chapter discusses in detail how radiation affects some insects and their larvae, and various important parasites.

Biological systems have the following basic components: carbohydrates, lipids (fats and oils), proteins, vitamins, and enzymes. Many of these have been separated as pure chemical substances of definite molecular composition. They can be arranged in systems of varying degrees of complexity and enzymes have been isolated in pure crystalline form. Viruses resemble complex enzymes in form but differ from them in that, given a suitable environment, they are capable of the fundamental life processes of self-reproduction and multiplication. Thus enzymes and viruses may be considered biological molecules on the borderline between the nonliving and the living. Microorganisms consisting of only one cell contain many molecules. A multitude of cells make up the more complex organisms of plants, insects, and animals.

EFFECT OF IONIZING RADIATIONS ON BIOCHEMICALS

Gamma radiation will ionize molecules, produce free radicals and excited molecules, and promote certain chemical reactions, as described in Chap. 7. The general nature of some of these reactions has been discussed, but little or no mention has been made of the effect gamma radiation has on complex organic compounds found in biological materials.

DIRECT AND INDIRECT ACTION OF RADIATION

The effect of ionizing radiation on biological material may be divided into two mechanisms: "direct" and "indirect" actions. The direct action is produced by hits on the target area of a molecule or cell as a result of interaction with ionizing radiation, while indirect action is dependent on the interaction of radiation with surrounding water or solvent molecules.

The concept of direct effect as a mechanism of biological action indicates that the molecule undergoing change becomes ionized or excited by interaction with ionizing radiation[1,2]. The comparison of the vital structure of the cell to a target, and radiation to a projectile, led to common expression of direct action as a target theory. This theory is discussed more thoroughly in a later part of this chapter.

The concept of indirect effect was first proposed by Risse[1,4] who stated that some of the effects of radiation might be attributed to the radiolysis of water. The primary products, and

those produced by further reactions, can cause changes such as breakdown of nutrients, formation of undesirable flavor compound, etc. In aqueous solutions, direct effects usually contribute only a small fraction of the total change, even up to concentration of 20 per cent of solute molecules according to Dale[3]. The effect of radiation on water is discussed in detail in Chap. 7. Reactive radicals are formed which possess both oxidizing and reducing ability.

Goldblith[5] has discussed the kinetics of radiation effects on solute molecules, and has compared the direct and indirect effects of radiation. If the reaction product of the solute molecule is not capable of further reaction with radiation-produced excited molecules and free radicals, the reaction is of zero order and hence a linear function (e.g., oxidation of ferrous sulfate). Where the reaction product can react further an exponential function occurs (e.g., oxidation of ascorbic acid). Goldblith has also compared the influence of concentration, temperature, and protection on direct and indirect actions of radiation[5].

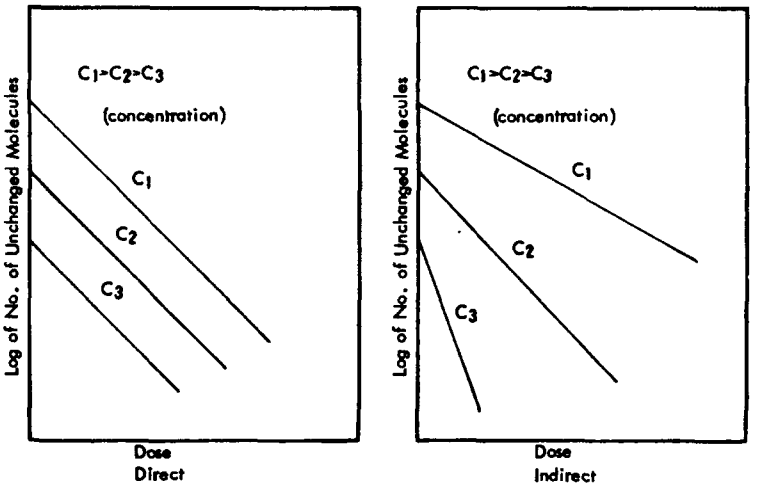


Fig. 8.1 — Function of concentration in direct and indirect action.[5]

Figure 8.1 shows the effects of concentration of solute (or organism). The parallel survival curves are obtained for direct action, with either the single-hit or the multi-hit organisms.

Figure 8.2 shows that temperature is not of importance in truly direct action. However, due to the inhibition of free-radical diffusion, temperature does have an effect in indirect action. Thus, reducing the temperature aids in minimizing undesirable side reaction.

Figure 8.3 shows that the presence of a protector material is beneficial in minimizing damage by direct action but is not effective in protecting against damage by indirect action. There is competition for free radicals and activated molecules by the various solutes and, therefore, one solute may offer "protection" to another. This offers a means of protecting nutrients and flavor compounds, if free-radical acceptors, scavengers, or compounds that more selectively react with free radicals are found, that can be used without producing other side effects. Uri has reported an extensive study on free-radical acceptors[6].

The significance of direct and indirect action with respect to the living cell is described by Hollaender and Stapleton[7].

In addition to the analysis given for the mechanism of indirect action there have been other theories, such as the poison hypothesis, which postulates the formation of poisonous substances of unknown identity in the cell as a result of irradiation[8].

PROTEINS

Proteins are complex molecules produced by living cells; they are composed mainly of amino acids in combination. Ionizing radiation appreciably affects the physical properties of proteins. The changes can be summed up by the term denaturation.

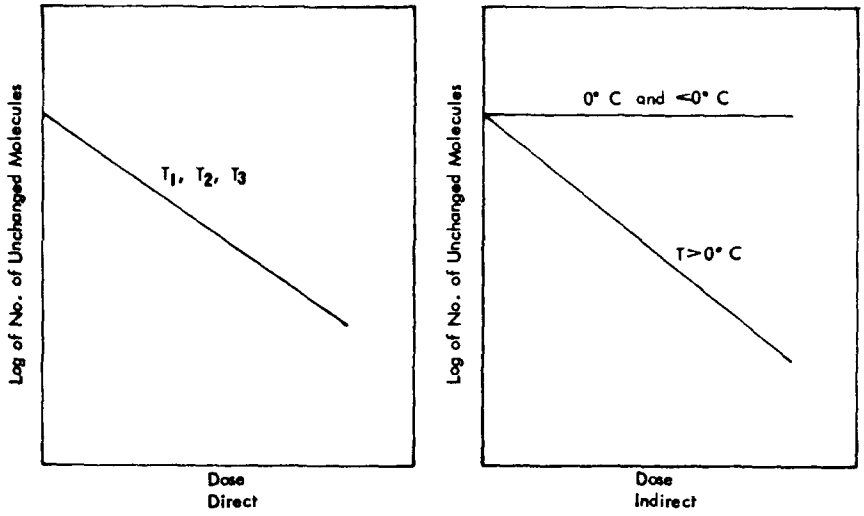


Fig. 8.2 — Function of temperature in direct and indirect action.[5]

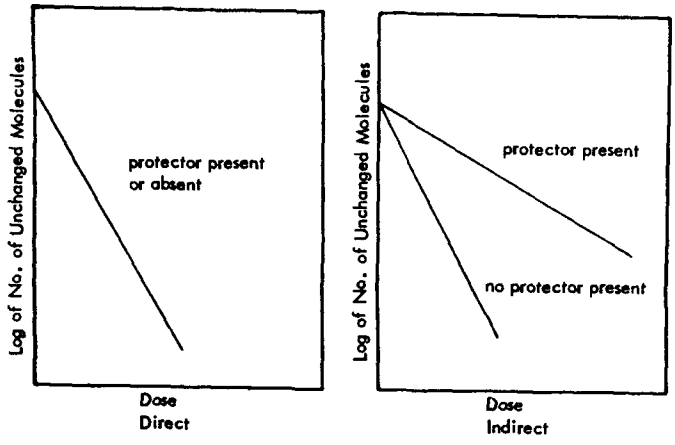


Fig. 8.3 — Function of protectors in direct and indirect action.[5]

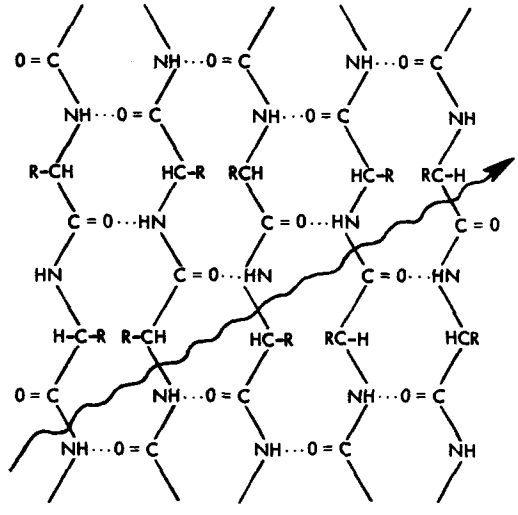


Fig. 8.4 — Modification of protein macromolecule by direct action of radiation (according to Kuzin).[9]

Proteins in the presence of water may be modified either by direct interaction of the radiation with the protein macromolecules or indirectly as a result of free radicals and excited molecules produced in water. Kuzin[9] states that in the direct process the ionizing radiation can simultaneously knock out n electrons, form a polyvalent ion, and so cause the rupture of n bonds in the macromolecule (see Fig. 8.4). He suggests that the indirect mechanism will predominate since "tissue proteins are, because of hydration, virtually permeated with water molecules."

When the blood proteins, hemoglobin and serum albumin, are exposed to ionizing radiation, their original molecular weights change both upwards and downwards. Another blood protein, hemocyanin, splits into two molecules when irradiated. All blood proteins, including fibrinogen, serum albumin, serum globulin and egg albumin, show some degradation plus some polymerization on being irradiated.[10] Polymerization has also been observed in irradiated gelatin, and this is believed to be a permanent cross linking. On irradiation, a clear gel first loses transparency, although it remains translucent. It then forms a permanent jellylike mass which does not melt in hot water. Formation of the permanent gel reduces the original volume of the gel from which clear liquid separates. Irradiated egg albumin[10] shows a decrease in the heat-denaturing temperature of as much as 30 to 40°F.

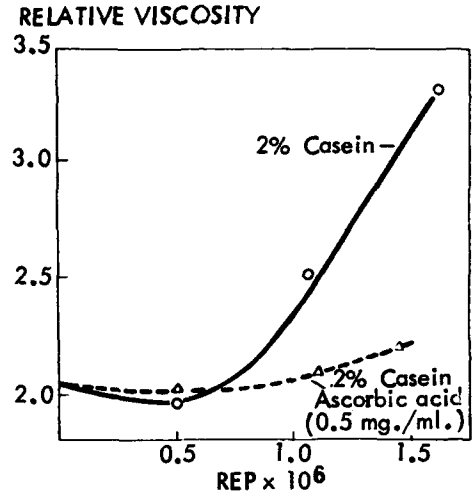


Fig. 8.5 — Effect of irradiation on relative viscosity of casein solution and protective effect of ascorbic acid[11]

Figure 8.5 shows the change in viscosity of casein as a result of irradiation with linear-accelerator electrons[11]. The initial drop in viscosity is due to decrease in molecular size and the subsequent rise to polymerization. Note the protective effect of ascorbic acid on polymerization. An increase in vulnerability to trypsin digestion, the formation of new sulfhydryl groups, and definite changes in electropheric patterns and mobilities were exhibited. Similar changes of viscosity and other properties were observed with irradiated egg albumin.

Irradiation of fibrinogen solutions also produced an increase in viscosity. Sedimentation studies showed a decrease in the concentration of the main fibrinogen component[12]. Drake et al.[13] studied the radiosensitivity of amino acids constituting protein insulin to determine the source of objectionable odors and flavors that often develop in food proteins sterilized by ionizing radiation. One per cent insulin in basic (pH 8.5) and acidic (pH 3) solutions was given 0, 10, 20, and 40 Mrep doses of gamma radiation. Insulin was used because it is available in a nearly pure state.

On analysis by paper chromatography, the irradiated protein was found to have undergone a marked change in molecular weight. Irradiation had destroyed the nitrogen-terminal amino acids, whose irradiation sensitivity is probably due to an irradiation-deamination reaction similar to that occurring in free amino acids. Figure 8.6 indicates reduction of amounts of

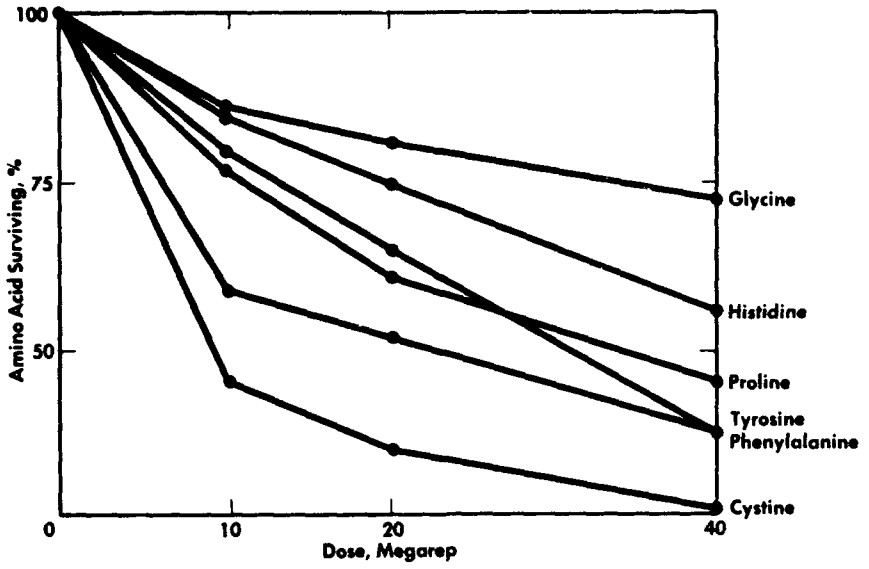


Fig. 8.6 — Amino acid value as percent of control (zero dose) for less resistant amino acids in irradiated 1 per cent insulin in pH 3.0 solution.[13]

amino acids as radiation is increased. There is an increase in per cent alanine and threonine with dose, apparently as a result of the degradation of more complex molecules containing these amino acids.

Biological activity is reported lost when reagents attack phenolic groups, carboxyl-terminal groups and disulfide linkages. The phenolic and disulfide groups were changed or destroyed by irradiation.

Only about 10 per cent of the biological activity of unirradiated insulin injected was "lost". After injecting a biological activity of 25 μc/mg of unirradiated insulin, the activity level actually found was 22.1 μc/mg; but after subjecting a sample to a 10-Mrep dose of radiation the loss was much greater — about two-thirds. The activity level of the injected irradiated insulin was 12.5 μc/mg: that actually found was less than 5 μc/mg.[13]

The amino acids bonded in the protein molecule and shown to be radiosensitive at the lower radiation doses of 10 to 20 Mrep are cysteine, tyrosine, proline, phenylalanine, and histidine. It is interesting that the last four are all ring-containing compounds. Leucine, valine, lysine, and arginine show a significant decrease at the 40-Mrep dose. These nine amino acids (plus possibly isoleucine, which was not included in the analysis, and tryptophan, methionine, and cysteine, which are not present in insulin) are indicated by these studies as possible sources of objectionable odor and flavor when food proteins are given sterilizing doses of radiation.

In addition to studies on irradiated proteins, there have been many investigations of irradiated protein foods. Meats, milk, and proteinaseous vegetables are complex systems containing many different proteins, lipids, and carbohydrates, as well as other biological chemicals and water. Subjecting proteins in these foods to ionizing radiation yields additional information.

Irradiated lean beef was studied by Batzer and Doty[14,15] who reported that radiation dosages of 0.5 Mrep or more reduced the percentage of soluble protein, increased the nonprotein nitrogen compounds, and partially destroyed the glutathione in the meat.

In further study Batzer and Doty[16] reported an increase of hydrogen sulfide and methyl mercaptan, and a decrease of glutathione and glycogen in irradiated meat. The nature and extent of most of these chemical changes are stated to be influenced by the fat content of the meat, the time and temperature of storage prior to irradiation, the irradiation dosage, and the storage time and temperature after irradiation. However, the exact relation of these chemical changes to changes in odor and flavor of cooked meat has not been established.[16]

Table 8.1—THE EFFECT OF GAMMA IRRADIATION ON BEEF[15]

Dosage, Mrep	pH	Glutathione, mg/100 g	Carbonyl, micromoles/g
0	5.68	13.5	12.7
2	5.70	9.0	43.1
4	5.72	6.3	66.0
6	5.80	4.1	86.3
8	5.97	4.5	112.0
10	5.99	4.0	115.0

Table 8.2—NUTRITIVE VALUE OF PROTEINS IN SOME IRRADIATED AND NONIRRADIATED FOODS[17]

Protein source	Test	Condition: Raw	Condition: Irradiated 3 Mrep	Condition: Heat processed
Beef	True digestibility, %	100	100	
	Nitrogen balance, mg/g N intake	578.5 ± 9.8	591.5 ± 21.0	
	Biological value, %	78.0 ± 1.0	78.6 ± 1.5	
Peas	True digestibility, %	92.5	91	92
	Nitrogen balance, mg/g N intake	368	275	354
	Biological value, %	59	50	58
Lima beans	True digestibility, %	73	75	85
	Nitrogen balance, mg/g N intake	106	163	332
	Biological value, %	50	51	68
Milk	Gain/g protein intake	2.74	2.5	
	Nitrogen balance, mg/g N intake	626	506	
	Biological value, %	90	74	
Milk and cystine	Gain/g protein intake		2.89	
	Nitrogen balance, mg/g N intake		661	
	Biological value, %		92	

Table 8.3—EFFECT OF GAMMA IRRADIATION ON AMINO ACID CONTENT IN GELATIN DESSERT POWDER[19]

Amino acid	Concentration, mg/g: Control (0 rep)	Irradiated (3 Mrep)	Irradiated (6 Mrep)
Arginine	9.6	9.6	8.4
Cystine	0.12	0.09	0.09
Glycine	29.9	29.5	30.2
Histidine	0.7	0.7	0.4
Isoleucine	2.6	2.6	2.3
Leucine	2.5	2.5	2.4
Lysine	7.3	6.5	6.4
Methionine	0.98	0.81	0.80
Phenylalenine	2.3	2.1	1.8
Threonine	3.0	3.7	2.6
Valine	4.1	3.6	3.3

Johnson and Metta[17] reported on the effect of a 3-Mrep gamma radiation dose on the nutritive value of protein in raw beef, garden peas, lima beans, milk, and milk with amino acid additions. The results are summarized in Table 8.2.

The gamma dosage of 3 Mrep had no effect on either digestibility or biological value of the protein of raw beef. In the case of the two vegetables, peas and lima beans, there was little difference between digestibility of the raw and irradiated proteins, whereas heat processing increased digestibility. Irradiation caused a loss of 9 per cent in biological value in the peas but not in the beans. Irradiated milk also showed a loss in biological value. This loss was believed to result from a deficiency in an essential amino acid. Various amino acids were added to the irradiated milk; it was found that cysteine restored the biological value but lysine and histidine had no effect. The conclusion was that biological value loss in irradiated milk is due to decreased availability of cysteine either by destruction or chemical combination.

Radiosterilization has no effect on the metabolizable energy contributed by proteins and other components of the diet. An 18-month feeding experiment with female rats showed that radiosterilized beef and flour are satisfactory for optimum growth and maintenance.[18]

Teply and Kline[19] reported the effects of 3- and 6-Mrep doses of gamma radiation on the amino acid content of gelatin dessert powder, as shown in Table 8.3. Note the slight but definite decrease in percentage for almost all amino acids as a result of irradiation.

Glycoproteins, such as collagen from connective tissue, are modified by ionizing radiation. Wet connective tissue tends to shrink and lose some of its ordered structure after irradiation. Irradiation makes dry collagen more water soluble.

Nucleoproteins are also modified by irradiation. Doses as small as 1 krep significantly reduce the viscosity of nucleic acid gels. Although the nature of the irradiation damage is not completely understood, it may result from one or more reactions such as slow rearrangement of the gel with elimination of polar groups, slow hydrolysis of phosphate esters, interaction of peroxide with deoxyribonucleic acid, or possibly decomposition of peroxide intermediate products. Both deamination and dephosphorylation are known to occur.[10]

Although the changes described above occur to an appreciable percentage in dilute aqueous solution, they usually amount to 1 per cent or less when Mrep doses are used on concentrated amino acid solutions or natural proteins. Figure 8.7 shows the influence of concentration on the decomposition of dl-phenylalanine by electron irradiation.[5]

PROTEIN PROTECTION BY COMPLEXING

Kuzin[9] has reported that the state in which high polymeric molecules are present in irradiated tissues influences their stability. He and coworkers found that pure myosin was highly sensitive to radiation but that complexes of myosin with glycogen had considerable stability. This protective effect is quite significant as demonstrated by the observation that a tenfold increase in dosage failed to produce changes in the myosin-glycogen complex that had been observed with pure myosin.

Danehy and Pigman[20] have anticipated Kuzin's observations with their work on protein-carbohydrate complexes. They show that much greater pH differences are required to dissociate the brown melanoidin's produced by the complexing of proteins, amino acids, and carbohydrates than for the uncomplexed substances. Furthermore, it was demonstrated that the reducing properties of the materials were not diminished by the complexing.

Gordy, Ard, and Shields,[21] using paramagnetic-resonance techniques, have shown that two characteristic patterns can be excited in irradiated proteins, one of which is clearly identified as owing to cysteine. Pollard,[22] interpreting Gordy's results, considers that the "irradiation which could hardly have been so intense as to excite the whole set of amino acids, should consistently give such a pattern, strongly suggests that a positive charge indeed does occur, resulting in its statistically settling in one or two favored spots." He proposes to call this effect the primary lesion. Subsequently the protein is exposed to water or to water and oxygen and exhibits a reactivity which he refers to as the chemical action. "This chemical action by breaking an $-S-S-$ (disulfide) bond or removing a side chain gives an altered molecule." Ingram,[23] in discussing Gordy's spectra from the standpoint of free-radical relations, believes that the spectrum produced is due to an unpaired electron localized on the $-S-S-$ bond of the

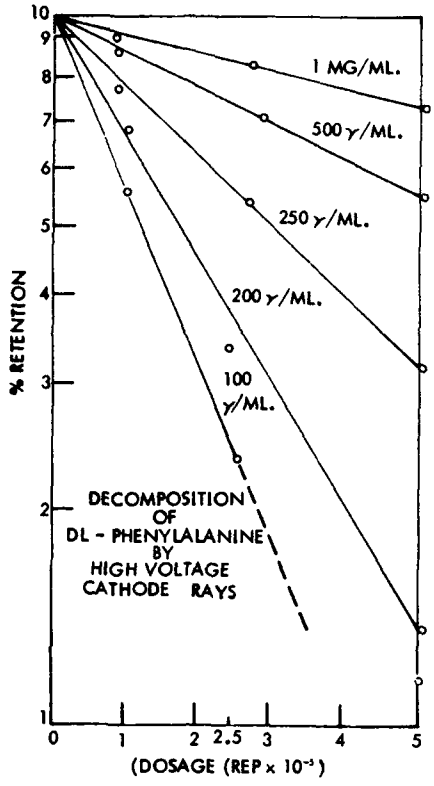
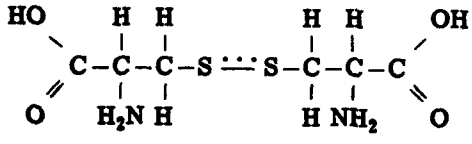


Fig. 8.7 — Effect of concentration on the decomposition of d 1-phenylalanine by high voltage cathode rays.[5]

cystine. If an electron is freed by irradiation from some point in the molecule, the vacancy probably moves to the S atoms to form an additional lower-energy three-electron bond, as represented by the structural formula



There is thus now a hole shared by the two sulfur atoms through exchange of their electrons, and the loss of an electron from the sulfur lone pair orbitals thus tends to strengthen rather than weaken the bond in this case. "If the three electron S–S bond represents a lower energy state, the cystine is likely to donate an unpaired electron to other ionized groups formed by irradiation." In this way the two sulfur atoms of the cystine molecule act as an electron reservoir and supply electrons to fill vacancies at other points in the protein molecule. And such a mechanism should reduce the damage produced by irradiation. This interpretation may serve to explain the radioprotective action of carotene-protein complexing. It is known that the –S–H– (sulfhydryl) bond is a much weaker bond than the –S–S– group and that biological damage *in vivo* is associated with the –S–H– group.[24] It is also known that beta-carotene forms complexes with proteins which are much more stable to dissociation than the uncomplexed proteins.[25] Beta-carotene is also known to be involved in the sulfur metabolism[26] and is radioprotective. It may be that the beta-carotene associated with the sulfhydryl group operates as an energy sink much as the –S–S– bond is supposed to do. Platt,[27] discussing the role of donor-acceptor complexes in photosynthesis, shows that the excited states of carotene, with respect to the donor or acceptor properties of free electrons resultant from excitation, differ sharply from differences in solvent and polarization effects. Carotene in chlorophyll grana is found invariably associated with and probably complexed to a protein. Platt considers that in this state the carotene-protein complex is important in electron transfer, and notes that

"such energy shifts and electron shifts might also be important elsewhere in biology." Bellamy et al.[28] found that the extremely radioresistant bacterium *Micrococcus radiodurans* is colored brilliantly red. This pigment is a carotenoid. Bellamy believes that it may serve as an "energy sink" and be important in radioprotection.

Dosages required for inactivation vary with the amino acid, as shown in Table 8.4.[29]

Irradiation of glycine results in deamination and production of ammonia and hydrogen. It

Table 8.4—RELATIVE RADIOSENSITIVITIES OF AMINO ACIDS IN AQUEOUS SOLUTIONS[29]

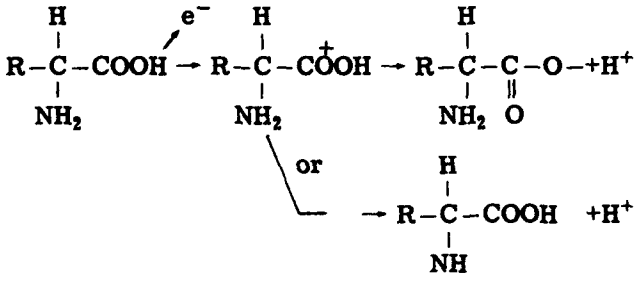
Amino acid	Specific inactivation dose, 10^9 rep/g/ml
dl-phenylalanine	1.18
l-histidine	1.25
l-cystine	2.40
l-tyrosine	2.60
l-leucine	3.60
dl-tryptophan	10.00

is believed that deamination is caused by simultaneous oxidation and reduction, possibly resulting from interaction with hydroxyl radicals and hydrogen atoms. Although the yield of ammonia by deamination of glycine appears unaffected by the oxygen concentration, the presence of oxygen has a significant effect in the deamination of serine. In the case of serine, a reduction in oxygen concentration increases the ammonia yield, indicating that this amino acid is more easily reduced than oxidized. For most amino acids, however, deamination generally increases as oxygen increases. The products may be aldehydes or keto acids. Leucine has been shown to yield isovaleraldehyde. This compound has a strong "goaty" odor and may be one of the sources of off-flavor in irradiated protein. Irradiation of solutions of tryptophan is believed to form some indole. Irradiated methionine forms sulfur products. Both have unpleasant odors. Side chains in the amino acid may be broken—for example, the imidazole ring of histidine and the aromatic rings of phenylalanine, tyrosine, and tryptophan. In general, amino groups in positions other than the alpha are not readily attacked. Thus, beta-alanine and beta-lysine are quite stable. However, the dipeptide glycyl-glycine is readily deaminated.[10]

AMINO ACIDS

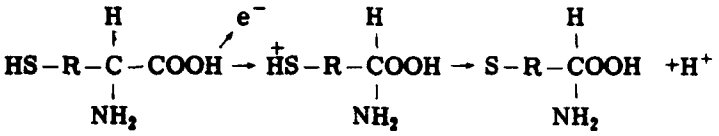
The amino acids are the primary building blocks of protein macromolecules and many of the effects of radiation on proteins can best be explained by considering the effects on individual amino acids. This is particularly true in regard to changes produced indirectly by radicals and excited molecules from irradiated water. Amino acids in proteins also may become ionized to form radicals and excited molecules, resulting in an infinitely great number of possible reactions. A few examples[9] are

By electron loss



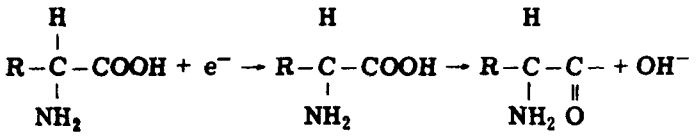
(8.1)

(8.2)



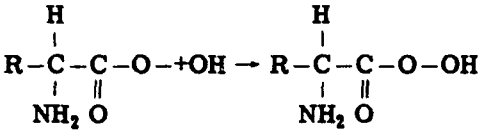
(8.3)

By electron capture



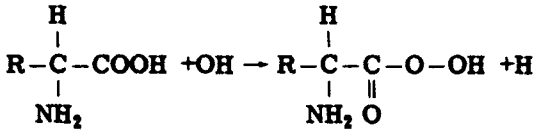
(8.4)

Radicals formed from amino acids by reactions such as those indicated by Eqs. 8.1 to 8.6 are surrounded by water. Therefore their most probable fate is additional reactions with H and OH radicals which may lead to restoration or the formation of peroxides[9]



(8.5)

Formation of peroxides can also occur by interaction of amino acids with hydroxyl radicals



(8.6)

For additional information on the effects of ionizing radiation on amino acids, see Refs. 30 to 42.

CARBOHYDRATES

Chemical and physical changes by irradiation of carbohydrates are not as conspicuous as the effects on proteins and lipids, because unpleasant flavors and odors are not produced. Some irradiated sugars do give off noticeable but inoffensive odors. Glucose converts to glucuronic acid, a preferential oxidation occurring at the C-6 position to form a COOH group. Also, sugar alcohols may be transformed to aldehydes, a reaction with possible commerical application.[10]

The effect of irradiation on mono- and disacharides is two-fold, namely hydrolysis and oxidation. The primary reaction of monosacharides in aqueous solution appears to be oxidation of the alcohol group.[43,44] D-glucose combines with amino acids and certain proteins to form polymerization products that are brown in color.

The disacharide sucrose is hydrolyzed to glucose and fructose by irradiation, at doses of 13×10^6 rep and greater. Hydrolysis of 50 per cent aqueous solutions of sucrose,[44] irradiated to 104×10^6 rep while cooled with (1) an ethanol-dry ice bath; (2) an ice water bath, and (3) ambient air, was 50.2, 39.3 and 33.6 per cent, respectively. (see Fig. 8.8).

Changes in many cellulose products, including cotton, paper, wood, and their products,[10] are slight at sterilizing radiation doses (about 3 Mrep).

However, cellulose products show appreciable degradation of the cellulose under high doses of radiation. Paper loses strength and after doses of about 100 Mrep it becomes brittle and crumbles easily. Plywood exposed to high doses of radiation shows similar degradation. The wood loses its fiber structure and may fail under load by brittle fracture, without splintering. This type of failure, however, is more characteristic of bark or cork than of fibrous wood. Carbon dioxide, water, mixed acids, and reducing compounds have been observed as decomposition products. Doses of 10 to 100 Mrep may depolymerize cellulose to products of lower molecular weights, such as sugar.

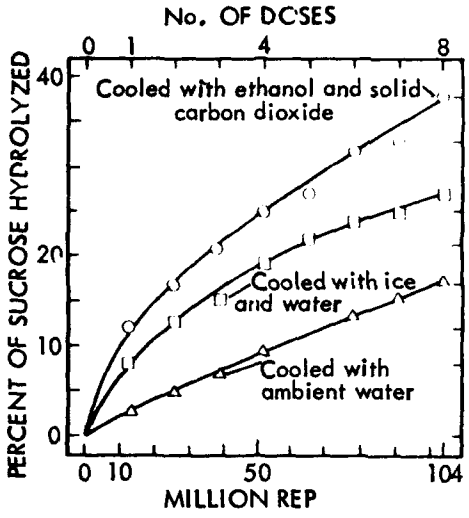


Fig. 8.8—Hydrolysis of sucrose by irradiation of 50 per cent aqueous solution.[44]

Phillips and Moody[45] have reported the products formed by the chemical action of gamma radiation on aqueous solution of carbohydrates. The products were identified and estimated by paper chromatographic techniques and radioactive tracer methods.

In dilute aqueous solutions D-glucose is degraded to D-glucuronic acid, D-gluconic acid, glyoxol, D-arabinose, D-erythrose, formaldehyde, saccharic acid, and 1 : 3 dehydroxyacetone. The main steps in the degradation of D-glucose in aqueous solution,[45] in the presence of oxygen may be represented as

D-Glucose

D-Glucuronic | D-Gluconic acid | Glyoxal + D-Erythrose

D-Arabinase | Glyoxal + Glycollic aldehyde

Dextran may be depolymerized by irradiation. A dose of about 0.3 Mrep appreciably reduces viscosity, with a corresponding reduction in molecular weight. Dextran may be irradiated either dry or in dilute solution. Studies of the products indicate that the carbohydrate chains degrade with no cross linking.[10]

Irradiation effects on the carbohydrates of potato cells were reported by Roberts and Proctor.[46] They found an alteration in the pectic constituents of the middle lamellae that softened the tissue. McArdle and Nehemias[47] investigated effects of gamma radiation on pectic constituents of fruit and vegetables; they found a relationship between radiation-induced softening of apple and carrot tissues and the changes in the pectic substances. Gamma irradiation caused a decrease in protopectin and total pectic substances, whereas soluble pectin and pectates increased. These effects were accompanied by depolymerization of the pectin, pectate, and protopectin molecules, as indicated by the decrease in viscosity shown in Table 8.5.

Wertheim et al.[48] described studies on radiation-induced browning in milk involving carbohydrates. Browning may be produced by overheating and has been a problem in the manufacture of dairy products. Also, browning develops rapidly in irradiated milk heated to 100°C. It was found that radiation-induced changes in lactose were largely responsible for the induced sensitivity to browning and that a lactose-protein complex was produced. The studies indicated that polyhydroxycarbonyls were probably formed by sugar dehydration in the presence of radiation-produced oxidative-free radicals and that these polyhydroxycarbonyls were the browning precursors. This theory was supported by the fact that radiation-induced browning did not occur when carbonyls were eliminated from the irradiated system.

Additional information on the effects of ionizing radiations on carbohydrates can be found in Refs. 49 to 52.

Table 8.5—RELATIVE VISCOSITY CHANGES IN PECTIC FRACTIONS INDUCED BY GAMMA RADIATION OF APPLES AND CARROTS[47]

Dosage, Mrep	Pectin		Pectate		Protopectin	
	Apple	Carrot	Apple	Carrot	Apple	Carrot
0.0	2.17	1.84	2.02	1.80	1.79	1.69
0.5	1.73	1.62	1.67	1.61	1.38	1.52
1.0	1.48	1.56	1.46	1.49	1.23	1.31
1.5	1.32	1.37	1.25	1.33	1.17	1.27
2.0		1.19		1.21		1.12

Table 8.6—EFFECT OF TEMPERATURE ON PEROXIDE VALUE OF BUTTERFAT[10,55] IRRADIATED WITH ELECTRONS, 2 Mrep

Temp. of sample, °C		Peroxide value, micromoles/g*							
			Days after irradiation				Days after irradiation		
During irradiation	During storage	Sample	0	2	8	Sample	0	2	8
−70	−70	A	3.4	4.2	5.0	B	2.7	4.1	3.9
	−20			16.6	17.3			17.3	14.1
	−20			5.0	6.6			5.1	7.9
	20			4.1	4.7			4.4	5.5
0	−70	C	4.0	2.8	4.9	D	2.1	3.2	4.1
	−20			17.3	16.9			19.6	17.7
	0			7.3	6.3			6.9	8.4
	20			2.6	4.1			3.5	7.9
20	−70	E	1.5	1.5	1.6	F	1.3	1.6	1.5
	−20			1.6	1.6			1.3	1.6
	0			1.1	1.0			1.1	1.0
	20			1.6	2.3			1.5	2.3
37	−70	G	0.2	0.6	0.6				
	−20			0.1	0.7				
	0			0.1	0.6				
	20			0.8	1.2				

* Total change in peroxide value plus arbitrary correction of 2 divided by number of ionizations.

Table 8.7—EFFECT OF DOSAGE ON PEROXIDE VALUE[10,55] OF ELECTRON-IRRADIATED BUTTERFAT STORED AT −20°C

Dose, Mrep	Peroxide value, micromoles/g,* after storing at −20°C				Effective over-all ionic yield†
	0 days	2 days	4 days	6 days	
0.10	1.4	2.2	4.1	4.7	8.0
0.25	1.65	8.0	9.0	8.1	5.4
0.50	1.95	14.6	20.5	23.0	7.6
2.00	2.0	23.5	27.0	26.0	2.2

* Total change in peroxide value plus arbitrary correction of 2 divided by number of ionizations.

† Measured by oxidation of ferrous ions in benzene-methanol solution.

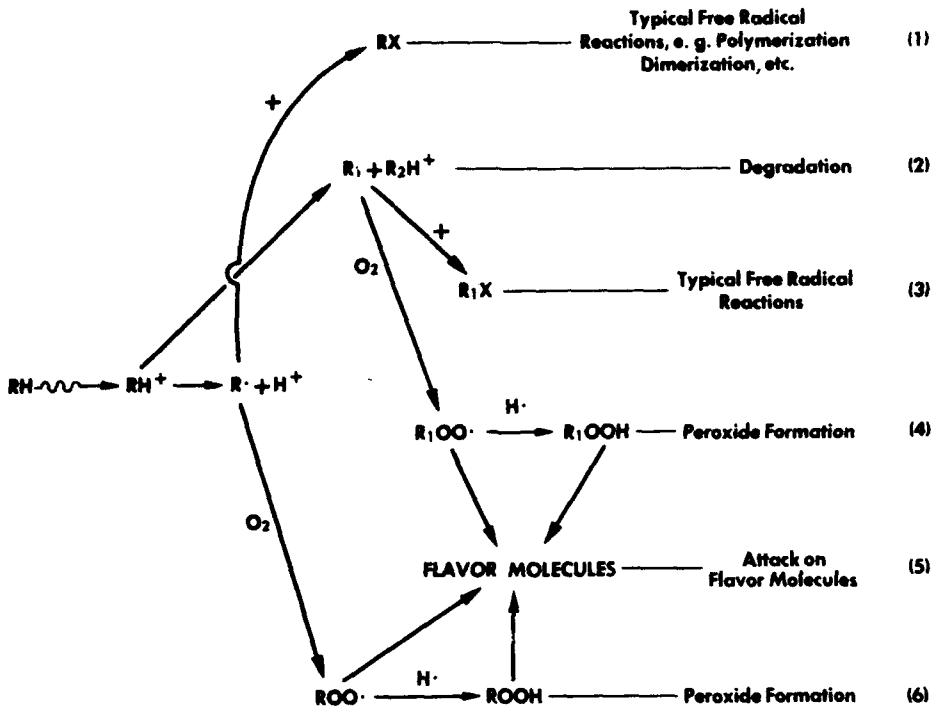


Fig. 8.9—Sequence of possible reactions in irradiated fats.[10]

LIPIDS

Ionizing radiation produces marked changes in lipids, making them of greater interest than the irradiated carbohydrates from the standpoint of nutrition. Irradiating an unsaturated drying oil such as linseed oil results in decreased iodine number and increased molecular weight. Some bleaching occurs and drying time is reduced. The reduced drying time is considered to result from destruction of antioxidants in the oil or autoxidation initiated by free radicals formed by the radiation.

Odor and flavor changes in vegetable and animal oils and fats have been observed after irradiation. On standing after irradiation, unsaturated fats tend to become rancid, but some oils have a pleasant (sweet-ketonic) odor, while others give off a burnt-metallic odor. The presence or absence of oxygen in the oil has an important bearing. When oils are irradiated in the complete absence of oxygen, irradiation odors and flavors are suppressed. However, adding common antioxidants such as propylgallate and butylated hydroxyanisole does not prevent characteristic irradiation odors from developing.

Hannan[10,53–55] describes the following changes that occur after fats are irradiated. Immediately after irradiation, tallowy odors and flavors develop, but this can be reduced about two-thirds by irradiation at dry-ice temperature for in the molten state (approximately 37°C for butter) instead of at room temperature. After irradiation, the peroxide value of fats is increased to a maximum value with a dose of about 0.5 Mrep. The peroxide value is lowest for fats irradiated in the heated state; it is greatest for fats irradiated at subzero temperatures. If the oxygen is completely removed from the fat before irradiation, peroxide value does not increase, nor do irradiation odors. Low-temperature storage of fats irradiated at low temperatures results in a progressive increase in peroxide value until a maximum is reached after 3 hr; the reaction during storage at low temperatures is prevented. Table 8.6 shows the effect of temperature during irradiation and during subsequent storage on the peroxide value of butter fat given a 2 Mrep dose of 1.2-Mev electrons. Table 8.7 shows the effect of dose on peroxide value of irradiated fat[10] before and during storage at –20°C.

Hannan[10] also gives a possible mechanism for the effect on fats, as shown in Fig. 8.9. Six different reactions are thought to occur after the formation of R, which represents a free radical of fat. The reactions (1), (2), (3), are decreased by the presence of oxygen and reactions

(4) to (6) are increased by the presence of oxygen. In reaction (2), R_1 represents a free radical fragment. In reactions (1) and (3), X represents any other molecule or free radical.

Doty et al.[56] have shown that the production of carbonyl compounds during beef irradiation is influenced by the fat content of the meat. These carbonyl compounds are formed even if oxygen is absent.

Recent work[57] also shows that since peroxide production is a chain reaction, the energy yield would be dependent on the dose rate and, experiments indicate, the higher the dose rate, the lower the peroxide production per dose.

Coleby[58] has discussed chemical changes produced in lipids by irradiation and Lea[59] has considered lipid oxidation under conditions when microbial spoilage is not limiting.

Additional information on the effects of ionizing radiation on fats is given in Refs. 60 to 63.

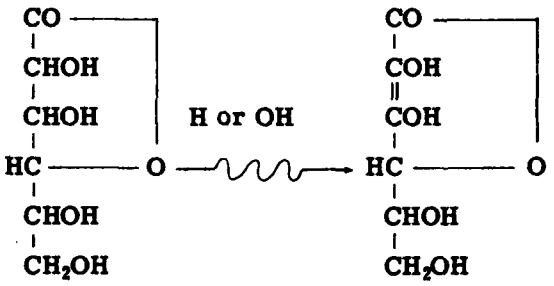
VITAMINS

Of all the nutrilities in food, vitamins are most susceptible to destruction by ionizing radiations. Low dosages (up to 100 krep) produce little damage in most vitamins but dosages high enough for radiosterilization significantly reduce the vitamin content of food. The percentage of destruction varies with the vitamin, the food or medium, and the dosage. For this discussion, the vitamins may be divided into two groups: water soluble and oil soluble.

Water-Soluble Vitamins

The principal water-soluble vitamins are vitamin C (ascorbic acid) and the B-complex vitamins (thiamine, riboflavin, niacin, p-amino benzoic acid and B_{12}). Ascorbic acid irradiation has been studied most. When irradiated in solution, it oxidizes to dehydroascorbic acid, which can be reduced back to ascorbic acid biologically. Irradiation oxydizes about half of the ascorbic acid in fresh fruit juice. The other half is destroyed. Diluting the ascorbic-acid solution increases the percentage destroyed by a given radiation dose. However, the total quantity destroyed by a given dose remains approximately the same. Ascorbic acid in pure solution is more susceptible to irradiation damage than that in natural fruit juice because of the protective effect of other constituents in the juice. Freezing the solution to subzero centigrade temperatures minimizes the destruction of ascorbic acid[5] by irradiation as shown in Fig. 8.10. Because ascorbic acid readily reacts with free radicals produced by irradiation, it is considered a free-radical acceptor and has been added to foods to lessen flavor changes by competing with flavor molecules for the free radicals.[10]

Although ionizing radiations are quite destructive to ascorbic acid, Coleby[64] has reported on the formation of ascorbic acids by ionizing radiations; D-glucono-γ-lactone and L-gulono-γ-lactone are converted into the corresponding ascorbic acids by the action of hydroxyl radicals on solutions of these compounds. One example of such conversion is indicated by Eq. 8.7.



(8.7)

Ascorbic-acid yield was found to depend on the concentration. As the concentration went from 5×10^{-5} M to 0.004, the ascorbic-acid yield or G (molecules produced per 100 ev of energy absorbed) increased from 0.16 to 0.94.

Coleby[64] comments that, as L-gulono-γ-lactone is the biological precursor of vitamin C, ionizing radiations, under some conditions, might increase the vitamin C content.

The other water-soluble vitamins appear to be more resistant to radiation than ascorbic acid. Thiamine can be destroyed by irradiation, but riboflavin and niacin are progressively

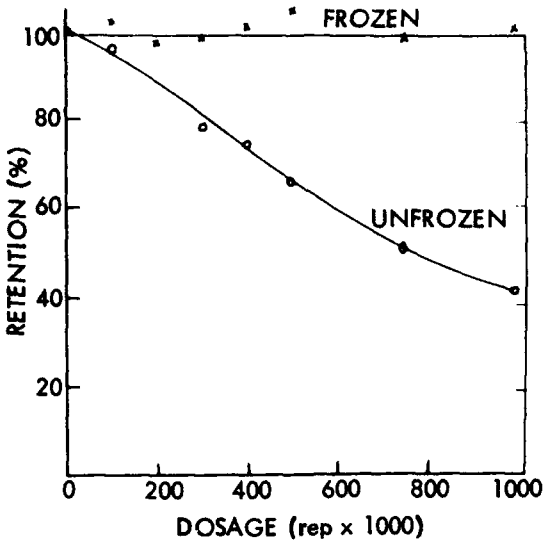


Fig. 8.10—Effects of 3-Mev cathode rays on l-ascorbic acid in frozen and unfrozen fresh orange juice.[5]

more resistant. Other food substances may protect vitamins somewhat from irradiation effects; in turn, vitamins such as ascorbic acid may protect flavor constituents in the foods.[77] Probably the most sensitive vitamin in the B complex is B_{12}. A dose of only 10 to 20 krep will destroy more than 50 per cent of this vitamin in pure aqueous solution. However, when the vitamin is in a food such as milk, the dose may be increased to 0.5 Mrep before there is equivalent destruction. This is another example of food constituents protecting one another.

Table 8.8—EFFECT OF 0.125 Mrep DOSE OF HIGH-VOLTAGE X RAYS ON NIACIN, ASCORBIC ACID, AND MIXTURE OF BOTH[66,67]

Solution	Concentration before irradiation, γ/ml		Retention after irradiation, γ/ml	
	Niacin	Ascorbic acid	Niacin	Ascorbic acid
Niacin	50		36.0	
Ascorbic acid		500		66.6
Niacin ascorbic acid mixture	50	500	44.0	83.6

The protective effect of niacin on ascorbic acid is shown in Table 8.8; that of milk on ascorbic acid and riboflavin is shown in Table 8.9.[66]

Studies by Kung et al. on the vitamin content of irradiated milk indicated considerable loss.[77] For example, a dose of about 0.5 Mrep given to vitamin-enriched milk was shown to destroy 70 per cent of vitamin A, 40 per cent of carotene, 61 per cent of vitamin E (tocopherols), 100 per cent of the reduced ascorbic acid, and 37 per cent of riboflavin.[10] The high percentage of water in milk is believed to result in an appreciable destruction from indirect process, due to formation of oxidizing and reducing agents by the irradiation of water. In general, results with other foods show appreciably less destruction.

Alexander et al.[68] reported the effects of a 3-Mrep dose of gamma radiation on the water-soluble vitamins in raw beef. They found that about 56 per cent of thiamine, 9 per cent of riboflavin and 24 per cent of pyridoxine were destroyed; no loss in niacin, choline, or folacin was

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Table 8.9—SPECIFIC INACTIVATION DOSE FOR ASCORBIC ACID AND FOR RIBOFLAVIN IN PURE SOLUTION AND IN EVAPORATED MILK[66]

Irradiated medium	Specific inactivation dose, 10^6 rep/g ml
Ascorbic acid, pure solution[65]	1.19–3.6
Ascorbic acid, evaporated milk[77]	24.0
Riboflavin, pure solution[65]	2.6–5.2
Riboflavin, evaporated milk[77]	370.0

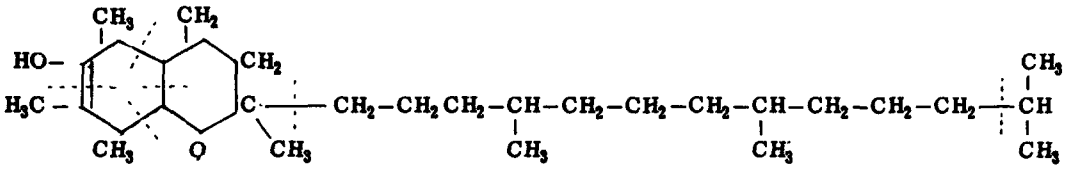
detected. Groninger et al.[69] also reported that thiamine was more susceptible to destruction by radiation than were the other B-complex vitamins (riboflavin and niacin) studied. It was reported that a 0.2-Mrep dose destroyed 21 and 18 per cent of thiamine in irradiated tuna and ham; that a 2.0-Mrep dose destroyed 75, 58, 84, 89 and 82 per cent of thiamine in irradiated pork, chicken, salmon, halibut, tuna, and ham, respectively. However, doses as high as 1 Mrep destroyed little or none of the niacin in pork and beef and a dose of 3 Mrep destroyed only about 30 per cent.[70]

Oil-Soluble Vitamins

The effect of irradiation on the oil-soluble vitamins (A, D, E, and K) has been studied less than effects on water-soluble vitamins. Vitamin A and carotene dissolved in hydrocarbon solvents are destroyed by irradiation, apparently by an indirect effect. Hence, indirect effects are not limited to aqueous solutions.[71]

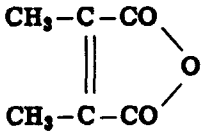
Vitamin E or α-tocopherol has the empirical formula $C_{29}H_{50}O_2$. The compound readily forms ethers and esters, indicating that one of the oxygens is in the form of a free hydroxyl group as shown below:

α-Tocopherol



The dotted lines in the formula above indicate points of probable scission as a result of oxidation with chemicals such as chromic acid.[72] The following products have been isolated as a result of such oxidations:

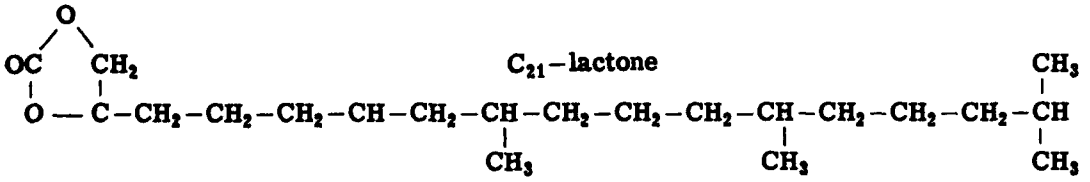
dimethyl-malic anhydride



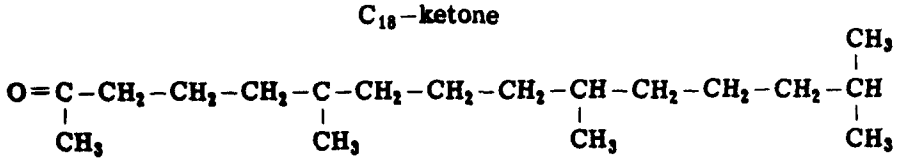
diacetyl $CH_3-CO-CO-CH_3$

acetone $CH_3-CO-CH_3$

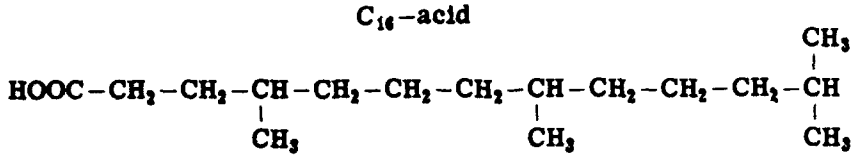
C_{21}–lactone



C_{18}-ketone



C_{16}-acid



Of the oil-soluble vitamins, E, (tocopherol) is probably the most susceptible to destruction since it is so easily destroyed by oxidation. Tocopherol is an antioxidant in natural fat systems somewhat equivalent to ascorbic acid in natural aqueous systems. If oxygen is present in either case, the antioxidant properties are lost quite readily. Poling et al.[73] and Brownell et al.[74,75] observed vitamin E deficiencies when feeding irradiated diets (2- and 4-Mrad doses, respectively) to rats. A large loss of tocopherol was observed by Tappel et al. when beef was irradiated at 3 to 30 Mrad in oxygen, but similar irradiations under nitrogen produced no apparent destruction.[76] This shows the great susceptibility of tocopherol to oxidation.

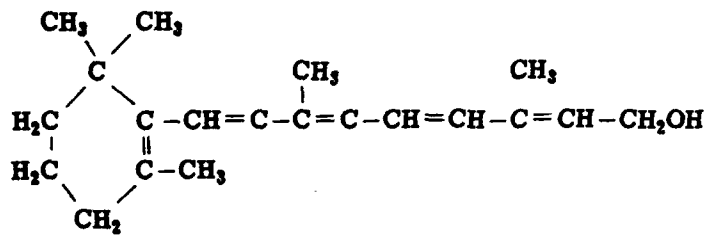
Kung et al.[77] reported high destruction of vitamin E as determined by bioassay of irradiated butterfat, whereas chemical assays for this vitamin showed little destruction. Groninger et al.[69] also reported little destruction (as determined by chemical assay) of vitamin E in meats given 3 Mrad radiation doses. Burns and Brownell[78] showed that a low dosage (10,000 rad) of gamma radiation did not destroy the effectiveness of irradiated whole wheat in supporting normal reproduction in four successive generations of rats. In this study, the irradiated whole wheat was the sole source of vitamin E, which is essential to normal reproduction.

Tocopherol acts as a protective agent to vitamin A and to precursors of vitamin A such as carotene. It is often added to vitamin preparations for this purpose.[72] In the additive mixture tocopherol is believed to protect the vitamin A in the mixture, to act as an oxygen scavenger and thereby aid in protecting against flavor change.

Voth et al.[79] have studied the interactions of α-tocopherol with proteins and amino acids. They find that, in the acid state of a pH of 7 or below, α-tocopherol binds to negative centers on the protein such as the carboxyl group. At distinctly alkaline states of pH above 10, there is binding to the amino group. Thus an increase in the net negative charge results in a greater binding of α-tocopherol, while blocking the carboxyl groups results in a loss of affinity. In their studies of the reactions with amino acids, they show that positive amino acids such as lysine, arginine, and histadine do not appear to bind α-tocopherol very well. Cystine also has a low affinity. This evidence indicates that, if α-tocopherol is active in masking sensitive areas on a protein against radiation damage, it is fairly specific and does not interfere with the probable activity of the carotenoids which have an indicated activity at the sulfhydryl linkage. The affinity for the carboxyl groups also gives some indication of the nature of the fat antioxidant properties of the tocopherols. If the carboxyl groups of the fatty acids of the glycerides are filled, to some extent with α-tocopherol, then the molecule is masked from further reactions with radiation-produced free radicals.

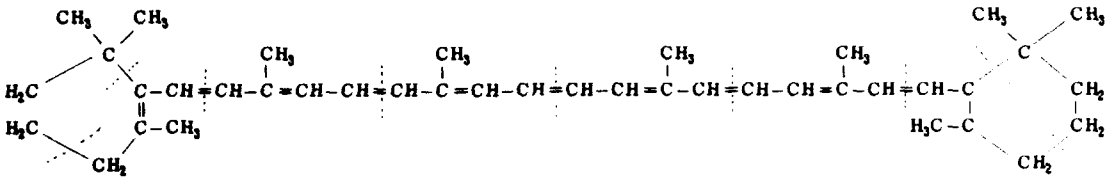
Vitamin A has the empirical chemical formula of $C_{20}H_{20}O$ and the structural formula shown below.

Vitamin A



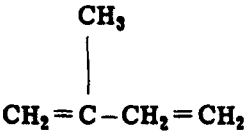
Vitamin A is an alcohol with the hydroxyl group at one end of the molecule, the β-ionone ring at the other end and a highly unsaturated hydrocarbon chain of isoprene units in between. This vitamin molecule represents approximately one-half of the molecule of β-carotene shown below:

β-carotene

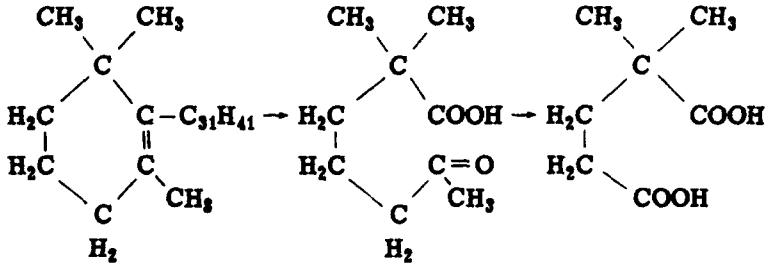


The formula above for β-carotene is marked with dotted lines to show how it contains skeletons of 8 molecules of isoprene, C_5H_8.

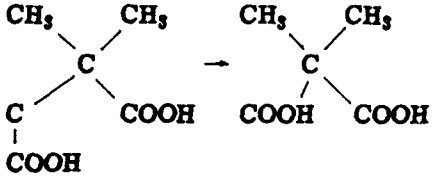
isoprene



Destructive oxidation of carotene gives successively: geronic acid, α, α-dimethyl glutaric acid, α, α-dimethyl sericinic acid, and dimethyl-malonic acid as follows:[80]



β-carotene — Geronic acid — α,α-dimethyl-glutaric acid



α,α-Dimethyl succinic acid — Dimethyl malonic acid

The most common natural source of β-carotene is the carrot root. Tomatoes which have been found to have a protective effect on flavor changes in irradiated cooked meat contain both carotene and a similar carotenoid, lycopene.[81] Both β-carotene and lycopene have the same empirical formula, $C_{40}H_{56}$, but in the case of lycopene both β-ionone rings are opened between 1 and 6 and 1′ and 6′ carbon atoms of the rings.

A dose of only 150,000 rad destroyed 80 per cent of β-carotene dissolved in petroleum ether.[82] Groninger et al. reported that in gamma-irradiated meats a Mrad destroyed 43 per cent of the carotenoids in beef, 58 per cent in chicken and 81 per cent in salmon.[69] At a 4-Mrad dose, the destruction of these carotenoids was increased to 76, 95, and 97 per cent, respectively. However, a 4-Mrad dose given to prawns destroyed only 50 per cent of the carotenoid astaxanthin and a 20-Mrad dose given to tomatoes caused less than 15 per cent loss.[76]

The resistance of carotenoids to destruction by radiation is apparently a function of the medium in which they are irradiated and the state in which they exist. Henry[82] comments that the carotenoids are found naturally in one of three categories: (1) dissolved in lipids, (2) attached to proteins such as the pigment coloring in crustaceans, and (3) in semicrystalline or amorphorus aggregates such as occurs in carrots and tomatoes. As has been stated, carotenoids dissolved in hydrocarbon solvents are very susceptible to destruction. When dissolved in lipids, they are less sensitive to destruction (except for salmon carotenoids in which a dose of 200,000 rad destroys 68 per cent).

Shrimp and prawns show examples of a carotenoid attached to proteins. This complex appears to be much more stable to the action of radiation. Greatest stability of carotenoids occurs when they are present in the semicrystalline or amorphorus aggregates as in carrots and tomatoes.

The complexing of carotenoids with proteins occurs with systems other than crustaceans. Moore states that it is highly probable that, in the utilization of carotene and vitamin A in the body, these materials are held in blood plasma by combination with protein.[80] In studies with blood plasma, carotene and vitamin A were precipitated with albumin and it was concluded that these hydrocarbons were carried in the blood attached to albumin. In other studies, carotene and vitamin A esters were found to be associated with the least soluble protein fractions, whereas the vitamin A alcohols were bound to the more soluble protein fractions.

Deborin et al.[25,83] state that carotene-containing lipoproteins are widely distributed in the organism. They point out that a number of observations support the view that protein-bound carotene is relatively resistant to oxidation but that denaturation of the protein is followed by the oxidation of the carotene. Deborin found that, while carotene is protected from oxidation when combined with a protein, oxidized carotene was not able to form complexes with the protein. Martinson and Fetinsenko have shown that carotene has an inhibitory effect on the proteolytic action of both cathepsin and pepsin. Balakhovski[26] found that carotenoids exert an activating effect on oxidation involving molecular or peroxide oxygen, and that this process appears to be regulated by reversible reactions between proteins and carotenoids.

The formation of carotenoid-protein complexes to produce a more radiation-resistant system is important in radiation protection against destruction of any component in the complex.

Since irradiation may also synthesize the D vitamins from sterol precursors, it is possible that under some conditions a limited dose of radiation might increase rather than decrease the vitamin D content. No evidence to support this possibility has been reported, but the optimum radiation dose for increasing vitamin D content probably would be considerably less than 3 Mrep.

Richardson et al. reported on the effect of a 3-Mrep dose of gamma radiation on vitamin K compounds (menadione, K_1, K_5), dehydrated alfalfa leaf meal, and on fresh spinach (natural sources of vitamin K).[85] Of the vitamin K compounds, menadione was more readily destroyed. However, there was practically no destruction of vitamin K when diets containing dehydrated alfalfa leaf meal or fresh spinach were irradiated. They concluded that "no animal, including humans, receiving irradiated diets would ever develop a hemorrhagic condition because of a deficiency of vitamin K in the diet."[85]

NUTRILITES

Long-term feeding and breeding experiments are required to explore the effect of gamma radiation on the biological value of nutrilites when mammals are used as the test subject. However, much information can be obtained in a short time if protozoa are used. Certain of these one-celled animals such as *Tetrahymena pyriformis* have approximately the same requirements for the essential amino acids and water-soluble vitamins as mammals. Using this microorganism, Elliott, Gross and Brownell[74,75,86] reported the effect of gamma irradiation on the nutrilites of a completely synthetic medium used to grow the protozoa. The analysis of the medium used is given Table 8.10.

The organism is classified as a *holotrichous ciliate* and is a member of the *Colpidium-Glaucoma-Leucophrys-Tetrahymena*-group. Fresh water is its natural habitat and it is holo-

Table 8.10—COMPOSITION OF THE BASAL MEDIUM USED TO GROW *TETRAHYMENA PYRIFORMIS*[75]

Compounds	Molarity at irradiation	Final concentration in medium, mg/liter
Amino acids		
L-Arginine · HCl	0.172	150
L-Histidine · HCl	0.142	110
D_XL-Isoleucine	0.152	100
L-Leucine	0.106	70
L-Lysine	0.048	35
D_XL-Methionine	0.046	35
D_XL-phenylalanine	0.061	100
D_XL-Serine	0.316	180
D_XL-Threonine	0.343	180
L-Tryptophan	0.019	20
D_XL-Valine	0.102	60
Carbon sources		
Glucose	1.4	1000
Sodium acetate	3.0	1000
Nucleic acid components		
Adenylic acid	0.0088	25
Cytidylic acid	0.0096	25
Guantidylic acid	0.0087	25
Uracil	0.0223	25
Vitamins		
Thiamine-HCl	8.25×10^{-4}	1
Riboflavin	7.5×10^{-5}	0.1
Ca pantothenate	0.96×10^{-4}	0.1
Niacin	2.03×10^{-4}	0.1
Pyridoxine · HCl	2.33×10^{-3}	2
Folic acid (PGA)	5.75×10^{-6}	0.01
Thioctic acid	4.3×10^{-8}	0.001
Inorganic salts		
K_2HPO_4	0.144	100
$MgSO_4 \cdot H_2O$	8.1×10^{-3}	10
$Zn(NO_3)_2 \cdot 6H_2O$	3.9×10^{-3}	5
$FeSO_4 \cdot 7H_2O$	4.2×10^{-4}	0.5
$CuCl_2 \cdot 2H_2O$	7.3×10^{-4}	0.5

zoic in nature; i.e., it feeds on particulate food. The name *Tetrahymena* comes from the four membranes in the oral opening.

A preliminary irradiation of the medium described in Table 8.10 was performed to determine the range of radiation effects and to compare radiation effects on a dry and on a liquid medium. A range of irradiation from 0.5 to 4 Mrep was employed in the initial screening experiment. The protozoa grew equally well in a medium prepared from the irradiated dry components and in the nonirradiated medium. On the other hand, the irradiated liquid medium failed to support growth of the ciliates when the irradiation dose was 0.5 Mrep or greater. When nonirradiated medium was added to these inhibited cultures, growth was recovered at a rate inversely related to the radiation level; i.e., recovery was greater when fresh medium was added to that which received only 0.5 Mrep than when added to medium irradiated by 4 Mrep. A dose-response curve based on the experimental data showed that the critical range of irradiation for the complete liquid medium was from 0.15 to 0.30 Mrep.

In the first experimental series to test separate components for irradiation effects, each was irradiated in the concentration usually employed in preparing the medium, as given in Table 8.10. Media were prepared by omitting one of the components, or several of them in groups. The five groupings used were: amino acids, vitamins, nucleic-acid components, glucose, and sodium acetate. Each component was individually irradiated with 1.0 Mrep. Then the compounds were pooled in the described groups and added to the experimental medium that was deficient in the test group. When the growth of *Tetrahymena* was tested in such media it was found that only the medium containing irradiated vitamins failed to support growth. All other media (except a control prepared entirely from irradiated components) yielded normal growth.

To determine which vitamins were destroyed at 1 Mrep of irradiation, an experiment was performed in which single vitamins, prepared as shown in Table 8.10, were omitted from the medium. An irradiated vitamin of the type omitted was placed in the medium. Growth experiments in this series indicated that pyridoxine and thiamine did not lose efficiency from irradiation, but that thioctatic acid, folic acid, riboflavin, niacin, and pantothenate were partly destroyed. That all the irradiated vitamins lacked toxicity was demonstrated by supplementing the nonirradiated control medium with irradiated vitamins, one type at a time, and obtaining growth equivalent to that in the control medium. Additional tests were made with irradiated individual components to determine the dosage of gamma radiation required to affect the various vitamins. High-level dosages in other omission experiments were observed to produce effects on other nutrilites. The results can be summarized as follows.

Thiamine, riboflavin, pantothenate, pyridoxine, folic acid, and thioctic acid were altered structurally by less than 1 Mrep, whereas 2 Mrep were required to produce changes in niacin. Most amino acids proved relatively radiation resistant: at the high levels of 10 and 23 Mrep, only serine and methionine, respectively, were damaged. All other amino acids remained biologically active even after receiving 23 Mrep, the highest level of radiation employed. The nucleotide guanylic acid was inactivated at 23 Mrep.

ENZYMES

Enzymes are complex protein molecules that in some cases approximate the size and molecular weight of the smaller viruses. Thus, the resistance of enzymes to effects of ionizing radiation might be expected to parallel that of the smaller viruses and, in general, this has been shown to hold true.

In foods a much higher radiation dose is required for total inactivation of most enzymes than for the destruction of bacteria, with the consequence that enzyme activity may persist in food which has been given sterilizing doses.

The radiation sensitivity of enzymes is affected by its state, i.e., in solution or dry, concentration, temperature, pH, and the presence of other compounds. Enzymes in general are inactivated by indirect action.

The inactivation dose for enzymes decreases with increasing temperature. For enzymes in aqueous solution the effect is very marked at the transition from the frozen state[125] as shown in Fig. 8.11.

The dosage required for inactivation of enzymes in aqueous solution is highest at a pH of 6 to 7 and decreases as pH changes above or below these limits.

Dale[87,88] made early studies on the decrease in activity of some enzymes caused by x-rays. He used low dosages, in the range of tens of thousands of roentgens. Some enzymes (carboxypeptidases) were found to resist inactivation in the presence of their substrates (materials whose reactions the enzymes catalyse). He also attributed a significant protective effect to the presence of carbohydrates and fats.[87,89] Dale showed that dilute solutions of enzymes are more easily inactivated than concentrated solutions and proposed several theories to explain these effects.[90,91]

Forssberg,[92–94] another early investigator, reported that X rays and environment inactivated the catalase process. His results on the dilution effect and substrate protection were in accord with those of Dale. Slight changes in environment yield dosage differences for equivalent inactivation of as much as 1000 per cent. Several mathematical expressions of inactivation as a function of dosage and dilution have been presented.[92] For the most part, early in-

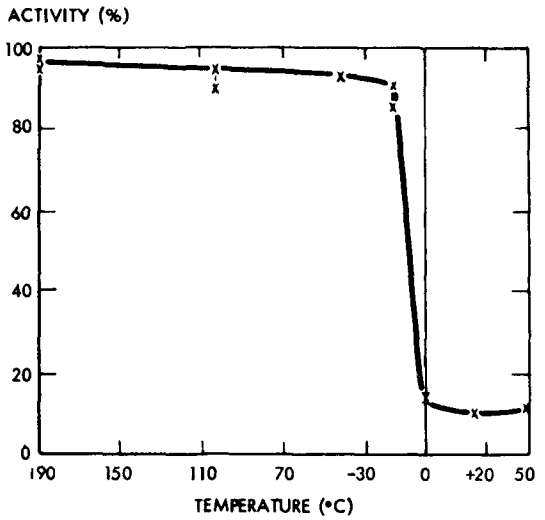


Fig. 8.11—Effect of temperature on the inactivation of 10 mg per milliliter solution of pepsin by 2.5-Mrep dose.[125]

vestigators of radiation effects on enzymes agreed that inactivation is an exponential function of dosage. Forssberg, although corroborating this expression for "high" concentrations of enzymes, offered a linear relationship for "low" concentrations.

Protection of an enzyme by its substrate is not a general rule. Laser[126] found the enzyme, glucose oxidase, to be very sensitive to radiation in the presence of glucose.

Pollard[95] reported the inactivation of dry enzymes (pepsin, trypsin and chymogen) by deuteron bombardment. An exponential relationship for survival was given, with dosages reported in terms of deuterons per square centimeter.

Barron,[96] working with the sulfydyl enzymes, stated that radiation did not destroy the enzyme but modified the active groups in the same enzyme molecule; as examples he gives the oxidation of the OH and SH groups. A number of investigators suggested that some changes accompanying irradiation of enzymes may be caused by changes in the colloidal properties of these compounds. In "solution," most enzymes are dispersed colloidally.

Table 8.11—PROTEINASE ACTIVITY* OF IRRADIATED BEEF EXTRACTS AND RESIDUES[15]

Dose, Mrep	Extract, mg/g		Residue, mg/g	
	Nonirradiated	Irradiated	Nonirradiated	Irradiated
1.6	0.085	0.019	0.087	0.080
1.6	0.046	0.012	0.059	0.037
0.5	0.035	0.035	0.062	0.056
0.5	0.034	0.030	0.063	0.058
0.5	0.037	0.021	0.076	0.075
0.5	0.028	0.033	0.076	0.076

* Incubation at 37°C for 24 hr at pH 4.6 with casein substrate. Results are expressed as tyrosine liberated.

The effect of gamma radiation on the proteinase activity in beef muscle tissue was reported by Doty and Wachter,[97] and additional studies of proteolytic enzymes were reported by Doty et al.[15] A dosage of 1.6 Mrep reduced the activity of the proteinase. It also reduced the amount of extractable tyrosine, as shown in Table 8.11, but left sufficient proteolytic activity to catalyse proteolytic changes in raw meat stored without refrigeration. Dosages of 0.5 Mrep had little effect on proteinase activity.

Lawrence[98] reported the effect of gamma radiation on the activity of phosphatase, an enzyme in raw milk used as an index of pasteurization because heat readily inactivates it. Phosphatase activity was measured by its action on disodium phenyl phosphate and expressed as milligrams of phenol per milliter, as given in Table 8.12. This table shows no significant reduction in phosphatase activity with radiation dosages up to 1.92 Mrep.

Table 8.12 — PHOSPHATASE ACTIVITY IN RAW MILK AFTER GAMMA IRRADIATION[98]

Radiation dose, Mrep	Phenol, mg/ml
0.0	0.55
0.14	0.55
0.36	0.48
0.90	0.51
1.92	0.43

In dilute aqueous solution, enzymes are much more susceptible to radiation damage, as shown by Lawrence in Table 8.13 for pepsin.[98] Cystine, and, to a lesser extent, sodium pyruvate, exert a protective effect upon dilute aqueous solutions of this enzyme.[98]

Table 8.13 — PROTECTIVE EFFECT OF ENZYME CONCENTRATION PEPSIN FOLLOWING GAMMA IRRADIATION*[98]

	Tyrosine for three enzyme concentrations, mg/ml		
Dose, Mrep	10	1	0.01
0.0	0.30	0.158	0.012
0.08	0.30	0.095	0
0.16	0.30	0.085	0
0.24	0.30	0.58	0

* pH of solution, 1.1; sample size, 4 ml; temperature, 25 to 26°C.

Setlow[99,100] has reported that the effect of ionizing radiation on dry catalase depends on temperature.

A thorough study of the inhibition of enzymatic activity in irradiated foods has been reported by Green et al.[101] The studies centered on enzymes in milk, beef liver, beef muscle, beef kidney, beef lung, beef heart, mushrooms, and cucumbers. Specific enzymes were: xanthine oxidase, alkaline phosphatase, succionoxidase, cytochrome oxidase, proteolytic enzymes, tyrosinase, and ascorbic acid oxidase. Results on several enzymes are given in Table 8.14.[101]

There appears to be a linear dose-inactivation of xanthine oxidase up to about 10-Mrep dose, at which approximately 85 per cent of the activity of the xanthine oxidase has been lost. Only 10 per cent more inhibition is recorded when the dose is increased to 20 Mrep. This characteristic break in the linearity of the dose-inactivation relation is observed in the data shown in Table 8.14.

RADIATION EFFECTS WITHIN A BIOLOGICAL CELL

The effects of radiation on biological components within the cells of living organisms and of foods with intact cells are believed to differ in some respects from the effects observed in solutions of biological materials.

Table 8.14—EFFECT OF IONIZING RADIATION ON VARIOUS ENZYMES[101]

Radiation dose, Mrep	0.1	0.2	0.5	0.6	1.0	2.0	4.0	5.0	6.0	8.0	10.0
Enzyme	Per cent inactivation										
Alkaline phosphatase of milk	3.7		32.3		45.3			92.3			100
Proteolytic activity of beef spleen		8.5		24.5	48.5	51.0	68.5				
Proteolytic activity of beef kidney		7.0		12.0	30.0	43.5	61.5			76.5	
Proteolytic activity of beef heart			7		12	29	43			58	
Mushroom tyrosinase	21		25		33	37	42		45		
Cucumber ascorbic acid oxidase			10		15	25	39		55	78	67

The discussion of the radiochemistry of water given in Chap. 7. considered the excitation of water molecules primarily from the standpoint of indirect action. It is understood that the energy of the initially excited species is transferred by a series of collisions to a sensitive site. Although it is generally agreed that the major damage to biological systems occurs through direct mechanisms, there is a growing body of opinion that considers indirect mechanisms to be more extensive than formerly realized.[140]

Szent-Gyorgyi[141] in 1957 proposed that biological energy transfer involves structured water: i.e., that ultra structures, proteins, and other macromolecules exist in a hydrated state, accompanied by a sheath of "frozen water." By frozen water Szent-Gyorgyi explains that there is a possibility that much of the water in biological systems exists in a quasi-crystalline condition, which is to say that water, being composed of dipoles that are attracted to surfaces and *other water molecules, will in a short time become so oriented as to present an array of loose* hexagonal structures whose electrical (dielectric) properties are different from random or disorganized water. He considers that water has two melting points, one at 0°C and the other somewhere between 30 and 40°C. A crystalline array is more efficient and direct in the transfer of energy, since the orbital electrons of the individual molecules fuse into continuous bands which present direct pathways for the migration of free electrons or other excitations.

Reid[142] states that "Energy migration along oriented stacks or chains of molecules could well account for the sensitivity of certain biological structures to radiation." In a recent paper Klotz[143] discusses the frozen water of proteins more extensively as it affects their general reactions. *One of the effects of simultaneous heating during irradiation may be the disruption of* structured or "frozen water" and so the disruption of direct pathways to sensitive chemical groups.

It is known that proteins evolve different off-odors in the lyophilized and in the hydrated state.[144,145] Recent work suggests that lyophilized proteins are partially denatured and exhibit caging effects with respect to their free radicals.[146] Hydration of such proteins may provide "bridges" to sensitive sites.

The radiation chemistry of proteins, direct excitation of protein molecules, the possible influence that some protective materials and structural water or frozen water of hydration may have on the action of radiation have been discussed. Klotz's excellent, *Protein Hydration and Behavior*, is a very thorough discussion of the concept, and contributes, we believe, an important perspective to the understanding of the interactions of radiation and proteins.[143] The importance of water in radiation damage to the components within tissue cells in meat has been described by Bellamy who demonstrated differences in the odors of lyophilized and hydrated proteins.[144,145] His work demonstrated that the characteristic off-odor of meats and proteins did not appear unless the protein was hydrated during or after irradiation.

A recent paper by Russian workers, Blyumenfeld and Kalmanson,[146] provides a possible explanation for this phenomenon as well as evidence to support the observation of Gordy and

others that there is transfer of excitation energy along a protein to a critical or quenching site, as has been suggested previously in this section. Most significant in their findings is that irradiated normal (lyophylized) proteins do not exhibit significant electron paramagnetic resonance signals which would indicate the presence of free radicals, but that thermally denatured and irradiated proteins such as oxyhaemoglobin pepsin, and casein have radiation-induced free radicals whose concentrations are several hundred times greater than nondenatured protein (excitation observed as doublets).

Blynmenfeld and Kalmanson comment that: "Thermal denaturation replaces the long hydrogen bond sub-chains by chaotic hydrogen bonds which do not form a single system. The conduction channels via which the ejected electrons combine with the holes are thereby destroyed and the free-radical concentrations hence raised. We consider that our data indicate that electron conduction dependent on hydrogen-bond network is present in native proteins."[146]

This information and observations of other investigators such as Bellamy indicates that water is involved in a process of energy transfer in proteins and should be considered in any attempted explanation of radiation damage within cells of biological systems.

EFFECT OF IONIZING RADIATION ON MICROORGANISMS

No organism ever tested has been found able to survive the more intense applications of gamma irradiation. Thus, gamma radiation is dangerous to man and dictates special handling. This also makes gamma radiation a valuable tool for processing biological materials, such as foods. Food is spoiled by the action of microorganisms such as yeasts, molds, and bacteria. In addition, it can be damaged or ruined by natural sprouting, as in potatoes, onions, and beans, or by insect infestation, as in cereals. Other biological materials such as pharmaceuticals and medical supplies must be sterile to prevent infection. Gamma radiation affords a new method of radiopasteurization and sterilization without heat. The advantages are obvious, particularly with heat-sensitive products. However, to weigh possible advantages and limitations, it is necessary to study effects of radiation on organisms involved, as well as radiation effects on complex chemical systems, and the gross effects on such qualities as odor, color, taste, biological value, and nutrition.

The simpler the organism, the more resistant it is to radiation (see Fig. 9.1 in Chap. 9). Also it can be stated that tissues that "grow rapidly" are more sensitive to the effects of radiation than are the slower growing—those whose component cells divide less frequently. Tissues may increase in bulk as the cells composing them grow larger or by increase in the amount of fluid between the cells; however, when we speak of tissues "growing" we refer, as a rule, to an increase in the number of cells. In man, for example, the most sensitive are the bone marrow cells (which produce blood corpuscles) and the cells lining the intestines. Nerve cells, which do not divide in adult organisms, are quite resistant to ionizing radiation. Tumor cells divide more frequently than normal cells. This fact is involved in the use of radiation for cancer therapy: just enough radiation is used to destroy the more sensitive malignant cells without destroying too many normal cells. Microorganisms, which divide at frequent intervals, such as vegetative bacteria, are usually much more sensitive to ionizing radiation than are bacterial spores. For example, the lethal dose for spores is 10 times that for most vegetative organisms. Some exceptions will be discussed.

EFFECT OF LOW DOSES

Microorganisms exposed to low doses of ionizing radiation exhibit slight changes in the rate of respiration and/or cell division. As radiation dose is increased, characteristic changes occur; different organisms exhibit varying degrees of sensitivity.

At irradiation little effect on microorganisms is detected, and some time must elapse before the effects of the irradiation become apparent. After irradiation, most cells will respire normally and show other normal biological characteristics such as motility. However, irradiation appears to interfere with cell division. Some cells may grow gigantically before dividing; when cell division does occur, the daughter cells do not redivide. Inability to reproduce by division results in death to the colony in a relatively brief time, because the life cycle of microorganisms is short.

The effect of low doses, for use in a radiopasteurization treatment, is to reduce the microbial count, and thereby increase the shelf life of various foods, particularly at lower temperatures. It should be remembered, however, that the residual microbial flora may be different from the normal, and any future spoilage may be of a different type from normal.[127]

TARGET THEORY

Many investigators have reported that the dose of ionizing radiation required to kill microorganisms is independent of duration or continuity of the radiation. These observations substantiate the "target" or direct-action theory—that death is caused by one particle only, although many ionizing particles may pass through the bacterium before it is killed. The fatal particle strikes a specially sensitive target in the organism, interacting with the target and killing the organism. Such one-shot killing would be independent of the length of time during which the shots were made. Further, if it were the cumulative effect of many ionizations, one could expect some recovery to be exhibited by the bacterium in time, and higher dosage would be required for sources at lower intensity.

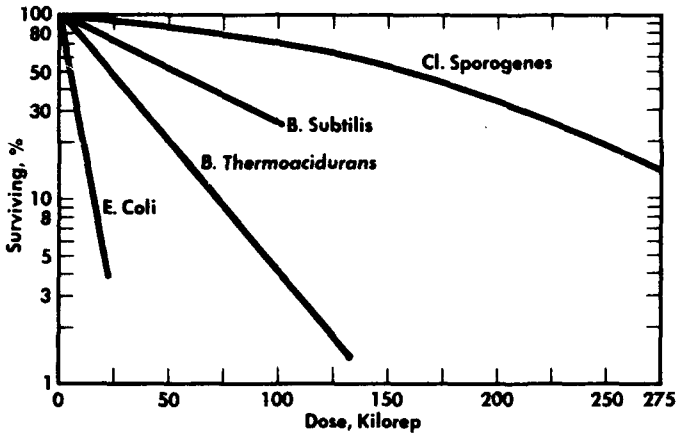


Fig. 8.12—Number of organisms surviving as a function of irradiation dose (semilog scale).[5,66]

The target theory is supported by the exponential characteristic of the survival curve for microorganisms exposed to radiation, as shown by Fig. 8.12 for the organisms *E. coli*, *B. thermoacidurans* and *B. subtilis*.[5,66] The nonlinear nature of some curves, such as those shown in Fig. 8.12 for *Cl. sporogenes* and in Fig. 8.13 for diploid yeasts, is discussed later.

Many observers have noted that, within limits, the percentage of the microorganisms killed by any dose is independent of the initial concentration of organisms. If the chance of hitting a target were 1 in 1000 for a given dose, then $1/1000$ of the bacteria exposed would be destroyed by that dose regardless of the number of bacteria exposed. Some investigators have reported a protective effect at high concentrations, but this might be due to secondary factors such as oxygen utilization in concentrated suspensions. Above the freezing point, wide variations in temperature seem to make little difference in the lethal effects of gamma radiation.

The distance between two ionization events in the absorption of ionizing radiation appears to have a significant influence. More lethal damage may be produced per ionization when the distance between events is sufficient to prevent overlap of effects, i.e., two photons affecting the same cell.

These phenomena can be explained by the target theory in which each cell is considered to have one or more sensitive zones or targets. If an ionization occurs at or near a sensitive zone, the organism is inactivated. For a given radiation dosage the probability of an ionization occurring within a target would be independent of temperature and rate of irradiation. Similarly, the fraction of the targets hit by a given dosage would be independent of the number of targets present.

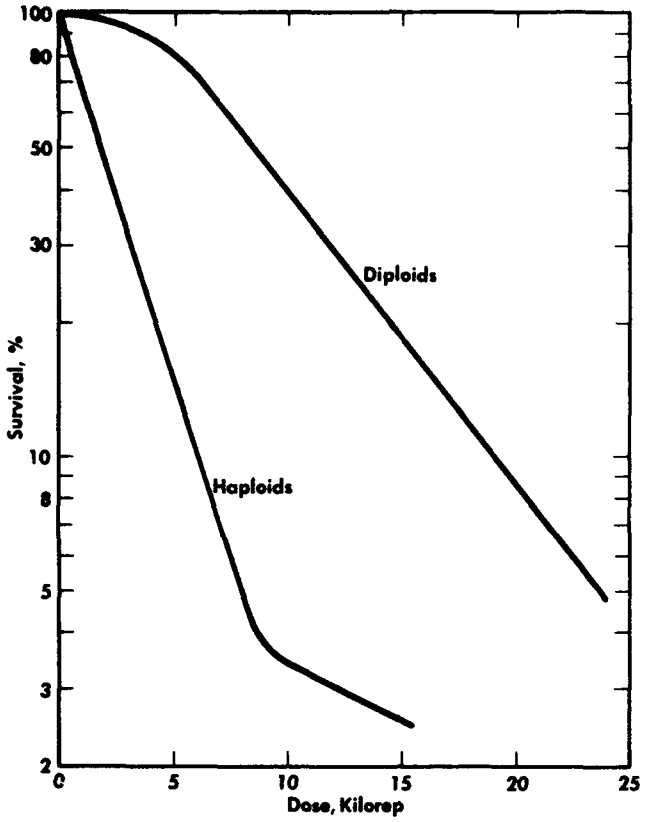


Fig. 8.13—Effect of X radiation on haploid and diploid yeasts of the same family (semilog scale).[103]

When many ionizations occur in the same volume, as in the absorption of alpha particles, more than one ionization may occur in the region of the given target, thereby decreasing the damage per ionization. The efficiency of each ionization event in destroying an organism can be used to estimate the target size. By this procedure, the target size of small viruses in a dry state was found to be of the same magnitude as its nucleoprotein molecule. Study of larger viruses and bacteria indicates that more than one target is involved. The vegetative organism *E. coli* contains an estimated 250 targets having an average diameter of 12 mμ. Note that this dimension is about the size of a gene, possibly indicating that the lethal effects of irradiation are caused by ionizations near the nuclear material of a cell.

An interesting extension of the target theory can be made with reference to the effect of radiation on haploid, diploid, and polyploid yeast cells. The haploids have one complete set of genes, whereas the diploids have two complete sets and the polyploids multiple sets. For an organism with more than one set of genes, it would appear that at least one hit in each set of genes is required to produce death. Mathematically, if only one hit is required to make a kill, the characteristic exponential curve is obtained, but, if two hits are required, the curve shifts to the right and when plotted on semilog paper gives a sigmoidal curve rather than the straight line characteristic of the exponential curve. Similarly, organisms with multiple sets of genes, such as the polyploids, would give still greater sigmoidal distortion in the experimental curve. The data shown in Fig. 8.13 demonstrate this effect.[103]

Although the target theory is essentially based on considerations involving physical effects, the mechanism may be chemical. Thus, if OH or H radicals form in cell near nuclear material, chemical reaction with this material may result. If this is true, the target area must be considered large enough to cover the effective range of mobile free radicals. As this effective range can vary, target size apparently changes with conditions, and there is evidence to this effect. Thus, the simple target theory probably requires considerable qualification to explain all the effects of radiation on microorganisms.

Fischer and Kurtz[147] considered direct action of radiation on microorganisms from the target theory and indirect action as transfer of energy through the medium of cell fluid by excited molecules and free radicals. They consider four possible methods of kill: "(a) A direct hit takes place within the cell and within the target resulting in a biological effect. (b) A direct hit takes place within the cell but outside the target area. Biological effect is due to passage of energy through the cytoplasm. (c) An event takes place outside the cell (indirect hit) but, within the target area, giving a direct biological effect. (d) An event takes place outside the cell and outside the target area. Any biological effect will then be due to the reaction between the cell and its environment."[147] Figure 8.14 illustrates these four possible mechanisms.

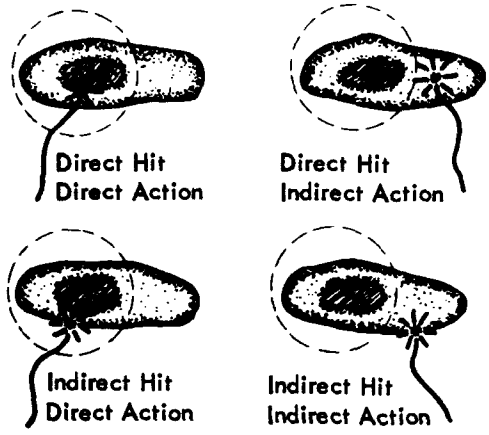


Fig. 8.14—Mechanisms which may cause biological effect.[147]

METHOD OF EXPRESSING DOSE

If the natural logarithm (ln) of the surviving population is plotted vs. dosage a straight line will be obtained for microorganisms following the target theory. This line may be expressed by

$$\ln \frac{N}{N_0} = \frac{1}{D_0} D \tag{8.8}$$

where N_0 = initial population
N = population after dose D
D = radiation dose given
D_0 = radiation dose necessary to reduce population by a factor of e or ~37 per cent survival $\frac{1}{e} = \frac{1}{2.718} = 0.37$

Lea[106,128] has proposed that the dose for 37 per cent survival (67 per cent kill) be termed the "mean lethal dose" and used as a measure of the resistance of microorganisms to radiation. It should be recognized that the "mean lethal dose" is much smaller than the dose for sterilization.

Katzin, Sandholzer, and Strong[129] proposed a similar procedure using common logarithms (Log_{10} which is more convenient when using semilog paper). In this case Eq. 8.8 may be modified to

$$\text{Log} \frac{N}{N_0} = -\frac{1}{D_D} D \tag{8.9}$$

where D_D = dose required to reduce population by a factor of 10 (10 per cent survival)
$= 2.303\ D_0$

The dose D_D is termed the "decimal reduction dose." If the initial portion of the curve is not a straight line on semilog paper as in the case of *Cl. sporogenes* in Fig. 8.12 and diploid yeasts in Fig. 8.13, Eq. 8.9 may be modified as

$$D = D_D \log \frac{N_0}{N} + L \tag{8.10}$$

where L = dose required to overcome lag in curve = dose at intercept of extrapolation of straight line portion of curve and initial population

Rayman and Byrne[130] have demonstrated Eq. 8.10 graphically as shown in Fig. 8.15.

The destruction of microorganisms by radiation is a statistical phenomenon and therefore no absolute value for a radiosterilizing dose exists. However, for practical purposes reductions in population of 10^6 to 10^{10} have been considered sufficient for "commercial" sterilization. If a value of 10^9 reduction in population is considered sufficient and if the initial population was 10^6 organisms per cubic centimeter the final population would be 1 organism per 1000 cm^3. If a can of food contained 1 organism it could be considered sterile because the single organism will not multiply. The reason for this is not understood because microorganisms multiply by cell division rather than by a sexual process. It is believed that a certain minimum population density is required to provide sufficient enzyme concentration produced by the microorganism to provide suitable conditions for growth and multiplication. A value of 4.5 Mrad is at present considered a radiosterilization dose for *Cl. botulinum*.[132]

As an example of calculation of the dose required for radiosterilization consider the following conditions:

$$D_D = 400 \text{ krad}, L = 100 \text{ krad}, N_0/N = 10^9, N_0 = 10^4, D = ?$$

by Eq. 8.10

$$D = 400 (\log 10^9) + 100 = (3600 + 100) \text{ krad} = 3.7 \text{ Mrad}$$

Table 8.15 lists values of D_D for various microorganisms.[130]

SPECIFIC MICROORGANISMS IN AQUEOUS MEDIA AT ROOM TEMPERATURE

Goldblith et al.[104] observed that a given dosage of gamma radiation produces equivalent biological effects to those of the same dosage from high velocity electrons. This might be anticipated from theory, because absorption of gamma radiation by photoelectric effect, Compton scatter, or pair production yields electrons with velocities similar to those produced by accelerators. Therefore, results from either of these types of radiation may be considered comparable. Some care must be exercised, however, in comparing dosages from different laboratories, to be certain the units of radiation absorption correspond.

Bacteria

Table 8.16 lists the data of Dunn, et al.[105] obtained by irradiating sporeforming and nonsporeforming bacteria with X rays. This table shows that the nonsporeformers, i.e., the vegetative bacteria, are the more sensitive to these radiations. Nonspore samples required dosages of 0.5 Mrep or less for destruction. The bacteria that form an inactive, resistant spore, i.e., the sporeformers, however, required doses up to 2.0 Mrep. Table 8.17 shows similar results obtained by Lawrence et al.[102] in studies of the effect of gamma radiation from Co^{60} on some vegetative organisms and some sporeformers. Figure 8.16 shows the series of agar plates for a sporeforming organism originally isolated from canned milk[102] and identified as M-S in Table 8.17.

Lea[107] found that for the vegetative bacteria the dosages required depended upon the type of radiation and the progression was as follows: beta particles, gamma rays, hard X rays, soft X rays, neutrons, and alpha particles. However, the difference between results obtained with beta particles (high-speed electrons) and gamma radiation was not great. Therefore, the

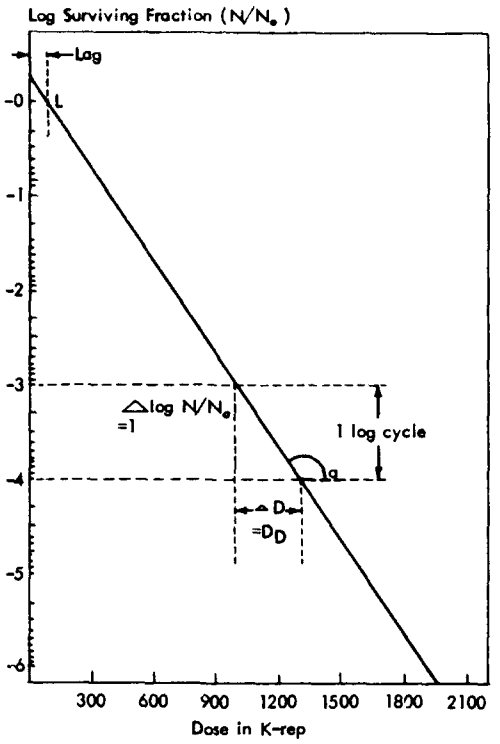


Fig. 8.15—Graphical illustration of Eq. 8.10.[130]

Table 8.15—RESISTANCE OF SOME MICROORGANISMS[130]

Microorganism	Irradiation medium	Type of radiation	D_D Krad	Refs.
Ps. geniculata	Nutrient broth	Gamma	5	Doty[15]
S. aureus	Nutrient broth	Gamma	20	Doty[15]
C. botulinum (spores)	10% gelatin	Gamma	200*	Kempe[132]
S. aureus	M/15 phosphate buffer	Beta	10–15	Bellamy[131]
S. aureus	Dry	Beta	65	Bellamy[131]
B. subtilis (spores)	M/15 phosphate buffer	Beta	75	Bellamy[131]
B. subtilis (spores)	Dry	Beta	80–95	Bellamy[131]
E. coli	M/15 phosphate buffer	Beta	10–15	Bellamy[131]
B. subtilis (spores)	Saline	Beta	260	Proctor[133]
B. subtilis (spores)	Pea puree	Beta	35	Proctor[133]
B. subtilis (spores)	Evaporated milk	Beta	170	Proctor[133]
E. coli	Nutrient broth	Gamma	15–20	Goldblith[134]
E. coli	Nutrient broth	Beta	10–20	Goldblith[134]
B. coagulans	Saline	Gamma	100	Goldblith[134]
B. coagulans	Saline	Beta	110	Goldblith[134]
E. coli	Nutrient broth	50-kv X ray	5	Fram[135]
A. aerogenes	Nutrient broth	50-kv X ray	3	Fram[135]
S. aureus	Nutrient broth	50-kv X ray	10	Fram[135]
S. marcescens	Nutrient broth	50-kv X ray	3	Fram[135]
Ps. aeruginosa	Nutrient broth	50-kv X ray	3	Fram[135]
Ps. fluorescens	Nutrient broth	50-kv X ray	2	Fram[135]
C. botulinum (spores)	Nutrient broth	Gamma	360	Morgan[136]

* L = 1 Mrad (see Eq. 8.10 and Figs. 8.15 and 8.21).

Table 8.16—EFFECTS OF X RAYS ON SELECTED BACTERIA[105]

Organism	Morphology	Gram reaction	No. of organisms per ml × 10^{-6}	No. of megaroentgens necessary for destruction	Special comments
Nonsporeformers					
Aerobacter serobenes	Rods	Neg.	238	<0.25 >0.10	
Escherichia coli	Rods	Neg.	700	<0.15 >0.09	Of sanitary significance
Pseudomonas fluorescens	Rods	Neg.	760	<0.14 >0.14	Produces fluorescence
Serratis marcescens	Rods	Neg.	380	<0.50 >0.25	Produces red pigment
Sarcina flava	Cocci	Pos.	2.4	<0.35 >0.14	Produces yellow pigment
Staphylococcus aureus	Cocci	Pos.	1000	<0.25 >0.10	Produces orange pigment
Spores					
Bacillus mesentericus	Rods	Pos.	6.2	<1.5 >1.0	Strain especially resistant to heat
Bacillus sterothermophilus	Rods	Neg.	75	<1.5 >1.0	
Bacillus subtilis	Rods	Pos.	4.8	<1.0	
Bacillus thermoacidurans	Rods	Pos.	240	<1.0 >0.5	Produces flat-sour spoilage of tomato juice
Canco No. 6B	Rods	Pos.	420	<1.0	Causes spoilage of canned foods
Flat sour (No. 1518)	Rods	Pos.	1.1	<1.0 >0.5	Causes flat-sour spoilage
Spore former from catgut suture	Rods	Pos.	13	<2.0 >1.5	Resistant organism

Table 8.17—RADIATION EFFECTS ON MICROORGANISM POPULATION[102]

Radiation dose, Mrep	Organism and count	Population reduction, %	Organism and count	Population reduction, %
	Proteus vulgaris		*Escherichia coli*	
0	1,666,000,000	0	1,976,250,000	0
0.071	4,500,000	99.7	1,600,000	99.8
0.149	3,680	99.99	500	99.99
0.205	55	99.99	0	100.00
0.289	0	100.00	0	100.00
0.425	0	100.00	0	100.00
	Bacillus subtilis		M-S sporeformer	
0	18,000,000	0	97,250,000	0
0.34	960,000	94.6	8,750	99.99
0.68	202,000	98.8	3,280	99.99
1.02	20,500	98.8	550	99.99
1.36	14,450	99.99	80	99.99
1.70	75	99.99	10	99.99
2.04	0	100.00	0	100.00

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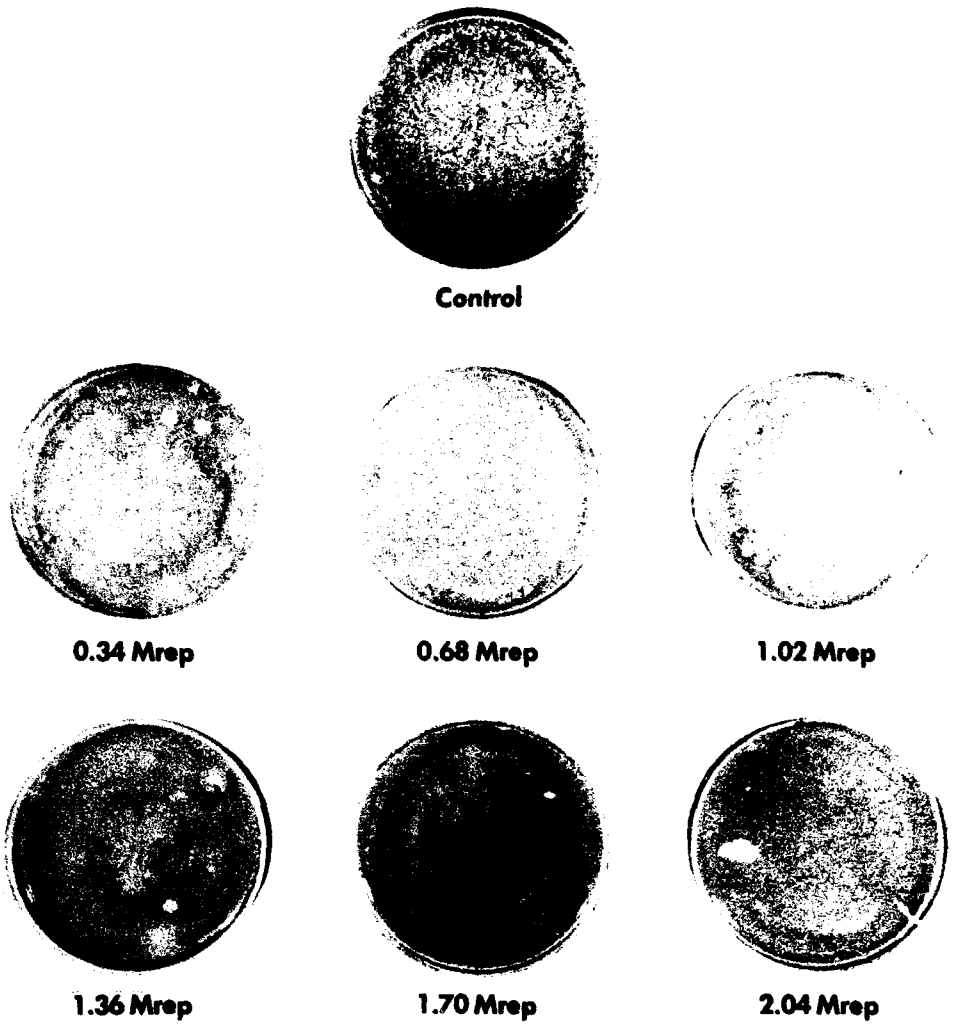


Fig. 8.16—Effect of Co^{60} gamma radiation on spore-forming (M-S) microörganisms in milk.[102]

Table 8.18—MEAN LETHAL DOSAGE, D_0, (37% SURVIVAL)* FOR *E. COLI* AND SPORES OF *B. MENSENTERICUS*[107]

Type of radiation†	*E. coli*, dosage in roentgens	Spores of *B. mensentericus*, dosage in roentgens
Beta particles	4×10^3	1.1×10^5
Gamma rays	5.2×10^3	1.3×10^5
Hard x rays	6.0×10^3	
Soft x rays		
1.5 A	6.5×10^3	1.3×10^5
4.1 A		1.1×10^5
8.3 A	7.5×10^3	1.5×10^5
Neutrons	7.1×10^3	0.26×10^5
Alpha particles	24×10^3	0.26×10^5

*See Eq. 8.8.
†Not all irradiations were carried out under identical conditions.

general observation by Goldblith et al.[104] of the similarity of effects of gamma radiation and electrons is consistent. Lea's results for the vegetative bacteria *E. coli* and the spore *B. mensentericus* are shown in Table 8.18. Note that with the spore the alpha rays are the most effective. Lea explains that for spores the effectiveness of the radiation increases when the specific ionization is increased, i.e., when more ion pairs are produced per centimeter path of the rays. This means that in irradiating spores, the lethal action either requires more than one ionization or at least is more effective if there are several ion clusters. Alpha-particle radiation produces high specific ionization. (It should be noted, however, that in Table 8.18 all the cultures were not irradiated in the same identical condition, some being in the dry state and some in aqueous suspension.)

Lawrence[98] reported some studies on the effects of gamma radiation on a mouse-virulent strain of *Mycobacterium tuberculosis* (Olsen strain) in concentrated suspension which was originally isolated from a human case of clinical tuberculosis infection. Table 8.19 summarizes the data and shows that a dose of 0.24 Mrep rendered the organisms incapable of infection when injected into test animals, but organisms receiving dosages of 0.08 Mrep or less were still capable of producing infection. Other data on the radiation resistance of pathogenic bacteria (as well as nonpathogenic bacteria, molds, and yeasts as determined by Moriarty)[65] are given in Table 8.20.

An important exception to the generally greater radiation resistance of sporeformers as compared to nonsporeformers has recently been reported by Anderson et al.[108] These investigators report that a radiation-resistant micrococcus resembling *Micrococcus roseus* and *Micrococcus rubens tetragenus* was isolated from raw pork and beef after the meat had been irradiated at dosages of 2 and 3 Mrep. The organism was isolated from the irradiated meat, and pure cultures were prepared which required radiation dosages of up to 6 Mrep for destruction. Additional pure cultures were prepared from nonirradiated raw meat and an identical resistance was observed, as shown in Fig. 8.17. This indicates that radiation resistance was not acquired as a result of irradiation but exists in the organism as found in the natural state.

The resistance of another micrococcus (*Micrococcus pyogenes var. aureus*) also is shown in Fig. 8.17 and it is seen that the radiation-resistant micrococcus is six to eight times as resistant. Probably the most interesting point is that this radiation-resistant micrococcus is not a sporeformer and yet has more resistance to radiation than any sporeformer.

Molds and Yeasts

Dunn et al.[105] report some results obtained in irradiating mold growths with high-voltage X rays. Although the term mold has no exact definition, it is generally taken to include all of the wooly, cobweb-like, or powdery growths which develop on food, etc.

A dosage of 250,000 to 500,000 r killed 100 per cent of the *Aspergillus niger* molds, while a dosage of only 50,000 r destroyed 99 per cent of the molds. However, a species of the genus Mucor, irradiated essentially the same way required a dose of 1,000,000 r to kill all the molds; a dose of 500,000 r destroyed 99 per cent of them. These data indicate that different mold species vary considerably in sensitivity to radiation. Table 8.21, reported by Lawrence et al., gives mold counts of two common molds, *Penicillium notatum* and *Aspergillus niger*, as a function of dose of gamma radiation, and Fig. 8.18 shows some of the agar plates used in this study.[102]

Yeasts examined by Dunn et al. appeared to be only slightly more resistant to X radiation than the nonsporeforming bacteria. A summary of these data on yeasts irradiated with X rays is given in Table 8.22 . Data on yeasts irradiated with X rays and with gamma rays are given in Table 8.23.[102]

Table 8.23 indicates that the reduction in count of viable cells by gamma rays emitted from Co^{60} and from X rays are comparable. While there appear to be some differences in the counts in the initial irradiated samples (0.085 and 0.086 Mrep, respectively), the percentage reduction variations are within the limits of error encountered in such a test.

Table 8.19—EFFECT OF Co^{60} RADIATION ON *M. TUBERCULOSIS* (OLSEN STRAIN)[85]

Radiation dose, Mrep	Growth on Petragnani's medium	Animals inoculated	Animals surviving test	Gross pathology	Acid-fast strains
0	Positive	5	2	Positive	Positive
0.08	Positive	5	5	Positive	Questionable
0.24	None	5	2	Negative	Negative
0.48	None	5	5	Negative	Negative
1.92	None	5	5	Negative	Negative

Table 8.20—DOSES OF ELECTRONS FROM A VAN DE GRAAF ACCELERATOR REQUIRED TO INACTIVATE SUSPENSIONS OF VARIOUS MICROORGANISMS[65]

Organism	Dose, Mrep
Bacteria, Nonpathogenic, nonsporeforming	
E. coli	0.100
S. marcescens	0.125
Ps. aeruginosa	0.250
Ps. fluorescen	0.250
Pathogenic, nonsporeforming	
M. pyogenes var. aureus	0.450–0.475
Strep. douchez	0.550
C. diptheriae	0.450
H. pertussis	0.525
D. pneumoniae, Type 1	0.450
N. meningitidis, Type 1	0.525
H. influenzae, Type B	0.550
Molds	
M. mucedo	0.625
R. nigricans	1.125
A. flavus	0.300
A. niger	0.400
P. notatum	0.240
A. terreus	0.350
A. chrysogenum	0.275
Penicillium sp.	0.275
N. crassa	0.600
F. moniliforme	0.275
Yeasts	
S. ellipsoideus	0.525
S. cerevisiae	0.550
Hansenula sp.	0.500

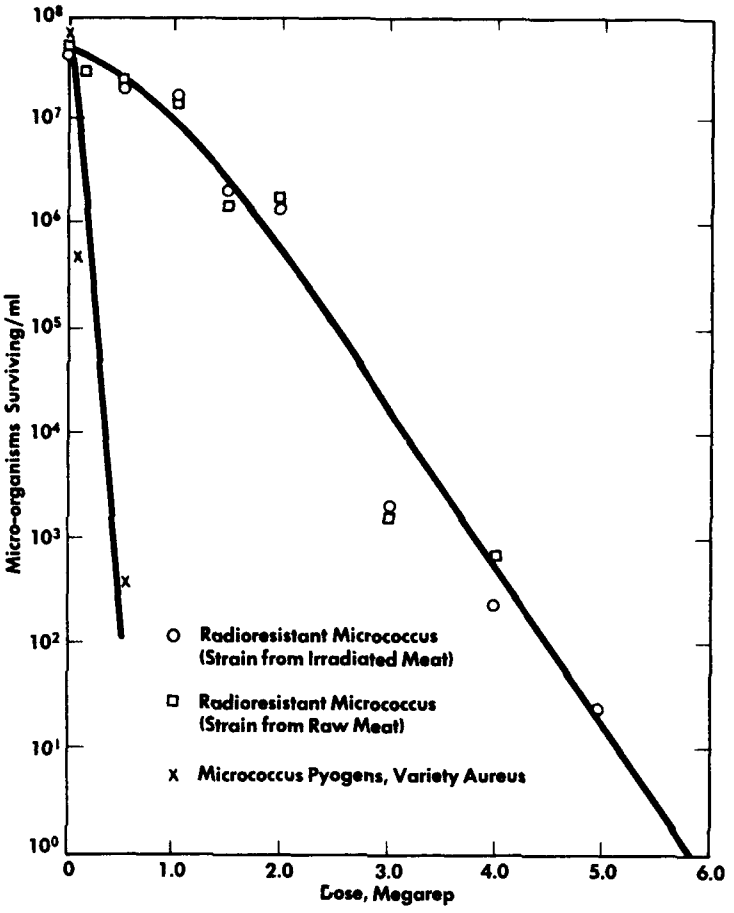


Fig. 8.17—Survival data on radiation-resistant micrococcus as compared to micrococcus pyogens var. aureus.[106]

Table 8.21—MOLD COUNT AS A FUNCTION OF RADIATION DOSAGE[102]

Radiation dose, Mrep	*Aspergillus niger*	Per cent reduction	*Penicillium notatum*	Per cent reduction
0	45,000,000	0	11,250,000	0
0.085	125,000	99.66	15,000	99.86
0.170	5,000	99.98	125	99.99
0.255	25	99.99	0	100.00
0.340	0	100.00	0	100.00

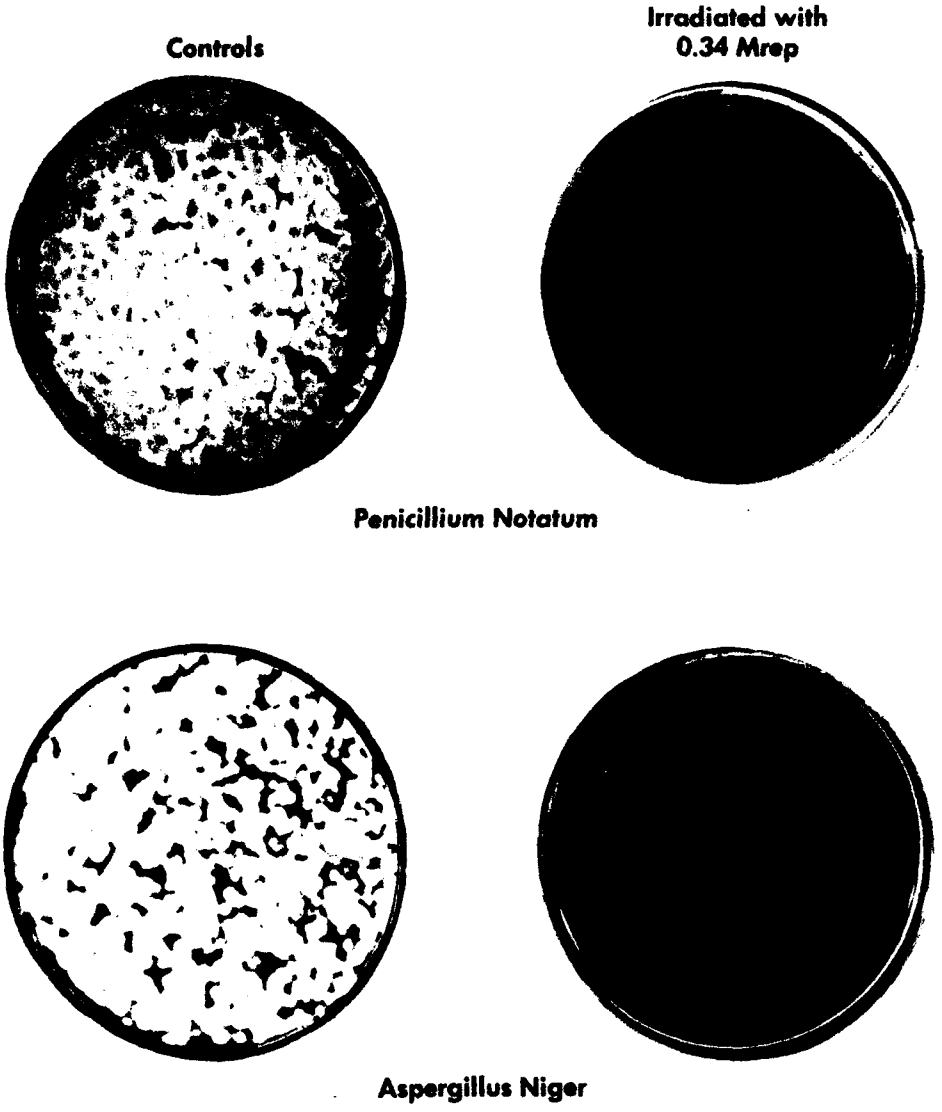


Fig. 8.18—Effect of Co^{60} gamma radiation on molds.[102]

Table 8.22—EFFECTS OF X RAYS ON YEASTS[105]

Name of yeast	Mrep required for destruction
Sacharomyces cerevisiae (strain 1)	0.25 to 0.5
S. cerevisiae (strain 2)	0.25 to 0.5
S. cerevisiae (strain 3)	0.25 to 0.5
S. cerevisiae (strain 4)	0.25 to 0.5
S. cerevisiae (strain 5)	0.25 to 0.5
S. cerevisiae (strain 4) (dry state)	0.50 to 1.0
Torulopsis pulcherrima	1.00 to 1.5
Torulopsis rosea	0.25 to 0.5

Table 8.23—NUMBER OF VIABLE YEAST CELLS *(SACCHAROMYCES CEREVISIAE)* AS A FUNCTION OF Co^{60} AND X-RAY DOSAGES[102]

Gamma dose, Mrep	Viable cells per ml	Per cent reduction	X-ray dose, Mrep	Viable cells per ml	Per cent reduction
0	2,750,000,000	0	0	3,250,000,000	0
0.085	200,500,000	92.72	0.086	3,255,250,000	98.30
0.170	26,250,000	99.05	0.132	10,000	99.99
0.255	32,000	99.99	0.218	9,000	99.99
0.340	25,000	99.99	0.344	5,500	99.99

Protozoa

Lawrence et al.[102] also reported on the protozoa *Trichomonas foetus* (causative agent of infectious abortion in cattle). Motility and number of viable organisms were determined by examining the treated and untreated organism suspensions under the microscope. The data are shown in Table 8.24.

In reference to Table 8.24, Lawrence et al. observed that in all the suspensions that had been given doses of 0.08 to 0.56 Mrep, the organisms appeared to be actively motile. On the other hand, it was only after the higher doses (0.40 to 0.56 Mrep) that there appeared to be some indication of a more sluggish activity than that noted in the nonirradiated controls. Examination following incubation for four days of the medium inoculated with the treated suspensions showed growth in those tubes inoculated with the organism exposed to 0.08 and 0.24 Mrep, but not in the cultures exposed to 0.40 and 0.56 Mrep. While immediate death of the organisms is not observed to take place as the effect of irradiation, some damage to the cells apparently occurs that prevents the organisms from multiplying.

Elliott and Slater[109] made studies of the effect of low dosages of gamma radiation on the protozoan, *Tetrahymena pyriformis*. Dosages up to about 0.15 Mrep had no effect on old cultures, and dosages up to 0.20 Mrep had no effect on young cultures indicating that the older cells were slightly more sensitive to irradiation. Some of the organisms survived dosages as great as 0.80 Mrep.

Table 8.25 gives the lethal dose of various types of radiation for different parasitic protozoa.[137]

Viruses and Bacteriophages

Many diseases are carried by foods contaminated with viruses. The sensitivity of viruses to radiation is therefore important in the irradiation of foods and in the sterilization of pharmaceuticals and medical supplies. Some consideration has also been given to the use of ionizing radiation to reduce a virulent virus to a nonvirulent condition in producing immunization.

Hodes, Webster, and Lavin[110] were able to inactivate rabies virus with ultraviolet radiation and yet retain its immunizing properties more effectively than with the chloroform- or phenol-treated virus. Huber and his associates[111,112] reported on the effects of high-intensity electrons on a number of viruses and also showed that an effective immunizing rabies virus could be prepared by using the capacitron. Table 8.26 shows some of the results of Brasch et al. for the "100 per cent" sterilization of a few viruses and bacteriophages exposed to high-intensity electron bursts from a capacitron. The dosages required to destroy viruses are higher than those reported by these investigators for destroying bacteria, molds, yeasts, and protozoa. This could be explained by the target theory, because the viruses are much smaller. The influence of size is seen in Table 8.26: for example, *Murine encephalomyelitis*, which has a diameter of 8 to 12 mμ, required 1.7 Mrep for 100 per cent sterilization, whereas *Vaccinia*, which has a diameter of 250 mμ, required only 0.6 Mrep for the same result. (The dosimetry methods used by Brasch et al. differ from those used by most other investigators and care must be exercised in using the absolute dosages. However, relative dosages are believed to be in better agreement.)

Table 8.24 — EFFECT OF Co^{60} RADIATION ON THE PROTOZOA *TRICHOMONAS FOETUS*[102]

Radiation dose, Mrep	Motility: Before irradiation	Motility: After irradiation	Growth in 4 days at 37.5°C
0	+	+	+
0.08	+	+	+
0.24	+	+	+
0.40	+	+	0
0.56	+	+	0

Table 8.25 — KILLING OF PARASITIC PROTOZOA[137] BY RADIATION*

Organism	Radiation	Criterion	Dose
Trypanosoma brucei	Soft X rays	Decreased infective power	6,000 r
	UV	Decreased infective power	$^1/_8$–$^1/_{16}$ HSE
T. gambiense	X rays	Visible change	100,000 r
		Failure to infect	12,000 r
T. cruzi	X rays	No visible change	100,000 r
		Decreased infective power	10,000 r
Plasmodium gallinaceum, sporozoites	UV 2537 A	Failure to infect	Not given
P. gallinaceum endothelial stages	X rays	No effect on infection	150 to 700 r
P. gallinaceum sporozoites	X rays	Failure to infect	8,000 r
P. gallinaceum trophozoites	X rays	Failure to infect	20,000 r
		Rate of development of the infection	4,000 to 8,000 r
P. malariae trophozoites	X rays	Failure to infect	5,000 r
P. lophurae trophozoites	X rays	Failure to infect	16,000 r
Eimeria tenella oocysts	UV	No infection of chicks	1 unit
	2537 A	100 per cent mortality *in vitro*	2 units
E. tenella oocysts	X rays	Failure to infect	13,500 r
		Effect on severity of infection	9,000 r
E. tenella	X rays	Distinct effect on severity of infection	9,000 r
		Some infection	13,500 r
E. perforans	UV	Inhibition of *in vitro* development of oocysts	Not given
E. stiedae	2537 A		
Adelina deronis	X rays	Abnormal oocysts	1,000 r
Endamoeba histolytica	UV	Decreasing in number excysting	Not given
E. histolytica trophozoites	X rays	Partial inhibition of growth *in vitro*	60,000 and 120,000 r
		Infectivity unchanged	120,000 r

* Compiled from various sources (see Ref. 137).

Table 8.26—TOTAL (100 PER CENT) STERILIZATION DOSES FOR VIRUSES AND BACTERIOPHAGES WITH HIGH-INTENSITY ELECTRON BURSTS FROM A CAPACITRON[112]

Strain	Diameter, mμ	Total (100 per cent) sterilization dose, Mrep
Viruses		
Murine encephalomyelitis, SK strain	8–12	1.70
Poliomyelitis,		
Aycock		1.55
Lansing	15	1.60
Equine encephalitis western, eastern	35	1.30
Fowl Plague, Newcastle	75	1.00
Mumps		0.80
Influenza,		
Human	100	0.80
Swine		0.80
Rabies	125	0.85
Vaccinia	250	0.60
Phages		
Coli templeton		0.65
Flexner I, type V		0.65
Staph. V.	50	0.60

Lawrence[102] reported some effects of gamma radiation on *Psittacosis* (strain 6BC) and *Human pneumonitis* (strain SF) viruses. The data are summarized in Table 8.27 and indicate that for these two viruses, which are considered to have a relatively large size, a dose of 0.24 Mrep is enough to inactivate the viruses.

Jordan and Kempe[113] reported the inactivation of some animal viruses with gamma radiation. Some of their data are given in Table 8.28.

These investigators[113] concluded that gamma radiation is effective for inactivating *Lansing Poliomyelitis, St. Louis encephalitis, Western equine encephalomyelitis,* and *vaccinia* viruses. They also stated that partly purified suspensions of viruses tested were more vulnerable to lethal effects of gamma radiation that were crude suspensions. In the crude and partly purified suspensions the smaller viruses required larger doses of gamma radiation for inactivation than did the larger viruses, a fact consistent with the results of earlier investigators. The rate of inactivation for viruses tested, within error experiment, is an exponential function of the radiation dosage, as shown in Fig. 8.19.

The bacterophages, which are parasites of bacteria, resemble the viruses in their resistance and several of their other properties, although they are different in their action. These phages, although about the same size as the viruses, require slightly lower dosages for sterilization. The phages investigated by Brasch et al. required doses of 0.60 and 0.65 Mrep for 100 per cent sterilization (see Table 8.27).

Lawrence and Graikoski also studied the effect of gamma radiation on *Staphylococcus aureus* bacteriophage.[102]

CHEMICAL COMPOSITION OF MEDIA

Microorganisms irradiated in aqueous media, as in the test results given in Tables 8.16, 8.17, 8.18, 8.19 to 8.20, are apparently more sensitive to radiation than are microorganisms irradiated in tissue, foods, and similar biological media as given in Table 8.15. For example, Kraybill[114] reported tests with *P. geniculata* and an unidentified species found in fresh meat. Both organisms belonged to the genus *Pseudomonas*. In one experiment a gamma radiation dose of 13 krep destroyed 99 per cent of the organisms in nutrient broth, whereas, for com-

Table 8.27 — EFFECT OF Co^{60} RADIATION ON PSITTOCOSIS AND HUMAN PNEUMONITIS VIRUSES[102]

Radiation dose, Mrep	Psittacosis	Human pneumonitis
0		
0.08	Active	Active
0.24	Inactive	Inactive
0.48	Inactive	Inactive

Table 8.28 — RATES* OF INACTIVATION OF POLIOVIRUS, ST. LOUIS AND WESTERN EQUINE ENCEPHALOMYELITIS AND VACCINIA VIRUSES IN WHOLE BRAIN, CRUDE SUSPENSIONS AND PARTLY PURIFIED STATES[113]

Virus	Diameter, mμ	Virus sample†	Nonirradiated controls	Mrep 1	1.5	2	2.5	3	3.5	4	4.5
Poliovirus	22–27	WB	4.3*	3.3	3.0	2.3	1.9	1.3	0	0	0
		CV	6.0	4.6	4.0	3.3	2.6	2.2	1.4	0	0
		PPV	6.0	4.2	3.3	2.5	1.7	0	0	0	0
SLE	20–30	WB	7.2	5.8	5.1	4.3	3.5	3.1	2.2	1.5	0
		CB	6.0	4.6	4.0	3.3	2.7	2.1	1.4	0	0
		PPV	6.0	4.2	3.3	2.5	1.6	0	0	0	0
WEE	53	WB	8.1	6.3	5.4	4.8	4.0	3.2	2.3	1.6	0
		CV	6.0	4.4	3.6	3.1	2.2	1.6	0	0	0
		PPV	6.0	4.0	2.8	2.0	0	0	0	0	0
Vaccinia	225	WB	5.2	3.7	2.6	1.8	0	0	0	0	0
		CV	6.0	4.1	3.0	1.9	0	0	0	0	0
		PPV	6.0	2.5	0	0	0	0	0	0	0

* All figures represent log of LD_{50} dilution.
†WB: whole brain; CV: crude virus; PPV: partly purified.

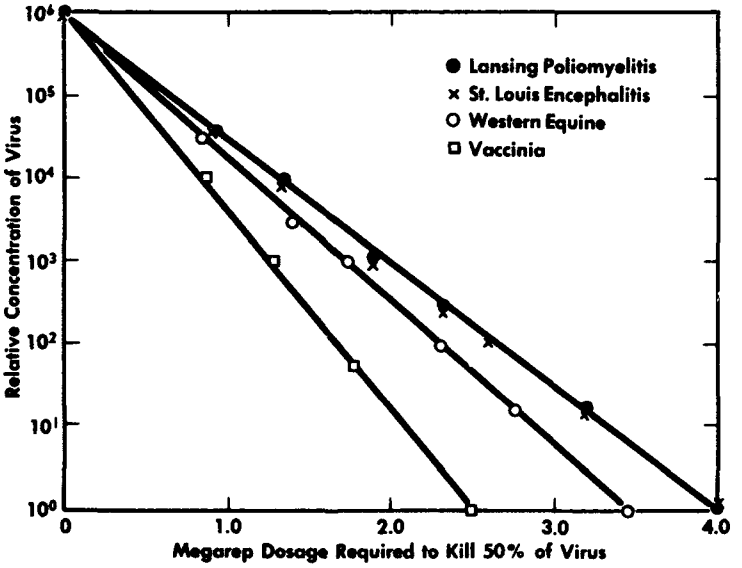


Fig. 8.19—Survival curves of gamma irradiated viruses in crude suspensions (semilog scale).[113]

parable destruction, a dose of 40.5 krep was required when the cells were in raw meat, 28.5 krep when the cells were in washed and cooked meat, and 55 krep when in the juice from cooked meat. Thus, the protective agent appears to reside in this case in the meat juices rather than in cell solids. In subsequent studies at the American Meat Institute Foundation, Doty et al.[15] reported similar protective effects by reducing agents such as ferrous sulfate and reducing sugars. (However this was true only if the ferrous ion was autoclaved with trytone or glucose and if citrate was used as a solubilizing agent.) Data on the influence of Fe^{++} concentration are given in Table 8.29.

Table 8.29—EFFECT OF FERROUS ION CONCENTRATION ON THE RADIOSENSITIVITY OF *PSEUDOMONAS GENICULATA*[15]

Ferrous sulfate, M in medium*	Mean lethal dose, rep	Dose necessary for 99% destruction, rep
5×10^{-2}	4,120	19,700
5×10^{-2}	5,750	20,200
5×10^{-3}	4,080	16,400
5×10^{-4}	1,640	8,220

* Contained sodium + citrate and glucose.

Generalized conclusions on the effect of the medium may be misleading; organisms respond differently when irradiated in different media. Pea puree medium apparently protects *E. coli* appreciably more than does a saline solution. However, the real variable may be the oxygen concentration of the medium.[66]

TEMPERATURE

Temperature has little influence on the lethal effect of ionizing radiation. Thornley, quoted by Hannan,[138] states that "the lethal effect on all types of microorganisms at normal temperatures is little affected by changes in temperature." However, Hannan has shown the protective effect of freezing in low dosage irradiation tests on minced chicken.[138]

A reduction in bacterial population of a factor of 10 is indicated for about every 0.1 Mrad (1 Mrep = 0.93 Mrad) of dosage even though the contamination of two samples differed by 10 (10^5 vs. 10^4 organisms/g). However, some of the more highly contaminated samples irradiated in the frozen state showed less reduction in bacterial population. The protective effect of freezing is believed to be due to immobilization of free radicals which reduces the secondary effects of irradiation. Data of other investigators on other organisms indicate different degrees of protection by freezing for different organisms. For example, freezing apparently has little protective effect on bacterial spores. The heat resistance of spores compared to vegetative organisms might be mentioned as another example of the lack of sensitivity of spores to temperature effects.

The influence of temperature is now being explored by Graikoski and Kempe at the University of Michigan. Their forthcoming publications will offer some information on conflicting reports of some investigators on effects of increased temperature on lethality of radiation.

PRETREATMENT WITH HEAT

Morgan and Reed[136] first reported on the effect of pretreating microorganisms with heat before irradiation. Studies in this field have been conducted by Kempe[115,132] who reported that pretreatment of canned meat with gamma radiation sensitized the remaining spores of *Clostridium botulinum* to subsequent heat processing. The reverse procedure of using a heat pretreatment did not sensitize the remaining spores of this organism to subsequent gamma irradiation. These results are shown in Figs. 8.20 and 8.21, respectively.

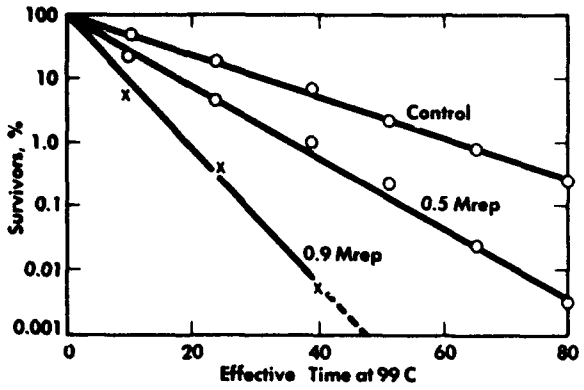


Fig. 8.20—Effect of preliminary gamma irradiation on heat resistance of *Clostridium botulinum* 213 B spores (suspended in 10 per cent gelatin at pH 7.0) Effective time includes time of desired temperature and allowance for heating and cooling.[115]

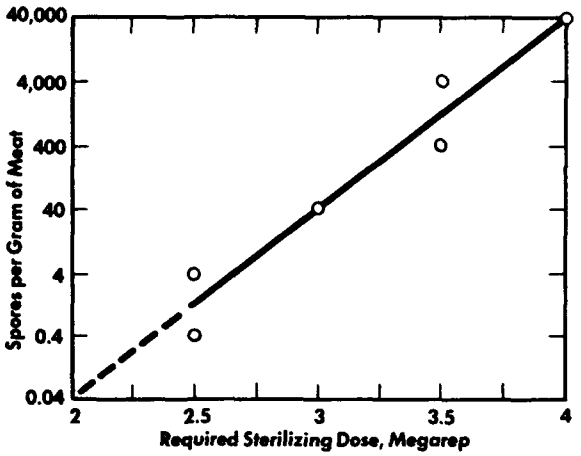


Fig. 8.22—"Sterlizing" gamma radiation dosage for *Clostridium botulinum* (Strain 62 A) in canned meat as a function of initial population (semilog scale).[116]

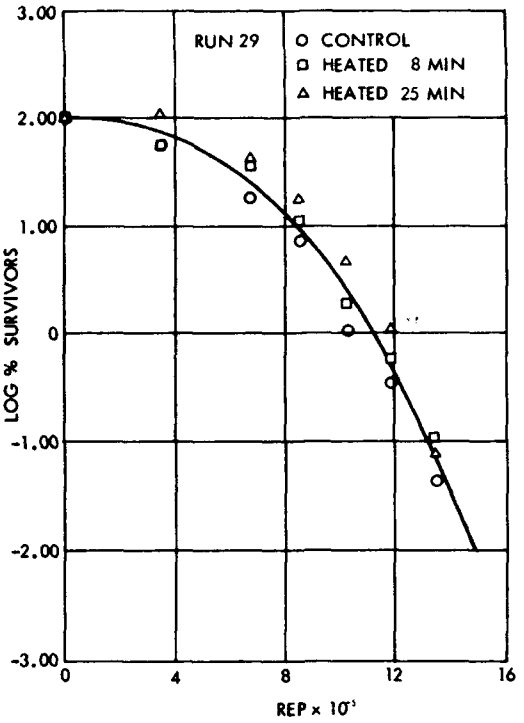


Fig. 8.21—Demonstration of absence of effect of preliminary heating at 99°C on subsequent radiation resistance of *Clostridium botulinum* 213 B spores suspended in 10 per cent gelatin at pH 7.0.[132]

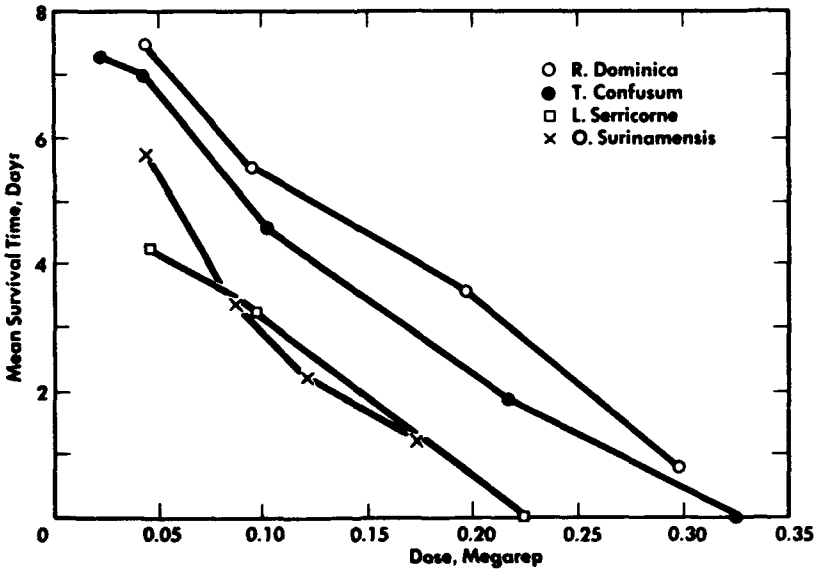


Fig. 8.23—Effect of gamma radiation on mean survival time for adults of four insect species.[139]

Heating at 99°C for 80 min reduces the population in the control 500 times (see Fig. 8.20). The same reduction can be obtained in about 50 min using an irradiation pretreatment of 0.5 Mrep and in about 25 Min using 0.9 Mrep. The obvious significance is that radiation followed by heat can possibly reduce the undesirable effects associated with either treatment alone and thereby improve the quality of sterilized canned foods.

OTHER FACTORS

Numerous factors may influence the lethal effect of gamma radiation on microorganisms. Age of the organism, pH of the medium, oxygen concentration, initial population, as well as postirradiation storage conditions, have an influence on the survival of microorganisms after irradiation. Figure 8.22 from the work of Kempe et al.[116] shows the effect of initial population and radiation dosage on the survival of *Clostridium botulinum* (strain 62A) in cans of cooked meat.

Oxygen increases appreciably the sensitivity of microorganisms to radiation. However, damage to food also is increased, which eliminates any advantages of increasing the oxygen concentration during irradiation.[127] The pH of the medium does not have an important effect.

EFFECT OF IONIZING RADIATION ON SOME INSECTS AND PARASITES

Hassett and Jenkins[117] reported the effects of gamma radiation from Co^{60} on eight species of insects. Most of these tests were made with doses sufficient to destroy adult insects. The General conclusions made for six species were that a dose of 64.4 krep was lethal to adult insects and that doses of 16 to 32 krep inhibited reproduction.

Proctor et al.[139] reported the effect of gamma radiation on the mean survival time of adults of four insect species, as shown in Fig. 8.23.

Dosages greater than 0.30 Mrep were required to kill the insects immediately, but one-tenth of this dose was sufficient to sterilize them.

An extensive series of tests was reported by Baker, Taboada and Wiant.[118,119] As a result of these studies the following conclusions, among others, were stated:

1. "An electron dose of 10 krep will sterilize flour beetle and granary weevil eggs, and this same dose will prevent the adults from reproducing."

2. "A dose of 500 krep was lethal to all adult flour beetles immediately after treatment. A dose of 250 krep was lethal to 92 per cent of adult flour beetles within 1 week after treatment."

MEXICAN FRUIT FLY

Brownell and Yudelovitch[120] studied the effect of gamma radiation on the eggs and larvae of the Mexican fruit fly infesting citrus fruit. An adult fly resting on a grapefruit is shown in Fig. 8.24.

The effect of gamma radiation at egg and first instar larvae stage was investigated, using gamma radiation treatments between 1 and 12 days after infestation. At the temperature at which the fruit was kept, egg eclosion occurred between 6 and 12 days after ovoposition. Within the 12 days after infestation, the gamma radiation acted mainly at an advanced embryological egg stage and probably upon few specimens at first instar larvae. Beyond the 12-day period, the majority of viable eggs were enclosed and some larvae were further than their first instar. The dosages tested at this developmental period were 2, 5, 10, 20, 30, and 90 krep. The results are listed in Table 8.30.

Insect damage or infestation and presence of pupae and fly emergence were evaluated by Brownell and Yudelovitch[120] by frequent observations and dissection of the fruit in checking larval damage or larval mortality in fruit that did not exhibit exit holes or did not present natural recoveries. As shown in Table 8.30 the 5-, 10-, 20-, 30-, and 90-krep dosage groups did not show the presence of insects nor any trace of insect damage in any of the samples dissected. In comparison, the control groups related to these dosages showed an infestation of almost 52 per cent, an abundance of pupae recoveries, and a 48.3 per cent of fly emergence.

Table 8.30 —DATA ON GAMMA IRRADIATED GRAPEFRUIT INFESTED WITH THE MEXICAN FRUIT FLY (AT EGG AND FIRST INSTAR LARVAE STAGE)[120]

Radiation dosage, krep	Number of fruit	Fruit with insect damage	Pupae recoveries	Flies emerging
0 (control)	52	27	151	73
2	10	1	0	0
5	13	0	0	0
10	32	0	0	0
20	13	0	0	0
30	33	0	0	0
90	13	0	0	0

The experiments at this developmental stage involved 166 grapefruit, all exposed to similar conditions of infestation. Only 52 fruit, randomly separated as control, were not irradiated. They proved to be the only group with infested fruit on which the insect normal cycle was completed with subsequent fruit damage. From the 114 fruit that were irradiated at various dosages, no further insect development and damage was found, with the exception on one fruit at the 2 krep dose, which exhibited damage and two dead larvae inside.

Brownell and Yudelovitch[120] also tested the effect of gamma irradiation on full-grown larvae. It was possible in this case to deliver radiation with a certain degree of accuracy within the developmental period and to apply radiation to fruit known to be infested. Infestation of fruit was carried out as in the developmental stages. In this set of experiments, radiation was applied at the time when exit holes were present and only to fruit showing these exit holes.

The fruit with exit holes was allocated randomly between the control and dosage groups. The doses tested were slightly different from those of the previous experiments. It was expected that a more differentiated dose would provide a better basis for evaluating results. Doses of 5, 15, 45, and 70 krep were used; results are listed in Table 8.31.

Table 8.31—DATA ON GAMMA-IRRADIATED GRAPEFRUIT INFESTED WITH THE MEXICAN FRUIT FLY (AT MATURE LARVAE STAGE)[120]

Radiation dosage, krep	Number of fruit	Fruit with insect damage	Pupae recoveries	Flies emerging
0 (control)	19	19	277	174
5	10	10	82	0
15	10	10	53	0
45	10	10	27	0
70	6	6	12	0

Table 8.31 shows many pupae recovered when the mature larvae were irradiated whereas no pupae survived when the irradiation was performed in the egg or first instar larvae stage, as is shown in Table 8.30. However, none of the pupae from irradiated fruit developed flies, whereas about 60 per cent of the pupae from the control fruit produced flies (see Table 8.31).

Brownell[120] and Yudelovitch report additional data on larvae irradiated at intermediate stages and on studies of dissected fruit. The conclusion was made that a gamma radiation dose of 5 krep (and possibly less, based on limited tests at 2 krep) will break the life cycle of the Mexican fruit fly, although the larvae stage may survive appreciably greater dosages.[120]

Table 8.32 gives a summary of the effects of different types of radiation on the reproduction of a variety of insects.[148]



Fig. 8.24—Enlarged view of adult Mexican fruit fly resting on grapefruit.[120]

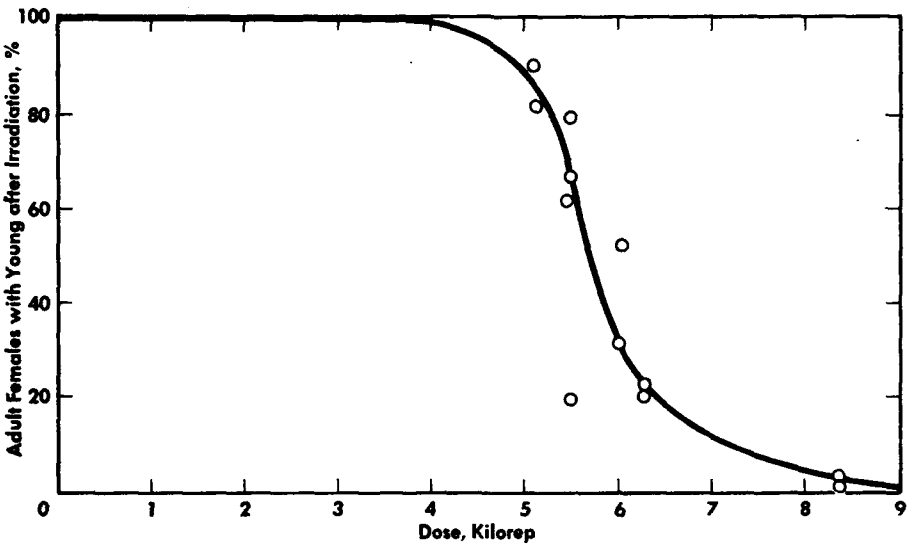


Fig. 8.25—Effect of fission product gamma radiation on trichina larvae in pork.[122]

Table 8.32—EFFECTS OF IONIZING RADIATIONS[148] ON REPRODUCTION IN INSECTS*

Order and species	Type of radiation	Effect on fecundity	Effect on fertility
COLEOPTERA:			
Lasioderma serricorne (cigarette beetle)	X		Adults sterilized by heavy doses
Lyctus planicollis	Gamma	Adults treated with 8050 r = many eggs; 16,100 r = many eggs; 32,200 r = no eggs	
Tribolium confusum (confused flour beetle)	Beta		10,000 rep = 100% adults sterilized
DIPTERA:			
Callitroga hominivorax	Gamma (Co^{60})	5000 r applied to 7-day old pupae; adult fecundity reduced	5000 r applied to 7-day old pupae; adult fertility reduced
			7000 r applied to 5-day old pupae; adults sterile
			12,000 r/hr/1 needed to sterilize 800–900 cc pupae
	X	2500 r applied to pupae; no effect	2500 r applied to pupae; males 90%, sterile, no effect on females
		5000 r applied to 7-day old pupae; adult fecundity reduced	5000 r applied to 7-day old pupae adult fertility reduced
Drosophila melanogaster	Gamma	8050 r = many eggs laid 16,100 r = few eggs laid 32,200 r = few eggs laid 64,400 r = no eggs laid	8050 r = few eggs hatch 16,100 r = no eggs hatch 32,200 r = no eggs hatch 64,400 r = no eggs hatch
	Gamma (Ra)		Males exposed 5.1 r/hr (chronic dose ca 1000 r/wk) capable of reproduction
	X	Decreases with increasing dose; at 10,000 r, 4% of normal	4000 r reduces fertility for 4 days; recovery in 1 week
		0–2500 r, 40% random variation. >2500 r decreases egg-laying; at 20,000 r, 2% normal; at 62,500 r, 0.1% normal	Fertility decreases with increasing dose; 10,000 r, hatch 1/8 normal

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		Portion of life used in egg laying reduced from 75% (normal) to 34%	Fertile period of male decreases with increasing dose
		4000 r reduces fecundity for 4 days; recovery in 1 week	Fertility affected by X rays
Drosophila robusta	X	2500 r (adult) stimulates then retards oviposition, recovery follows. 5000 r (adult) stimulates then retards oviposition, no recovery	2500 r (adult) = poor fertility 5000 r (adult) = poor fertility
HYMENOPTERA:			
Habrobracon juglandis	X	Negative correlation of fecundity with X-ray dose	<1600 r (adult) = no effect 1694 r (adult) = 77.4% fertility 3850 r (adult) = 58.6% fertility 8000 r (adult) = 43.8% fertility $Fertility_{50}$ (adult) = 5900 r
		Preventing egg deposition after irradiation tends to decrease egg loss expected 6–7 days after exposure; caused by repair or resorption of damaged eggs	3300 r (adult) produces temporary sterility; 4800 r (adult) produces permanent sterility 5000 r (adult) produces permanent sterility of female
Mormoniella vitripennis	X		Male mated successfully after exposure of from 568 r to 5112 r
LEPIDOPTERA:			
Bombyx mori	X		Decreased fertility after exposure to X rays
Prodenia eridania	Gamma		Male and female adults sterilized by sublethal doses
ORTHOPTERA:			
Blattella germanica	Neutrons	Irradiation of virgin females causes stimulation of oviposition	Irradiation of adult males causes temporary sterility; irradiation of adult females causes permanent sterility, eggs are nonviable; irradiation of nymphs does not affect fertility

THE PARASITIC NEMATODE, *TRICHINILLA SPIRALIS*

Nematodes such as *Trichinella spiralis* (trichinae) are much more complex than microorganisms. They have the physical size and general biological characteristics of small insects.

Taylor and Parfitt[149] report that, although meat inspection methods have greatly reduced the risk of transmission of parasites to man through the consumption of butcher's meat, parasites like *Trichinella spiralis* and *Cysticercus bovis* (causative agent of beef tapeworm) remain as hazards even in developed countries. Additional information on other parasites is given in Chapter 10. One might estimate that the resistance of nematodes to ionizing radiation would compare to that of insects. This has been substantiated by extensive studies by Gomberg, Gould et al.[121,122] These investigators report that to kill the trichinae *in situ*, a dose of about 0.75 to 1.0 Mrep is required. The lethal dose for isolated larvae was determined from motility measurements on larvae irradiated at 0.27, 0.51, 0.77, and 1.02 Mrep.[121] At the end of each period of irradiation, 2 hr later, and 24 hr later, 200 of the irradiated larvae were examined on a warm stage (110°F) for life as evidenced by motility. In each case the percentage of motile forms was recorded.

One experiment showed that 20 hr after irradiation, death occurred using half the dose required 2 hr after irradiation. Additional tests showed that a gamma radiation dose of only 20 krep is sufficient to prevent maturation of the larvae to adults and a dose of only 12 krep will sterilize the females, thus preventing reproduction. Figure 8.25 shows some of the data obtained by irradiation of trichinous pork with Co^{60} gammas. Similar data were also obtained using gamma radiation from reactor slugs.[122]

In obtaining the data shown in Fig. 8.25 Gomberg and Gould[122] determined the radiation effect by reduction in the ability of the irradiated larvae to mature in 6 days in the intestinal tract and by the absence of trichina cysts in the muscle tissue of test rats 30 days after ingestion of trichina larvae. Each rat was tube-fed with 5000 larvae. For each radiation dose level, whether applied to isolated trichinae or rat muscle infected with trichinae, at least six test animals were used. After 6 days, three of the six rats were killed and the contents of the small intestines examined for adult worms. Gomberg and Gould point out that the very rapid reduction in the number of encysted trichinae is significant. Data appear to give a straight line, implying that inhibition of encystment by trichina larvae is a "one-hit" effect as in the target theory. It is not known whether the effect noted is due to damage of the germ cells in the female larvae so that a smaller number of young are born, or if the young developing from these germ cells are defective and cannot follow the normal course of invasion and encystment. In this connection, the excellent and very complete work of Alicata[123] is of interest, particularly in the photomicrographs showing a reduced number of fully developed egg cells in female adults that received less than the sterilizing dose.

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Chapter 9

EFFECTS OF RADIATION ON PLANTS, FOODS, AND PHARMACEUTICALS

Nuclear radiations can kill living organisms. Working with this radiation is hazardous, and strict precautions must be taken to safeguard life and health. The lethal effects of radiation have given rise to many of the therapeutic uses. They are also the basis for employing radiation to preserve foods. In addition, ionizing radiation can change the genetic make-up of living organisms. From irradiated plants have come mutant strains, some with valuable characteristics of hardiness, such as quick growth, heavy fruiting, and disease resistance.

This chapter describes studies on the treatment of plants, plant seeds, foods, and pharmaceuticals with ionizing radiation. The various applications of radiation are divided into groups on the basis of the radiation dosage applied. The effects of low-dosage irradiation on plants and plant products are treated with special reference to potatoes, fruit, and grain. Problems associated with the irradiation of foods, such as milk, meat, and fish, are discussed together with important studies of the flavor and odor of irradiated foodstuffs. Sterilization of pharmaceuticals by irradiation and its effects on their potency are briefly considered. A discussion of investigations into the possibility that radiation might induce radioactivity in foods, cause toxicity, impair nutritive value, or have carcinogenic effects is included. The chapter ends with a brief review of selected experiments in feeding irradiated foods to animals, and short-term feeding studies with human beings.

DOSE RANGES FOR IRRADIATION

The various biological applications of ionizing radiation can conveniently be grouped into various dose ranges, depending upon the particular objective desired. For example, a dosage of 60 rads is enough to sterilize sexually some mammals, and the effect is considered permanent in females. About 2000 to 5000 rads are needed to destroy the eggs of insects. The larvae and pupae of insects die during pupation after receiving a dose of 5000 to 10,000 rads. Similar doses also can be used on plants and tubers to inhibit sprouting, and on plants and seeds to increase the frequency of mutation or produce modified forms for use in plant grafts. The reproductive sterilization of adult insects in bulk-stored grain and certain packaged products can be achieved with doses of 10,000 to 20,000 rads. The life cycle of certain parasites in meat, e.g., *trichinella spiralis* in pork and *cysticerous bovis* in beef, may be broken by doses of 25,000 to 50,000 rads. Doses of about 70,000 to 100,000 rads kill insects and multicellular parasites.[1] Similar dosages can also be used to produce plant mutants leading to the development of improved varieties.

A dose of radiation of 10,000 to 25,000 rads may be termed "subradiopasteurization" because such doses will not destroy a sufficient quantity of the population of vegetative microorganisms found in fresh foods, such as meat, to affect appreciably the storage life at either refrigeration or room temperatures. This range of radiation dose, however, has very little influence on flavor and color and would be economical to employ because of the high radiation

capacities possible at such low dosages. A summary of the effects of different doses on various forms of life as described by Proctor[2] is given in Fig. 9.1.

A subradiopasteurization dosage has been used for the development of new varieties of beans, oats, and barley through mutation, and could be used for the treatment of potatoes, onions, and other tubers for sprout inhibition; for beans, wheat, and other grain and cereal products for insect control; to break the cycle of trichinosis in pork; to control tapeworm from beef, pork and fish; and to control a number of other diseases caused by parasites. The irradiation of potatoes and wheat, as well as other grains, is considered to be feasible on a mass scale from the viewpoint of both engineering and economics. Because of the tremendous quantities of these staple foods, the application of radiation would be on a large scale.

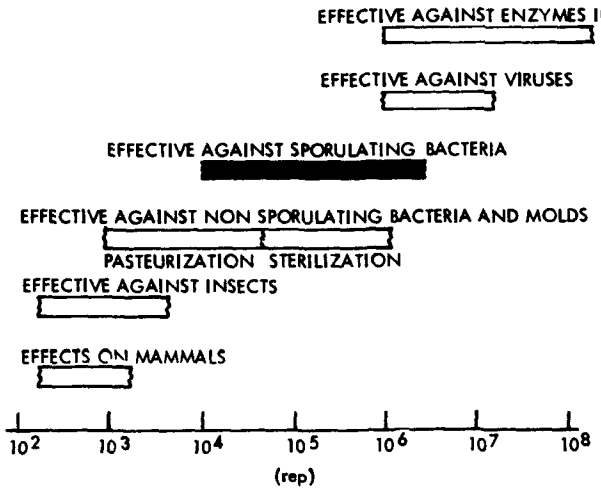


Fig. 9.1—Dose levels of ionizing radiations required for various biological effects.[2]

A dose of radiation less than that required for radiosterilization and in the range of about 100,000 to 500,000 rads might be termed a "radiopasteurization" dose in that, like thermopasteurization, most but not all of the microorganisms are destroyed. Radiopasteurization doses equal to 2 to 5 per cent of radiosterilizing doses can be used to reduce the population of microorganisms 90 to 99+ per cent without producing undesirable changes in food. Such a radiopasteurization treatment could be used to extend the refrigerator shelf life of perishable foods which might make possible new methods of handling.

The organisms causing salmonella food poisoning in egg products, fish, and fresh meats can be inactivated in frozen egg pulp and meats by a dose of 500,000 rads. Similar doses destroy staphlococci and streptococci organisms which also cause food poisoning. The commercial exploitation of radiopasteurization of egg yolks is being considered.

A still higher dose range of about 1×10^6 rads has been termed "high-radiopasteurization." This dosage destroys food poisoning organisms except for spores of *Clostridium botulinum* which can be prevented from growing by refrigeration. Also, yeasts which cause food fermentation and most molds can be controlled with this dosage giving several weeks of storage (with suitable packaging and refrigeration) without freezing. This amount of irradiation can be used with a large number of foods and "ready-to-eat" food preparations without inducing undesirable changes in flavor and odor.

The largest number of studies of food irradiation are directed toward the use of "radiosterilizing" doses in the range of 2 to 5 (depending on the product) Mrad. With this dose and suitable packaging, food may be kept sterile at room (and higher) temperatures for months. Sterilizing doses of radiation may produce undesirable changes in foods, particularly in the odor and flavor of meats and dairy products, and these changes in radiosterilized foods have been a major problem. However, the gains from complete sterilization offer great rewards.

LOW-DOSAGE IRRADIATION OF PLANTS AND PLANT PRODUCTS

It has been found that ionizing radiations have pronounced effects on biological materials. As mentioned, the effects on the seeds, fruits, and tubers of various plants have been studied, to develop improved, mutant strains and to increase storage life. These applications will be considered more completely in the following sections.

PRODUCTION OF MUTANTS

There are three chief uses of radiation in plant breeding that may be beneficial. The first is the production of a desirable somatic "mutation" in asexually reproduced plants where the desirable characteristic is propagated by grafting. Using this technique Bishop[3] produced a Cortland variety apple with higher red coloration that has high economic value.



Fig. 9.2—Leaves from irradiated and control Thomas-Variety black walnut seedlings. (Courtesy of A. L. Barlow)[8]

The second use is the production of useful mutants in crops that reproduce sexually. A number of mutants, particularly cereal grains, with superior resistance to disease have been isolated. Oats with resistance to Victoria blight and crown rust were reported by Konzak.[4] A short-straw strain of rice was developed by Jodon.[5] Mertens and Burdick[6] isolated a strain of tomatoes having increased green weight of plants and earlier development.

A third use of radiation in producing mutants is by causing species chromosomal interchanges. By this technique it is possible to transfer genes between hybrids in which there is a lack of homology between chromosome complements. A wheat with rust resistance was developed by Elliott through this method.[7]

To explore the use of gamma radiation to produce new mutants in asexually reproduced plant strains, seeds of a large variety of plants have been tested. Seeds of one type (Thomas-variety) of black walnuts were irradiated at The University of Michigan;[8] the results are being evaluated by the Northern Nut Growers Association. Since data on the nonlethal limit for gamma irradiation of walnut seed were not available, a preliminary study was conducted with several lots of 50 nuts each using the following amounts of radiation; 1, 5, 10, 20, 30, 40, and 50 krep.

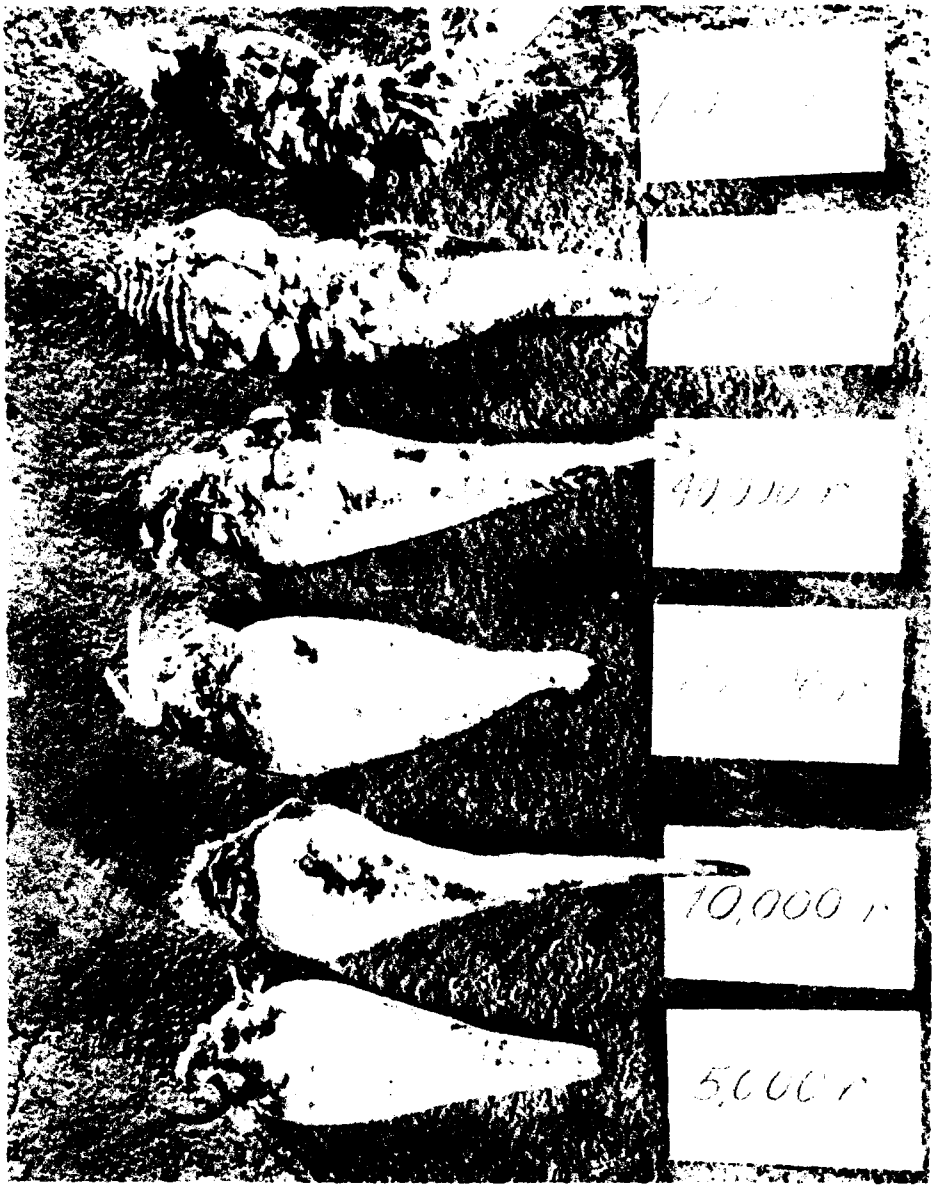


Fig. 9.3—Sugar beet roots grown from gamma-irradiated seeds. (Courtesy of H. E. Brewbaker)[8]

After irradiation, the walnuts were labeled and placed on a mat of sawdust, then covered with several layers of damp burlap to hasten germination. Inspections were made twice weekly to check germination progress and to dampen the walnuts and burlap. When the walnuts germinated, they were planted in plots. When growth above ground was first observed, many irradiated seedlings were greenish and several of the second and third leaves were considerably distorted in shape, as shown in Fig. 9.2. However, leaves coming out later were normal. Damaged leaves do not necessarily indicate plant mutations. Most abnormalities would disappear in the second generation if trees were grown from nuts. Desirable abnormalities, such as thin shells or large nut sizes, may be maintained by grafting. Table 9.1 summarizes early results.[8]

TABLE 9.1— OBSERVATIONS ON THE GERMINATION OF IRRADIATED WALNUTS[8]

Number stratified, Mar. 27	Dosage, krep	Germinations, May 24	Germinations total, July 3	Number growing, Aug. 8	Number growing, Aug. 22
50	0	3	7	5	5
50	1	2	4	4	3
50	5	3	6	6	6
50	10	2	8	7	7
50	20	2	10	9	9
50	30	2	16	12	10
50	40	3	16	3	1
50	50	3	18	1	0

Sugar beet seeds were irradiated for the Great Western Sugar Company. Radiation dosages varied from 5 to 100 krep. The seeds were planted and the beets grown. Typical roots are shown in Fig. 9.3. As beets are grown from seed, true mutants with desirable characteristics must be separated by growing successive generations and inbreeding.

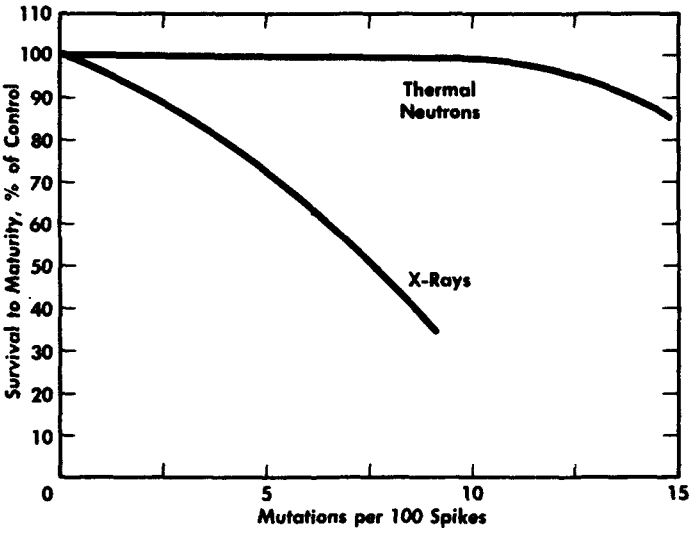


Fig. 9.4—Survival vs. mutation frequency for barley seeds irradiated with X rays and thermal neutrons.[9]

In reference to Fig. 9.3, the distortion in root shape increased with dosage up to 100 krep (there were none at 5 and very few at 10 krep). Most roots showed some distortion at 20 krep. Up to 20 krep, there was little apparent effect on growing beets. At 40 krep, the stand was probably not reduced but the growth of the tops was depressed. The stand was heavily reduced at 50 and all beets were badly affected at 100 krep, with about 99 per cent loss. At the two

higher dosages of 200 and 400 krep, no beets matured, although some germination was observed.

The result of greatest interest—incidence of mutations—will largely be determined in the second generation. After a 40-krep dose, a few roots grown in the greenhouse showed considerable disturbances in pollen formation. Most appeared normal.

It is reported that irradiation with thermal neutrons has an advantage over electromagnet (X or gamma) radiation because the ratio of survival to mutation frequency is higher, as shown in Fig. 9.4.

Plant and animal mutations are produced by radiation damage to the chromosomes in the seeds that carry genetic characteristics of the embryonic cells. When chromosome structure is changed, it may produce undesirable or even lethal characteristics. Since plants are highly specialized organisms, adapted to certain environmental conditions, any change is more likely to be harmful than beneficial. However, the one desirable mutant in a thousand or more makes the irradiation experiments well worth while.

In one experiment using thermal neutron irradiation, seven original pairs of chromosomes were rearranged into two pairs and one large ring of ten produced by the breaking and recombination of five of the original pairs.[9]

Plants are attacked by many diseases. In the case of grain, one strain of wheat stem rust has been particularly devastating. In 1950 and 1951 this disease swept through the Midwestern States. It destroyed plantings of the Lee variety of wheat which is susceptible to this disease. Since then, Hsu and Ausemus at the University of Minnesota have developed by irradiation a mutant Lee wheat that resists rust.[9]

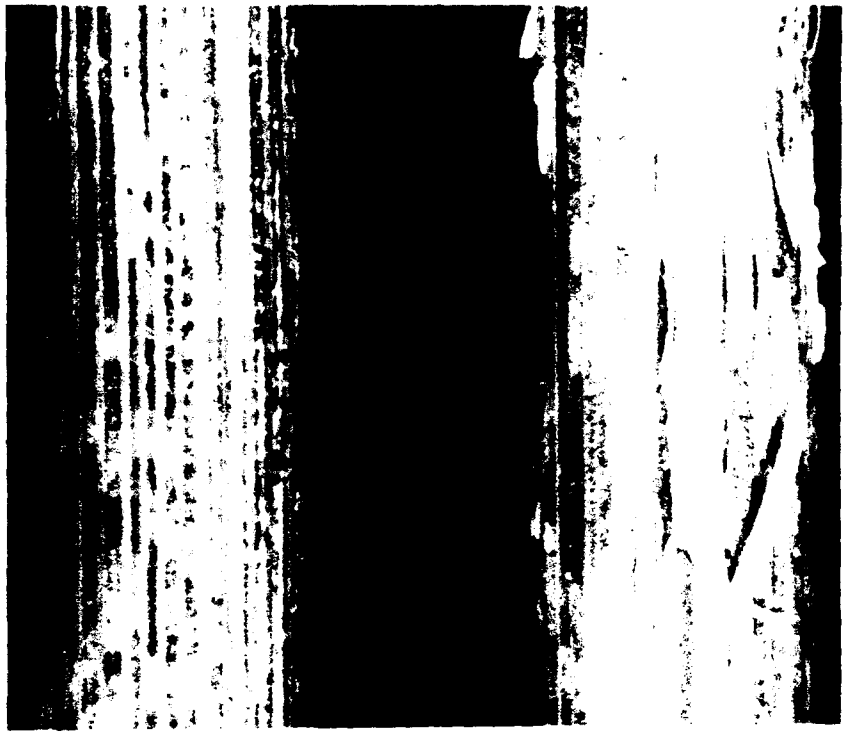


Fig. 9.5—Left: Stem of thermal neutron-induced, rust-resistant Floridaland-variety oats. Right: Stem of rust susceptable parent. (Courtesy of A. T. Wallace, Florida Agricultural Experimental Station, Gainesville, Florida)[9]

Similar studies have been conducted on the Floriland variety of oats at the University of Florida by Wallace.[9] Figure 9.5 shows stems of the original rust-susceptible Floriland oats (right) and the thermal neutron induced mutant (left).

Besides resistance to disease, other desirable characteristics such as early maturity may be induced by irradiation. Early maturing is particularly desirable to adapt plants to regions with short growing seasons. In other climates, late maturing varieties may have advantages. Figure 9.6 shows examples by Wallace[9] of maturity differences in oats induced with thermal neutron irradiation.



Fig. 9.6—Maturity difference in oats induced with thermal neutron irradiation. Center: Typical Florida oat. Left: Induced late long panicle. Right: Induced late panicle. (Courtesy of A. T. Wallace, Florida Agricultural Experimental Station, Gainesville, Florida)[9]

Figure 9.7 shows another example of desirable radiation-induced characteristics. In this case Down and Anderson at Michigan State University developed by X-radiation a new strain (Sanilac) of bean plant from the vine type Michelite variety.[9] This new type is in bush form and has numerous advantages. Caldecott comments that this is one of the best examples "of what has been done with radiation in agricultural crops to produce literally a crop that is more valuable to mankind."[9]

The bush type bean plant shown in Fig. 9.7 has an open crown and is not as susceptible to fungus disease and rot as the vine type Michelite bean. Because it grows upright, it can be

harvested mechanically more easily than vine plants. It is expected that the new Sanilac variety will replace the Michelite variety in more than 400,000 acres in Michigan.[9]



Fig. 9.7—Two plants of the sanilac-bush bean developed from the Michelite-variety by use of X rays. (Courtesy of E. E. Down and A. L. Anderson, Michigan State University)[9]

IRRADIATION OF POTATOES

Early Studies at the University of Michigan[10,11] and Brookhaven National Laboratory[12,13] indicated that low-dosage gamma irradiation of potatoes prevents sprouting during storage. In

1955 and 1956 a study was conducted with support from the Quartermaster Corps of the U. S. Army to investigate more completely the storage properties of irradiated potatoes. Research was conducted jointly at the University of Michigan and Michigan State University.[14]

Weight Loss

In the first phase of the study, Idaho Russet Burbank certified seed potatoes that had been kept in storage for 9 months were given a radiation dose of 10 krep in early June. Storage was continued with control lots of the same variety under temperatures of about 35, 40, 50, 55, and 65°F and room temperature (75 to 80°F). Additional lots of Russet Burbank potatoes were irradiated with doses of 5, 15, 100, and 200 krep; all were stored at 45°F. They were weighed every 2 weeks and checked for sprout development. Sprouts on the control tubers were of maximum size after 3 months. Figure 9.8 shows irradiated potatoes and sprouted controls stored at 60°F.

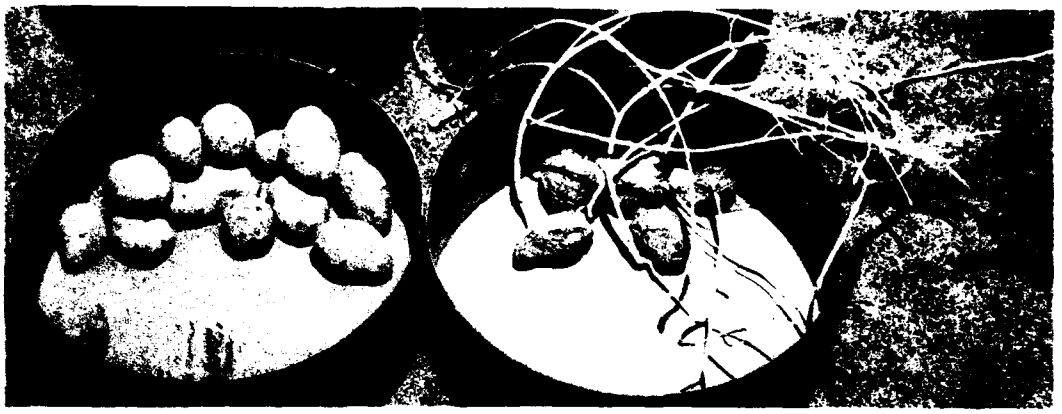


Fig. 9.8—Left: Irradiated Idaho seed potatoes; Right: Control Idaho seed potatoes. Stored at 60°F, at maximum sprout growth (August of year after harvest).[14]

In one experiment irradiated potatoes stored for 140 days at 90 per cent relative humidity (RH) and 50°F, showed that increasing radiation dosage reduces weight loss up to a dosage of about 20 krep. Increasing radiation dosage above this equilibrum level has little additional effect on weight loss. Figure 9.9 shows some of the data and further demonstrates that the equilibrium value weight loss that is independent of radiation dose varies with storage time and relative humidity.[14] The potatoes lose weight for a number of reasons, but, generally speaking, losses may be divided into two groups, physical and biological.

Both living and dead potatoes will lose weight if stored in a dry atmosphere. The skin of the potato tuber acts as a membrane through which water and oxygen, carbon dioxide, etc., may diffuse. The physical weight loss results largely from evaporation of water diffused through the skin, and varies with the partial pressure of water vapor in the storage air, and the velocity of air circulating around the stored tubers. These and other variables which influence drying also effect physical or radiation-independent weight loss. If this value of radiation-independent weight loss is subtracted from total weight loss, one obtains the remaining biological weight loss, which is radiation-dependent.

For the Idaho potatoes used in these studies, the radiation-dependent component of weight loss showed a logarithmic reduction with radiation dosage. This phenomenon tends to support a hypothesis of weight loss proportional to the number of living cells, if the cells are made inactive by a random, statistical process. This is similar to the target theory of radiation effects on microorganisms. Extensive data from many laboratories on a wide variety of unicellular organisms indicate similar exponential radiation inactivation phenomena (see Chap. 8).

Later studies indicated that there is a reverse effect in which weight loss increases with radiation dosage. This effect, although not observed in the Idaho seed potatoes, was controlling in the case of Michigan-grown (Sebago variety) potatoes.

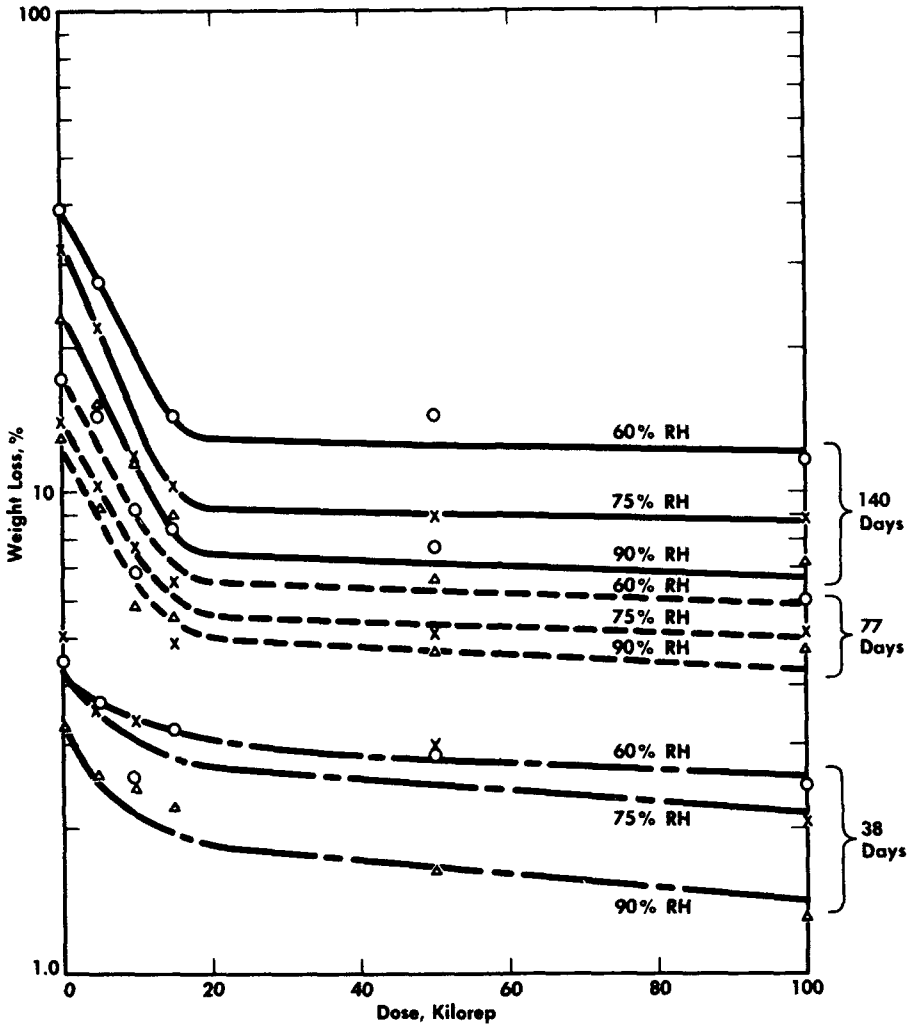


Fig. 9.9—Weight-loss data for Idaho seed potatoes as a function of radiation dose, length of storage and relative humidity.[14]

Suberization and Periderm Formation

In studies conducted at Michigan State University by D. Isleib,[14] Sebago, Russet Rural, and Katahdin varieties of potatoes were irradiated and then damaged by cutting with a knife to study the healing processes. At 6 hr after damaging only the control tubers showed any suberization (healing characterized by cork-like growth). Traces of suberization were apparent in treated sections after 36 hr, and at 48 hr there was little to distinguish the suberization in treated sections from that in control sections. Periderm formation (a new second skin produced by cell division) in controls was not evident at 48 hr, but was well developed after 96 hr. There was no evidence of periderm formation in either of the two treated series even after 196 hr; cell division was apparently inhibited completely in treated potatoes. Beginning at the 48-hr stage and continuing through 192 hr, suberization was essentially the same in both treated and control potatoes.

Results indicate that sprouting inhibited by irradiation is not caused by decreased activity of growth hormones or increased amounts of growth inhibitor. Actually there appeared to be

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an increase in growth hormone and a decrease in inhibitor as a result of irradiation. These results are supported some by earlier studies,[10] in which low doses of radiation (10 to 50 krep) caused irradiated onions and walnuts to sprout faster than controls. However, the sprouts on the irradiated samples were short-lived, dying after reaching a length of about ½ in; whereas the later-sprouting controls continued to grow. These limited observations support the theory that growth hormones may be increased and/or growth-hormone inhibitors decreased as a result of gamma irradiation.[14]

Studies on periderm formation in cut potatoes also indicate that gamma irradiation inhibits cell division. This checks with general observations of the effect of low doses of gamma radiation on microorganisms. At the time of irradiation, there is little observable effect on the microorganisms; some time must elapse before any effects become apparent. Most cells continue to show normal biological characteristics such as respiration and motility. However, the irradiation appears to interfere with the process of cell division. Some individual cells may grow to giant size before dividing and, when cell division does occur, the daughter cells do not redivide.[14]

The hypothesis is proposed by Brownell et al.[14] that sprouting is inhibited in irradiated potatoes by an induced mechanism which interferes with cell division. Growth hormones may be stimulated as a response to cell damage caused by irradiation.

Carbohydrate Analyses

In the same study irradiated and control tubers were analyzed for the percentage of reducing-sugar, sucrose, starch and ascorbic acid content. Results showed that increasing the storage temperature of potatoes irradiated with 10 krep of gamma radiation decreased reducing-sugar content from 2.7 per cent for potatoes stored at 35°F for 6 months to 0.35 per cent for potatoes stored at approximately 80°F. Potatoes receiving doses of radiation ranging to 200 krep, but stored under the same temperature conditions for 5 months, showed no consistent change in reducing-sugar content. Table 9.2 gives values of reducing sugar, sucrose, and starch for control and 15-krep irradiated Russet Rural variety tubers stored at various temperatures for 8 weeks.

Table 9.2 indicates that low radiation doses have little effect on the concentrations of reducing-sugar, sucrose, and starch, even under extended storage. In general, higher dosages

Table 9.2—CARBOHYDRATE ANALYSES ON RUSSET RURAL POTATOES[14]

		Control				Irradiated 15 krep			
Time, weeks	Temp., °F	Reducing sugar, %	Sucrose, %	Starch, %	Total, %	Reducing sugar, %	Sucrose, %	Starch, %	Total, %
0		0.43	0.19			0.43	0.30		
8	35	1.71	0.47	13.1	15.3	1.25	2.71	13.4	17.4
8	45	0.50	0.28	20.7	21.5	0.40	0.81	21.4	22.6
8	55	0.27	0.31	20.4	21.0	0.43	0.45	20.4	21.3
8	65	0.37	0.25	20.9	21.5	0.55	0.20	20.9	21.7
8	Room (70 ± 10)	0.36	0.46	22.5	23.3	0.37	0.49	17.1	18.0

and lower storage temperatures increase the sucrose concentration. Studies on enzyme systems indicated a sharp reduction in ascorbic acid content at radiation doses of 10 krep, and an additional but more gradual reduction at higher doses.

Respiration

Studies of gamma-radiation doses of 5, 10, and 15 krep on potato-tissue slices showed no significant effect on respiration. However, doses of 50, 100, and 200 krep resulted in higher

respiration rates. This effect continued with time of storage. Respiration studies on whole tubers gave results similar to the tests on slices for doses of 50, 100, and 200 krep. Low doses of 5, 15, and 25 krep showed a drop of about 30 per cent in respiration rate during the first day after irradiation; respiration increased about 60 per cent on the 2nd day, and gradually declined to about 5 per cent at the 13th week of storage.

Problem of Potato Rot

Studies by Hooker at Michigan State University[14] on Sebago variety potatoes field-infected with different rot organisms indicated that gamma radiation decreases incidence of ring rot on potatoes stored at 20°C but has no significant effect on the same kind of tubers stored at 1°C. However, various other forms of rot increased at both storage temperatures with increased radiation dosage.

Later Observations

Further investigations were conducted in 1957 at Michigan State University and at Purdue University. The studies were designed to include potatoes grown in four areas of the country, dosages between 5 and 15 krep, various curing schedules, and various storage temperatures. In general it was observed that if the potatoes are irradiated after a lapse of time following digging, the detrimental effects are decreased.[22]

In 1959 Tripp[15] reviewed the results of the studies on potato irradiation supported by the U. S. Army Quartermaster Corps. He stated that "Sizeable quantities of sprout-inhibited potatoes of the 1956 crop are still in good condition. These are Russet Burbank tubers which were treated with relatively low doses of radiation in the range of 7500–10,000 rads. Several tons of potatoes from the 1957 crop are in excellent condition. In both these lots sprouting, shrinkage and decay are negligible."

Studies of Potato Irradiation in Europe

Studies on irradiated potatoes have been underway in Russia since 1955. Extensive investigations have been made on the chemistry, odor, taste, and cooking qualities. On the basis of these tests the USSR Chief of Public Health Inspection authorized the use of potatoes given a dose of 10,000 rads for human consumption.[16] A large capacity radiation facility has been constructed and the process is in use.

Norwegian studies[17] indicated that radiation doses as high as 16,000 rads could be used on potatoes. Similar studies in France indicated that doses higher than 10,000 rads increase spoilage and that doses of about 7500 rads are to be preferred.[18] In 1959 Vidal[19] of France commented that "For short term storage doses of 5000 to 6000 rads are sufficient but for long term storage (12 months or longer) it is necessary to use doses of 7500 to 10,000 rads."

Burton and de Jong[20] of England reported extensive studies on irradiated Ware variety potatoes. They stated "The effect of irradiation (high level) is thus to stop sprouting completely but not to prolong storage indefinitely . . . Irradiation at a low level—say 5000 rads—either does not induce harmful side-effects or does not induce them so soon but it may not always stop sprouting completely. The irradiation dose should be the minimum which will give commercially acceptable sprout suppression—certainly less than 10,000 rads."

American studies have shown that the permissible dose varies appreciably with variety, length of storage before irradiation, and storage conditions. This may explain some of the differences in doses recommended by European investigators.

IRRADIATION OF ONIONS

Preliminary studies were reported by Brownell et al.,[8,10,11] on the irradiation of White Pearl and Bermuda onions. In the tests with Bermuda onions, four groups of onions in 50-lb mesh bags were placed in storage in May, at 50°F and 50 per cent humidity. One group was the control group (not irradiated); three groups had been exposed to gamma radiation of 7, 21, and 42 krep, respectively.

Onions and potatoes seem to respond differently to irradiation. During initial storage both irradiated and control onions developed small sprouts. In the case of the White Pearl onions the irradiated onions sprouted first. In all cases sprouts on irradiated onions grew only a short distance (about ½ to 1½ in.) and then withered and died. On the control onions, sprouts continued to grow up to 6 to 8 in. All irradiated onions remained dry and firm (but lost some crispness) whether or not they had sprouted; control onions became moist and soft.

The contrast in appearance of the control and irradiated Bermuda onions is shown in Fig. 9.10. This photograph was taken in the storage room, 4 months after irradiation. It shows



Fig. 9.10—Comparison of irradiated and nonirradiated Bermuda onions showing the sprouts of nonirradiated onions protruding several inches through the mesh bags.[8]

the long sprouts of the control onions protruding several inches through the mesh bags and no sprouts on the irradiated onions.

Tests of the irradiated onions after storage indicate some loss in crispness and some loss in pungency. Brownell et al. concluded that a 7-krep dosage of gamma radiation may not completely prevent initial sprouting, but it inhibits the continued growth of sprouts, keeping the onion more firm and increasing storage life.[8]

Additional studies on extending onion storage life by gamma irradiation have been reported by Dallyn et al.[21] and in 1959 Bednarczyk[23] reported on tests made in Poland with irradiated onions. It was found that with this crop it was advantageous to minimize the time between harvesting and irradiation. Two weeks delay was considered to be the maximum permissible to avoid darkening of the vegetative cone.

IRRADIATION OF OTHER ROOT CROPS

Studies in Norway on irradiated carrots indicated that doses of 5000 to 10,000 rad were beneficial and decreased rotting. If the dose was increased to 15,000 rad,[17,24] the rotting increased markedly (see Table 9.3).

Mikaelsen also reported that studies had been made in Norway with irradiated red beets, turnips, and Jerusalem artichokes.[24] In all cases sprout inhibition was obtained with doses of 5000 to 15,000 rads of gamma radiation. As in the case of potatoes and onions the extent of rotting increased with dosage.

Table 9.3—THE EFFECT OF GAMMA-IRRADIATED CARROTS STORED FOR SIX MONTHS IN PLASTIC BAGS*[24]

Dose, rads	Growth, % control	Weight loss, %	Rotting, %
0	100	38	53
5,000	25	36	21
10,000	14	33	9
15,000	0	56	73

*Note the sharp break in results at 15,000 rads similar to that shown in Fig. 9.9.

IRRADIATION OF SEEDS

Kuzin[25] reported Russian studies of the irradiation of seeds to stimulate the initial stages of growth and to raise crop yields. This work is reviewed here briefly because any beneficial effects of the irradiation of seeds appears to be related to the effects of irradiation observed on potatoes, onions, and walnut seeds. These observations indicated a two-fold effect of radiation: (1) interference with cell division at doses of 5000 rep and greater as indicated by inhibition of continued growth of sprouts; (2) stimulation of growth hormones as shown by more rapid sprouting of irradiated onions and hormone studies on potatoes. However, if the radiation dose is kept sufficiently low, it is believed that stimulation of growth may be effected without significant interference with cell division.

In the Russian studies the most beneficial dosages usually ranged from 300 to 1000 roentgens.

Rye seeds given a dose of 750 rep, produced roots having a diameter of 393 μ on the 3rd day of development as compared to 304 μ for nonirradiated rye seeds used for control purposes. This observation is theoretically important because the increase in root diameter is due not to the growing size of the cells, but to the increase in their number. For example, the number of cells in the subepidermal layer of shoot roots was 68 for the rye seeds given 750-rep dose as compared to 40 for the control rye seeds given 0-rep dose. The root length on the 4th day of development was 52.2 mm for the 750-rep rye seeds as compared to 43.5 mm for the control.

Similar results on stimulation of plant growth have been observed with other species. On the 5th day of development radish, pea, and cucumber shoots from seeds given an X-ray dose of 500 rep had shoot lengths of 72.2, 61.2, and 89.5 mm, respectively; whereas the respective untreated seeds had shoot lengths of 50.0, 53.6, and 82.6 mm, respectively.

Yields per unit field area were also increased. In the case of radishes grown in a greenhouse a dosage of 1000 rep gave a 30 per cent increase in total weight of tuber yield as compared to the controls. Field grown radishes from seeds receiving the same dosage gave a 40 per cent increase as a result of irradiation. Cabbage seeds given a dosage of 1000 rep produced plants that ripened earlier than the control and gave a 19 per cent increase in yield per area. The harvest yield of peas given an X-radiation dose of 350 rep produced 10 per cent more pea seeds per plant. Furthermore, the weight of 1000 pea seeds was 16 per cent greater for the yield from irradiated plants than from the controls. The irradiation of soaked pea seeds with Co^{60} gamma radiation gave up to 50 per cent increase in weight of peas per plant and up to 24 per cent increase in the vegatation mass as compared to untreated controls. Irradiation of rye seeds (750 to 1000 rep) resulted in 21 to 22 per cent increase in the weight of 1000 harvest grains.

IRRADIATION OF FRUIT

Experiments have been reported on the irradiation of various fruits: apples, berries, cherries, peaches, plums, and tomatoes.

Apples

Apples, unlike most foods, are reported by some investigators[8] to improve in flavor upon irradiation. However, unblanched apples given radiation doses of 2 Mrep or more will discolor. These dosages produce a brown color like that of rotten apples, throughout the fruit. However, initial tests with considerably lower dosages (50 krep) have been quite promising.

In one experiment several varieities of apples grown in Michigan were wrapped in polyethylene film (without sealing), irradiated at doses of 50, 100, and 200 krep and placed in commerical storage in December with nonirradiated (control) apples wrapped in the same manner. Varieties used were Delicious, Golden Delicious, and Northern Spy. The apples used for the major experiment had been held in cold storage from September to December; after irradiation they were returned to cold storage. One year from the harvest date, about half of the apples irradiated at 50 and 100 krep were still crisp, juicy, and in excellent condition. A slightly lower percentage of those irradiated at 200 krep were also in excellent condition. The remainder were spoiled by rot and mold. Apples with blemishes rotted, whether irradiated or not. All control apples had spoiled by the end of May following harvest.

In a supplementary experiment, apples were irradiated immediately after harvest prior to cold storage. The irradiated apples kept only slightly longer than the controls; however, only a few samples were employed in this experiment. Storing apples for over a year with the use of radiation is promising, but requires additional study.

Kertesz[26] studied the reaction of apples to ionizing radiation and found an increasing loss of texture with increasing radiation which was correlated with changes in pectic substances.

McArdle and Nehemias[27] measured the changes in texture of apple tissue exposed to gamma radiation using a Magness-Taylor pressure tester (see Fig. 9.11).

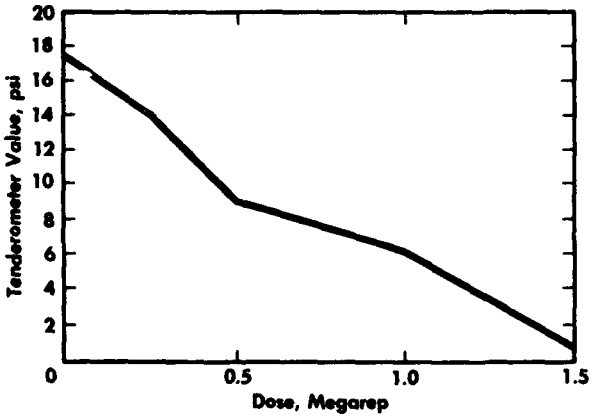


Fig. 9.11 — Effect of gamma radiation on texture of apple tissue.[27]

Delicate fruits, such as berries, are damaged when irradiated with a sterility dose. They may lose cell fluid, bleach, or deteriorate in appearance and texture. Fresh fruits and vegetables soften; a certain amount of cell destruction takes place, evidenced by increased tenderness, decreased crispness, and the loss of some fluid from the cell. Irradiated peas cooked for 3 min are as tender as nonirradiated controls cooked for 6 min.[28]

Cherries and Peaches

Studies on irradiated cherries and peaches were reported by Brownell and Nehemias.[28] The fruits were individually packaged in polyethylene and saran bags and given dosages of

radiation from 200 krep to 2 Mrep. The irradiated packages of fruit were immediately stored, either at room temperature (70°F) or at refrigerator temperature (40°F), in such a way as to allow visual inspection. They were then inspected daily, and, if mold was detected on a sample of fruit, that sample was removed. No other criterion was used in this series of experiments.

For cherries stored at room temperature, the nonirradiated control samples molded after 8 days. At doses higher than 1.5 Mrep a substantial majority of the cherries were still free of mold even after 20 days at room temperature. At doses below 1 Mrep the irradiated fruit molded faster than the control. Mold occurred much more slowly under refrigeration. With doses less than 1 Mrep, mold formed faster than in untreated fruit. Some cheries had not developed mold after 90 days.

Nonirradiated samples in saran bags stored at 40°F molded more slowly than the treated samples, even up to 2 Mrep.[28]

If increasing the radiation dosage decreases the number of organisms present, an apparent anomaly exists. One explanation might be that radiation changes the fruit, physically or chemically, so that mold grows more readily. At lower doses this effect apparently stimulates growth rates enough to more than compensate for the fact that fewer organisms are initially present. At higher doses, the probability of killing completely all mold organisms on a particular sample of fruit increases, and more and more samples will be free of mold formation regardless of growth-promoting factors. Under most of the experimental conditions studied, radiation doses above 1 Mrep definitely retarded mold formation,[28] as compared to untreated samples stored at either 40 or 70°F.

Unfortunately no data were taken by these investigators on fruit at radiation doses lower than 0.2 Mrep. If cherries and peaches follow the pattern indicated by apples, a radiation dose of 0.05 Mrep or possibly less might prove beneficial in increasing the storage life of the fruit.

Plums

Hannan[29] reported the effect of low doses of 200-kv X radiation on the condition of plums (Giant Prune variety) irradiated in the near-ripe condition and stored at room temperature (about 18°C). The irradiated samples for each dose, together with an equal number of controls were stored under greaseproof paper to prevent excessive wilting. The results are given in Table 9.4.

Table 9.4—EFFECT OF LOW DOSAGES OF RADIATION ON THE STORAGE OF GIANT PRUNE PLUMS[29]

	Percentage sound fruit					
Storage time, days	Irradiated, 1 krep	Control, 0 rep	Irradiated, 10 krep	Control, 0 rep	Irradiated, 50 krep	Control, 0 rep
0	100	100	100	100	100	100
3	100	100	100	100	100	92
5	90	90	95	88	92	92
6	90	70	85	77	84	58
7	90	70	80	65	84	58
8	90	70	75		84	58
10	75	45	58	48	67	58
12	65	50	53	40	67	38
14	50	25	48	40	67	50
16	40	25	38	27	58	42
19		20	33	27	42	34

In 1959 Clarke[30] reported that plums given a 400,000-rad dose had a microbial count 5 days after irradiation of 0 mold and 140 yeast as compared to 260 mold and 17,000 yeast for the nonirradiated controls.

Strawberries

Extensive studies have been made at the Wantage Laboratory in England on irradiated strawberries.[30] Doses above 500,000 rad were found to have a marked effect on quality. Texture and flavor suffered severe destruction and the berries were bleached considerably. At a dose of 200,000 rad little change in flavor was observed. Data on the incidence of mold are given in Table 9.5.

Table 9.5—PER CENT INCIDENCE OF MOLD ON IRRADIATED STRAWBERRIES (CAMBRIDGE SPECIAL)[30] STORED AT 20°C

Days stored	0 rad	200,000 rad	300,000 rad	400,000 rad
0	0	0	0	0
3	65	0	0	0
4	100	8	0	0
5	100	20	8	0
7	100	40	18	5

An interesting observation in these studies was that irradiated berries stored at 1°C for 40 days showed no mold regardless of dose (same used as in Table 9.5). The nonirradiated controls began to mold in 5 days and reached 100 per cent molded berries in 24 days. Clarke[30] comments that "Future research should disclose whether the mold spores which can develop at low temperatures have been killed or whether the remaining viable spores cannot recover from some metabolic damage induced by the radiation treatment."

Other Fruits

Vidal[31] reported that studies in France showed that doses of 150,000 to 200,000 rads followed by storage increased the storage life by a factor of 2 to 3 for a number of fruits. Green tomatoes which were given doses of 500,000 rad followed by storage at 4°C had a marked increase in storage life as compared to controls. The stored tomatoes were given a heat-treatment for ripening before use.

Black currants are not an important fruit crop in this country but are grown extensively abroad. The irradiated fruit has been studied in England[30] and Denmark.[32] Some of the observations of these studies are mentioned here because they differ in some respects from those made with other fruits. Most fresh fruits and vegetables suffer a lose in ascorbic acid content as a result of low doses of radiation but black currants given doses up to 600,000 rads suffered no loss. Also, most fruits (strawberries in particular) are bleached by irradiation but the juice extracted from irradiated black currants had more color than the juice from the controls.[30,32]

Unexpected results were also obtained with irradiated pears. With increasing doses up to 550,000 rads the texture of the fruit was less firm than that of the controls, the loss in firmness increasing with dose. This would be expected from studies made on other fruits. However, after storage at 20°C for 13 days, the relation was reversed, the irradiated fruit being more firm and firmness increasing with dosage. Although all fruits became more soft with continued storage the fruits receiving the highest dose (550,000 rads) were still the most firm after 40 days. Doses above 400,000 rads resulted in the development of a purple pigment in the irradiated pears[30] after 24 days storage at 20°C.

Grapes receiving a dose of 200,000 rads have a slightly modified flavor and a loss in firmness but the changes are not considered to be sufficient to affect acceptability.[33] Irradiation of grape juice has been used in Yugoslavia to control the growth of undesirable fungi. The dose required varies with the organism and is over 600,000 rads for *Botrytis cinera*. It is possible to age wines with low doses producing a madeira type product. Also a poor quality wine can be changed to a vermouth type by irradiation.[33]

The storage life of oranges, lemons, and certain varieties of peaches can be extended appreciably at both ambient and refrigerator temperatures. Softening of bananas can be delayed

by using doses less than 50,000 rads, but some damage to the skin results from this treatment.[30]

Fruits and vegetables, in general, are less sensitive to irradiation than animal products. However, there is a threshold level above which detrimental changes occur in texture, color, and appearance, thus reducing the quality and acceptability. Radiopasteurizing dosages, on the other hand, could be well used to extend refrigerated shelf life.[34]

IRRADIATION OF CEREAL PRODUCTS

At Kansas State College, Lloyd et al.[35] reported studies on the effect of irradiation on the biological, biochemical, and technological properties of wheat; they found that irradiating gluten, either dry or wet, reduces its viscosity. In another study at the same laboratory Yen et al.[36] showed that irradiating damp or dry whole grain with doses between 125 and 625 krep will eliminate fungal respiration without changing the protein solubility of grain extracts. However, doses in excess of 625 krep produced undesirable effects, especially after storage.

In a third study at Kansas State College, Milner and Yen[37] reported the effect of ionizing radiation on the bread-making and related properties of wheat.

Milner used a hard red winter wheat (Concho variety) and doses of 0, 0.125, 0.250, 0.50, and 1.00 Mrep of gamma radiation from MTR fuel elements. The irradiated wheat was conditioned and milled to an average flour extraction of 60 per cent. The biological and biochemical properties of the irradiated wheat and the biochemical properties of flour milled from irradiated wheat are given in Table 9.6 and 9.7 respectively.

Table 9.6—PROPERTIES OF IRRADIATED WHEAT[37]

Dosage, Mrep	Germination, %	Fat acidity, mg KOH/100 g	Fluorescence, scale units	Transmittancy, %
0	95.0	14.0	20.3	17.2
0.125	34.0	14.5	20.1	17.0
0.250	5.0	14.5	20.3	16.8
0.50	4.0	13.5	20.6	16.1
1.00	1.0	14.0	20.9	16.6

Table 9.7—PROPERTIES OF FLOUR MILLED FROM IRRADIATED WHEAT[37]

Dosage, Mrep	Moisture, %	Protein, %	Ash, %	Sedimentation value, ml	Maltose value, malt/log	Maximum amylograph viscosity, B. U.
0	12.9	13.8	0.39	41.1	179.0	960
0.125	13.2	13.6	0.37	37.4	185.5	520
0.250	13.5	13.8	0.37	34.9	207.6	330
0.50	13.7	13.9	0.37	29.5	222.3	180
1.00	13.8	13.8	0.35	21.8	249.3	90

In reference to Table 9.6 the only property of the wheat significantly affected by irradiation is germination. Even though 34 per cent of the wheat germinated after a 0.125-Mrep dose, it is doubtful that the sprouts would live or develop into undamaged plants.

The data in Table 9.7 give considerably more information. The consistent decrease in sedimentation value with increase in dose indicates a corresponding decrease in gluten imbibitional capacity. The increase in maltose value shows a corresponding increase in autolytic sugar production. The drastic decrease in maximum amylograph viscosity indicates a similar decrease in starch gelatinization viscosity. This damage to the starch observed by Milner may explain the drier texture and increased need for liquid noted in other studies with highly irradiated cake flour.[8,38]

Doughs mixed with irradiated flour required shorter development time and had a tendency toward accelerated breakdown with overmixing. As the radiation dose was increased Milner observed that the doughs became less extensible and increasingly stiff, with a loss in characteristic gluten strength.

In spite of the changes noted in the flour and dough, the quality of the bread made from flour given 0.125 Mrep-dose showed an apparent improvement in all loaf qualities as compared to the controls. However, dosages higher than 0.250 Mrep decreased crumbling and reduced loaf volume. A summary of these data[37] is given in Table 9.8. In studies at Michigan[8,38] bread, cake and all-purpose flours were each given a range of doses of gamma radiation; then bread, biscuits, and cakes were prepared from the irradiated flours and taste-panel tests were made.

Table 9.8—QUALITY CHARACTERISTICS OF BREAD MADE FROM IRRADIATED WHEAT[37]

Dosage, Mrep	Loaf volume, ml* (at various $KBrO_3$ levels, %)			Loaf score† (at various $KBrO_3$ levels, %)		
	0.000	0.001	0.003	0.000	0.001	0.003
0	2633	2962	3000+	82	92	94
0.125	2745	2970	3000+	83	93	95
0.250	2702	2972	3000+	79	89	88
0.500	2745	2907	3000+	78	84	85
1.000	2490	2890	2938	66	73	73

* Replicate loaves.

† Maximum score equals 100.

Bread loaves made with 20-krep irradiated bread flour were equal to those made with the nonirradiated flour in all respects except in total volume; the irradiated products were slightly smaller. Bread flour given a dose higher than 50 krep was considered to be undesirably altered. Cakes made with irradiated flour were of progressively poorer quality with increasing doses of gamma radiation. Figure 9.12 shows photographs of cut sections of cakes made in these experiments.[8,38]

Biscuits made with irradiated flour (20 to 150 krep) required more fluid to obtain a workable dough. This dryness of irradiated flour was also noticed when mixing cakes. Biscuits made with irradiated flour had a satisfactory appearance but the quality and flavor of those given higher radiation dosages were not equal to those of the controls.

In summarizing these tests, cake flour, all-purpose flour, and bread flour were considered to be unchanged when given a dose of 25-krep gamma radiation or less. Cakes made with these different flours were similar to the controls in all respects.[8,38]

In 1959 additional studies were reported by Jackson et al.,[39] on the baking qualities of irradiated flour used for bread. An increase in density of the loaves was reported of 0.42 to 7.13 per cent as the dosage was increased from 10,000 to 75,000 rads.

RADIOPASTEURIZATION OF MILK AND MILK PRODUCTS

Thermal pasteurization of raw cow's milk has been practiced for many years to control diseases transmitted to human beings by microorganisms. Many investigators have studied the effects of ionizing radiation on milk with the hope of producing a sterile product of high quality. However, the flavor of milk has been so sensitive to radiation that the studies have shown only limited promise.

In early tests at the University of Michigan[40] about 100 samples of raw and pasteurized milk were irradiated. The results showed that milk may be sterilized by irradiation and stored at room temperature (70°F) for a number of weeks without spoilage from microorganisms. Observations on storage of irradiated milk are listed in Table 9.9.

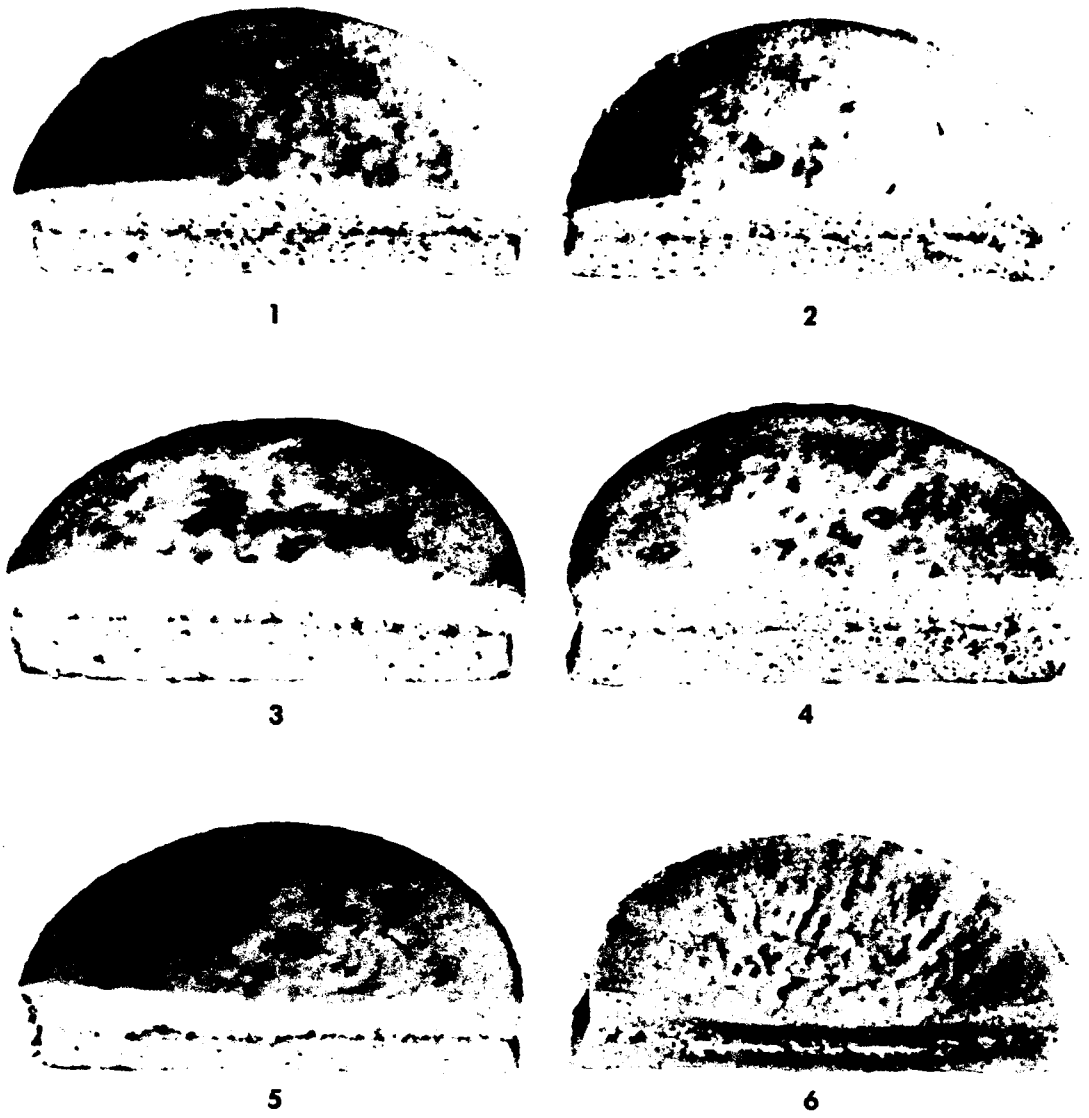


Fig. 9.12—Cakes made with irradiated bread flour; control (O rep), 0.10, 0.15, 0.20, 0.50, and 1.00 Mrep, respectively.[8,38]

Table 9.9—OBSERVATIONS ON IRRADIATED MILK STORED AT ROOM TEMPERATURE[40]

Type	Container	Temp., °F	Dosage, Mrep	Time to curdle	Remarks
Pasteurized	Test tubes	77	0	48 hr	Large white curds
			2.04		Not curdled after 6 weeks
Pasteurized	Polyethylene bags	77	0	72 hr	Large white curds
			2.04	6 weeks	Brown discoloration, small curds
Pasteurized	Polyethylene bags	77	0	24 hr	Large white curds
			1.44	8 weeks	Brown discoloration, small curds
Raw	Polyethylene bags	77	0	24 hr	Large white curds
			2.04	3 weeks	Red growth on milk (blood colored)
Pasteurized	Evacuated test tubes	70 ± 10	0	48 hr	White curds
			2.04		Opened after 8 weeks; no spoilage evident

Taste tests reported in these studies on irradiated pasteurized milk showed that most individuals could detect an off-flavor in milk which has received a dosage of only 80 krep of radiation. The off-flavor was described as a burnt or sharp taste or as a "chalky taste."

In subsequent tests attempts to control the flavor change in irradiated milk and milk products by means of various chemical additives were not successful. Skim milk given an irradiation dose of 1.9 Mrep had a goaty odor and a flavor somewhat similar to that of irradiated fresh meats. Irradiated cream smelled rancid and tasted like oxidized fat. Fresh creamery butter gave the same result. Irradiated cottage cheese (1.9 Mrep) had such poor flavor and odor that the product could not be considered edible.[41]

Freeman[102] discussed the effect of radiopasteurizing doses of gamma radiation on the bacterial flora of liquid milk. Total counts and coliform counts were markedly reduced at various dose levels. Spore formers which survived were found to increase in about 4 to 11 days after irradiation.

In 1959 Glew[103] reported on the effects of radiation on liquid whole milk and whey protein. Flavor changes, alkaline phosphatase enzyme activity, and acid soluble nitrogen were determined for gamma irradiated whole milk. A dose of only 25,000 rads produced detectable flavor changes but 25 Mrads was required to inactivate the enzyme. By combining irradiation with heat treatment lower radiation doses can be used for enzyme inactivation. He commented that "The results presented here do not, as yet, indicate any clear applications for the use of radiation in preserving milk."

Bierman et al.[42] report that the threshold doses producing off-flavor in cream and various types of whole and skim milk ranged from as low as 7 to 25 krep (cathode rays). High-temperature short-time prepasteurization of milk did not significantly alter susceptibility to radiation-induced off-flavors. However, homogenization of pasteurized whole milk was beneficial. The off-flavors induced by radiation increase with moisture content and fat; they decrease as total solids increase. In cream both the total solids and fat content are increased and effects of these tend to compensate each other.

The susceptibility of milk and milk products to flavor change as a result of irradiation has been reported by other investigators.[43-45,51] Huber et al.,[45] reported the elimination of off-flavors in flavored milk beverages by stripping with inert gases. However, this technique adds to the cost of processing and must be performed carefully to prevent recontamination with microorganisms. Also raw milk must be thermally pasteurized to inactivate the enzyme phosphatase.

In 1959 Proctor[2] reviewed the problem of the high susceptibility of milk to flavor change on irradiation. He also described the procedure of using low-temperature vacuum distillation

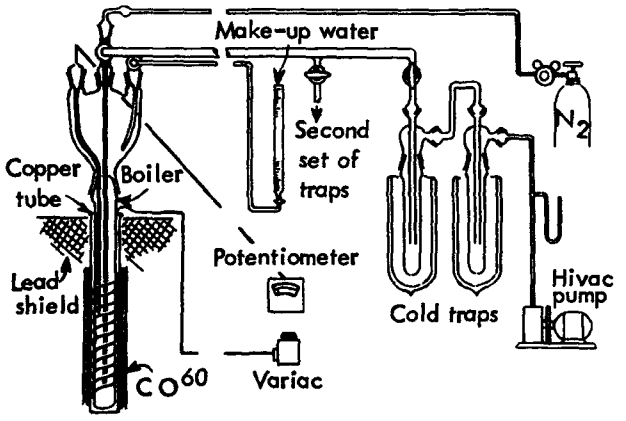


Fig. 9.13—Radiation distillation apparatus for milk.[2,142]

equipment concurrently during irradiation to strip the off-flavor components from irradiated milk (see Fig. 9.13). The sterile milk obtained by this process is claimed to be free of off-flavor and to possess a bland taste.[2]

RADIOPASTERUIZATION OF EGG PRODUCTS

Brooks, Hannan, and Hobbs of Cambridge (England) have radiopasteurized egg products and have reported that a dose of 0.25 Mrad of electrons will destroy (reduce by a factor of 10^7) *Salmonella, S. paratyphi* and *S. typhicurium.*[104,105] A dose of only 0.1 Mrad is required for *S. seftenberg.* A dose of 0.3 Mrad of gamma radiation also produced whole egg magma free of these organisms. When this product was used to prepare scrambled eggs an off-flavor could be detected. Spray drying of the radiopasteurized egg magma minimized the difference between the irradiated product and control. No off-flavor was observed in sponge cakes made from the irradiated frozen eggs.

Radiopasteurizing of egg products is considered quite promising as a commercial process, particularly for frozen products because it is not necessary to thaw the products.

RADIOPASTEURIZATION OF MEAT, POULTRY, AND SEAFOOD

Most meats are susceptible to flavor changes induced by irradiation but the off-flavors in many cases are not severe. Although radiation doses as low as 20 krep can be detected in milk and dairy products, this is not the case with meat. Extensive studies on irradiated meats have been conducted in many laboratories with varying degrees of success.

TASTE PANEL TESTS ON PORK

To explore the feasibility of radiopasteurization, microbiological and taste panel studies were conducted at the University of Michigan.[10,106] In the taste panel tests, studies were made both with fresh ground pork and fresh ground beef. Ground meat was used as it has a shorter refrigerator shelf life than standard meat cuts. One experiment, a taste panel test with ground pork, will be briefly described here.

In the first taste panel tests with fresh pork relatively lean loin was obtained, put through a conventional meat grinder twice, and divided into four portions. One portion was stored in closed glass containers in a refrigerator at 40°F. The other three portions were placed in closed glass containers and irradiated in the radiation cave at approximately 45°F with dosages of 30, 60, and 100 krep, respectively, after which they were placed in a refrigerator with the controls.

The ground pork was made into patties and fried slowly in a neutral hydrogenated vegetable shortening until well done. Each sample was fried separately and care was taken to use the same dosage of cooking in each case. The observation was made that the irradiated samples had a slightly different odor which was most noticeable when the glass containers were first opened. However, with the low radiation dosages used, this odor was not objectionable and disappeared on cooking.

The raw irradiated meat developed a slight color change by losing some of its redness immediately after irradiation. In addition, the outer surfaces of the raw ground pork developed a grayish tinge. The cooked meat had no off-color or off-flavor.

Table 9.10 shows the statistical analysis of the first pork-storage experiment; no significant differences in flavor preference was detected by the panel until the 5th day of storage, at which point, the control was definitely inferior to the irradiated food, especially to that given the higher doses.

Fresh control samples were substitued for the remaining taste tests. After 8 days of storage, there appeared to be no significant difference in preference between the fresh control and the pork given the two highest doses, but the 30-krep sample appeared to have developed an undesirable flavor. This experiment was stopped after the 8th day since the irradiated samples of meat had been consumed. However, these data indicate that ground pork irradiated at levels between 60 and 100 krep will keep for at least 8 days at 40°F.

As a result of further tests over a longer time, it was concluded that irradiated fresh ground pork can be expected to have a refrigerator storage life of around 2 weeks using a radiation dose between 60 and 80 krep. Although no tests were made with standard cuts of pork, it is expected that the refrigerator shelf life would be as long or longer.[10]

Table 9.10—RANKING OF TASTE OF IRRADIATED PORK BY MEANS OF PAIRED COMPARISONS[10]

Test No.	Storage time, days	Dose, krep, and total scores for each dose* 0	30	60	100	P	Significant difference
1	1	26	30	24	28	0.37	No
2	1	26	26	29	27	0.87	No
3	3	22	23	20	25	0.49	No
4	3	25	18	23	24	0.12	No
5	5	36	29	21	22	<0.01	
		0.00	0.04	0.55	0.41		Yes
6	5†	30	30	25	23	0.11	No
		23		17	14	0.01	Yes
		0.04		0.27	0.69		
7	8	21	28	19	22	0.02	Yes
		0.28	0.05	0.45	0.22		
			20	12	13	0.02	Yes
			0.00	0.60	0.40		
		14		14	17	0.61	No
8	8	20	19	15	18	0.40	No

*Where significant differences exist, estimates of the preference ratings are displayed below the total scores. Sum of total scores is either 90 or 108 as the number of judges varied from 5 to 6.

† At this point the control food was definitely spoiled; hence, frozen control samples were used for the remaining sittings.

Table 9.11—RANKING OF IRRADIATED BEEF BY MEANS OF PAIRED COMPARISONS[8,10]

Test No.	Storage at 40°F, days	Dose, krep 0	50	80	110 Score*	P	Significant difference
1	1	22	20	25	23	0.49	No
2	1	26	29	28	25	0.70	No
3	4	22	22	22	24	0.94	No
4	4	23	20	25	22	0.49	No
5†	7	29	24	28	27	0.58	No
6	7	27	25	25	31	0.29	No
7	11	18	28	26	18	0.0004	Yes
		0.47	0.03	0.02	0.47		
8	11	20	34	29	25	0.0002	Yes
		0.71	0.02	0.08	0.19		
9	14	19	23	33	33	<0.0001	Yes
		0.83	0.17				
10	14	22	23	31	32	0.002	Yes
		0.49	0.39	0.07	0.05		

*Where significant differences exist estimates of the preference ratings are displayed below the total scores.

† Frozen control was substituted at this point because fresh control had spoiled.

TASTE PANEL TESTS ON BEEF

Similar tests were conducted with ground beef using the technique of ranking by means of paired comparisons; results of one series of tests[8,106] are given in Table 9.11.

After 4 days of storage the control samples stored at 40°F were replaced with control samples stored in the frozen state because the fresh control had spoiled.

The data given in Table 9.11 indicate that after 7 days of storage at 40°F there was no significant difference in the rankings but after 11 days storage the beef given 50 and 80 krep had developed decided off-flavors.

The panel showed a decided preference for the nonirradiated frozen ground beef after storage for 14 days, but the irradiated beef given 50 krep was reported to have a good flavor and was definitely preferred to the beef given 80 and 110 krep. The beef given the highest dosage of irradiation (110 krep) tasted rancid to most of the panel members.[8] Although these tests were not considered conclusive a storage life of 10 days at 40°F appears reasonable for both raw ground lean pork and raw ground beef given a 50- to 80-krep dose.

SEAFOODS

Fish, shell fish, and white meats such as chicken appear to be less susceptible than beef to off-flavors and odors produced by radiation. For example, fresh iced crabmeat was irradiated, stored for 1 week under refrigeration, then inspected for flavor and bacteria count at the Crisfield Company.[8] The results listed in Table 9.12 indicate that a dose of 120 krep gave an "excellent" flavor.

Table 9.12—RESULTS OF TESTS ON IRRADIATED FRESH CRABMEAT AFTER ONE-WEEK STORAGE[8]

Dose, krep	Condition	Odor	Flavor	Bacteria count, organism/g
0*	Spoiled	Off	No taste test	3×10^6
40	Slightly spoiled	Slightly off	No taste test	3×10^6
80	Very slightly spoiled	Slightly off	Slightly off	2×10^6
120	Excellent	Excellent	Excellent	0.78×10^6
1000	Dry cooked appearance	Musty	Musty	3×10^3

*Control

In 1959 Shewan[107] reported on studies made in Scotland on radiopasteurized fish. A large number of types of fish were irradiated and checked for quality to determine the most suitable type for treatment. The maximum permissible doses that can be given to various seafoods[107] are given in Table 9.13.

Over 20 species of fish were tested and of these, cod and halibut were superior from organoleptic considerations. Fish with a high fat content such as herring tend to become rancid. Pigmented fish such as salmon are bleached and some white fleshed fish develop brown discolorations. Cooked crab, shrimp, and blanched oysters tolerated high doses of radiation and keeping qualities were greatly improved.

In some of the tests radiopasteurizing doses of 0.25 to 0.85 Mrads were given to a variety of fish both raw and with various heat treatment and different methods of packaging. The extension in shelf life as compared to untreated controls was determined and the results are given in Table 9.14. Shewan[107] comments "Here in main feature is the remarkable extension of shelf life obtained when the product is held at chill temperatures.... Provided the more obvious defects in the present process can be overcome, it seems likely that this new technique could revolutionize the present methods of marketing unfrozen fish to inland populations far from ports."

Table 9.13—THE MAXIMUM PERMISSIBLE IRRADIATION DOSES THAT CAN BE GIVEN TO VARIOUS FISH AND FISHERY PRODUCTS (FROM VARIOUS SOURCES)[107]

Species of fish	Irradiated dose,* Mrad	Species of fish	Irradiated dose,* Mrad
Butterfish	0.50–0.70	Salmon	0.5
Butterfish (blanched 140°F for 5 min)	1.5	Tuna	1.0
Cod	0.8–1.5	Whiting	0.20–0.25
Cod (blanched 140°F for 5 min)	1.5	Whiting (blanched 140°F for 5 min)	0.80
Haddock	0.60	Ocean perch	0.80
Halibut	1.0–2.0	Kippered herring	1.0
Lemon sole	0.5–1.0	Kippered sable fish	1.0
Ling	0.5–1.0	Kippered sturgeon	1.0
Mackerel	1.0	Crab meat	1.0
Pollack	0.5–0.70	Shrimp	1.0
Pollack (blanched 140°F for 5 min)	0.80	Pacific oysters (blanched)	5.0

* Doses in excess of those given above produce undesirable organoleptic changes, e.g., in flavor and odor and/or in appearance, e.g., bleaching of pigment in salmon.

Table 9.14—EXTENSION OF SHELF LIFE OF FISH AND FISHERY PRODUCTS (FROM VARIOUS SOURCES)[107]

Species	Irradiation treatment	Extension of shelf life in days over the untreated controls
Cod, pollack, butterfish	Blanched, vacuum packed in tins, at 0.5 Mrad	20 days stored at 1.5°C
Pollack and butterfish	Raw, vacuum packed in tins at 0.25 Mrad	10–20 days stored at 1.5°C
Cod	Raw, vacuum packed in tins at 0.25 Mrad	About 70 days stored at 1.5°C
Cod	As above at 0.25 Mrad	About 40 days stored at 0°C
	As above at 0.50 Mrad	About 40–50 days stored at 5°C
	As above at 0.75 Mrad	About 50–100 days stored at 0°C
Pacific cod fillets	Raw, in polythene, at 0.20 Mrad	About 40 days stored at 1.5°C
Haddock fillets	Raw, in cellophane, at 0.6 Mrad	About 30 days stored at 1 to 4°C
Pacific cod fillets	Raw, at 0.25 Mrad	About 40 days stored at 0°C
	Raw, at 0.75 Mrad	About 100 days stored at 0°C
Cod fillets	Raw, in polythene at 0.25 Mrad	About 20 days stored at 0°C
Crab meat	Raw, at 0.50 Mrad	About 50 days stored at 0°C
Crab meat	Cooked, at 0.40 to 0.85 Mrad	About 40 days stored at 0°C

MICROBIOLOGICAL TESTS ON BEEF

In the microbiological tests conducted at Michigan[10] fresh lean ground beef was inoculated with a suspension of a psychrophilic gram-positive bacterium originally isolated from spoiled ground beef. This organism grew readily at refrigerator temperature (40°F). Noninoculated control samples were kept to establish the meat's normal population of microorganisms. Samples of the ground meat were given various dosages of gamma radiation after inoculation. Irradiation and subsequent storage was carried out at 4°C. Immediately after irradiation and at frequent periods during storage, plate counts were made. Table 9.15 shows the aerobic microbial counts for the meat samples immediately after irradiation. No significant differences in counts are apparent[10] up to a dosage of 40 krep.

Table 9.15—EFFECT OF GAMMA RADIATION[10] ON AEROBIC MICROBIAL FLORA OF GROUND BEEF INOCULATED WITH 32,000 PSYCHROPHILIC BACTERIA PER GRAM

Dose, krep	Count, thousands of microorganisms per gram	Survivors, %
0 (control)	80	100.0
20	140	100.0
40	68	85.0
80	7.5	9.0
160	9.5	12.0

Figure 9.14 shows counts per gram of meat after subsequent storage at 4°C. Data are typical of population growths. With a highly contaminated sample, the population increases rapidly as a power function until a saturation point is reached. In the data shown the population is increasing by a factor of 10 about every 30 hr. There is no direct correlation between the bacterial count and spoilage of food. However, soon after the count begins to rise, off-flavors and off-odors are usually produced by the metabolic activity of the organisms and the food is considered spoiled. There are many exceptions, such as cheese, pickles, and fermented beverages, in which the flavor and odor changes and chemical products produced by the microorganisms are desirable. Beginning of spoilage might be expected within 24 hr.

No detectable differences in either color or odor were noted in the samples immediately after irradiation. After 5 days of storage, both the inoculated and noninoculated controls had a pronounced putrid odor. Initiation of spoilage in the sample receiving a radiation dose of 20 krep was indicated by a slight off-odor, whereas none of the other irradiated samples had an off-odor. After 13 days of storage, all the irradiated samples except that which received a dose of only 20 krep were still free of off-odors and there was no indication of spoilage.

A study of the microbiology of fresh beef stored at 2°C under aerobic conditions and high humidity was conducted at the American Meat Institute Foundation.[108] The results of this study indicate that the lethal dose of gamma radiation for two representative cultures, *P. geniculata* and an unidentified species, is approximately 30 krep.

MICROBIOLOGICAL TESTS ON FISH

Shewan[107] reported microbiological studies on irradiated fish. Radiopasteurization greatly reduced the total count of organisms as shown in Table 9.16.

In addition to reducing the total count radiopasteurization had a marked selective action on the types of organisms remaining. The *Pseudomonas* group of organisms are very active in fish spoilage and were almost completely eliminated leaving the *Achromobacter* group in predominance. During storage at 0°C the *Pseudomonas* group gradually recovered and multiplied. After three weeks they predominated again and constituted about 90 per cent of the population of microorganisms in the stored irradiated fish. Shewan[107] states "It seems safe to conclude

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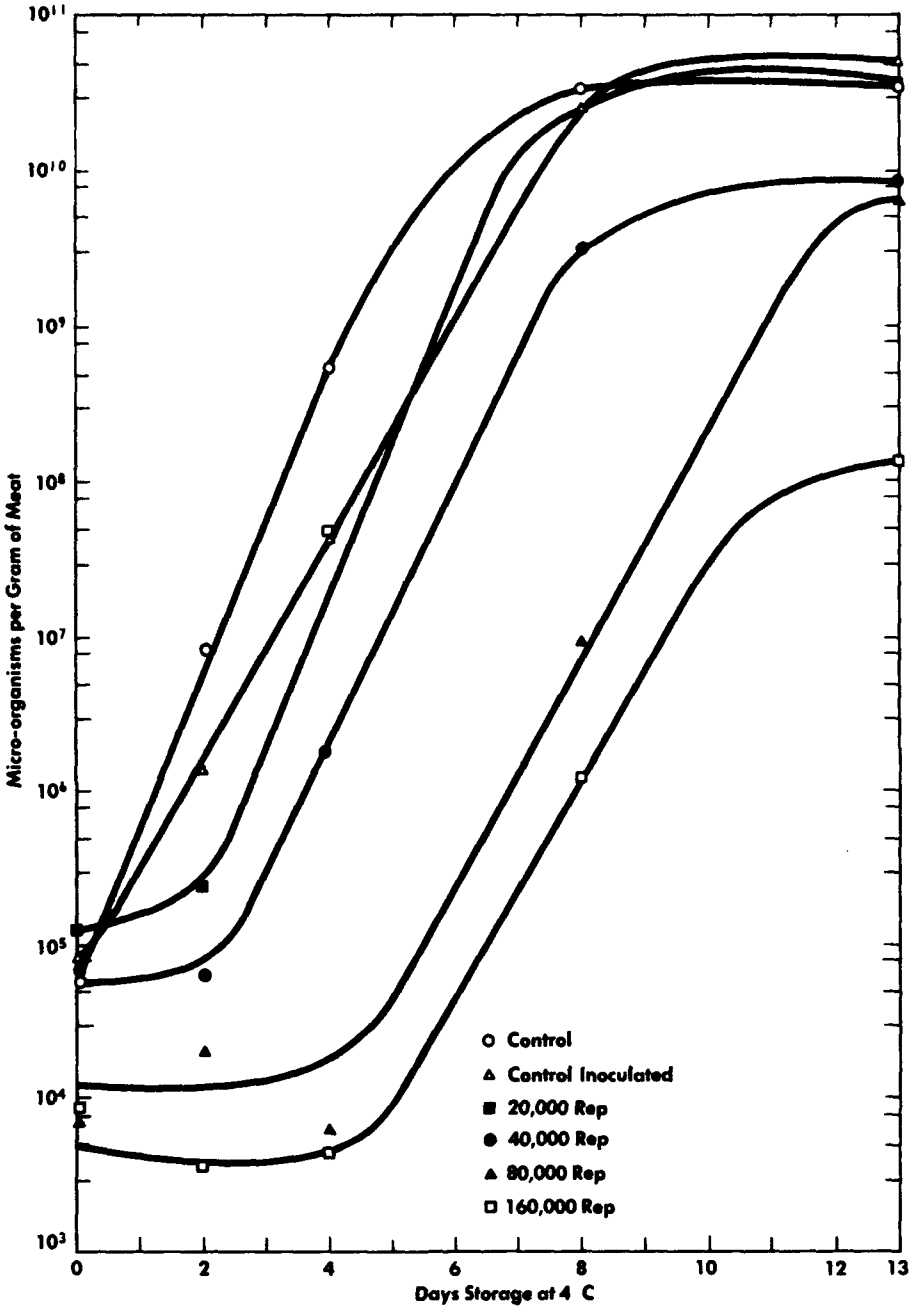


Fig. 9.14—Semilogarithmic plot of effect of gamma radiation on aerobic microbial flora of ground beef.[10]

Table 9.16—REDUCTION IN THE BACTERIAL LOAD IN FISH MUSCLE IMMEDIATELY AFTER IRRADIATION*[107]

Species of fish	Unirradiated controls	Irradiation doses, Mrad						
		0.1	0.2	0.25	0.4	0.5	0.6	1.0
Haddock fillets	5.5				2.5		1.75	
Cod fillets	6.5	4.0	2.0					
Cod fillets	6.3			2.7		2.0		†

* Expressed as log number per gram of muscle.
† Commercially sterile.

that this enhanced storage life must be related to the elimination or suppression of the active spoilage types belonging to the *Pseudomonas* group." Table 9.17 gives data on the reduction of various groups of organisms in cod fish receiving various treatments and stored at 0°C for various lengths of time.

Table 9.17 also includes data on codfish receiving a combination of radiopasteurization and a treatment with the antibiotic aureomycin. Aureomycin is effective in reducing the count of the *Pseudomonas* group and the combination of processes virtually eliminates all organisms except yeasts. Fish treated with the combination of processes and stored for 3 weeks were considered to be of good quality organolystically.[107]



Fig. 9.15—Inspection of irradiated foods at the radiation laboratory of the Low Temperature Research Station, Cambridge, England. (Courtesy of R. S. Hannan)

MICROBIOLOGICAL TESTS ON POULTRY

Hannan and colleagues at the Low Temperature Research Station, Cambridge, England, have carried out extensive studies on radiopasteurized fresh chicken.[109,110] Figure 9.15 shows

Table 9.17 — QUALITATIVE ANALYSIS[107] OF THE BACTERIAL FLORA OF IRRADIATED FISH DURING STORAGE AT 0°C

	Normal untreated fish			0.5-Mrad-treated fish			0.25-Mrad-treated fish			Aureomycin dipped fish				Aureomycin dip and 0.25-Mrad-treated fish		
Bacterial flora from plates incubated at 20 and 0°C	1 day after filleting	16 days at 0°C	23 days at 0°C	1 day after treatment	13 days at 0°C	21 days at 0°C	1 day after treatment	13 days at 0°C	21 days at 0°C	1 day after dipping	9 days at 0°C	16 days at 0°C	23 days at 0°C	6 days after treatment	13 days at 0°C	20 days at 0°C
Total organisms examined	100	200	200	40	100	100	35	100	100	75	100	100	100	50	90	100
Pseudomonas, %	40	55	72	5	52	100	3	7	99	32	5	7	7	2	89	97
Achromobacter, %	30	40	13.5	40	47		63	93		35	16	9	6	2	11	
Flavobacter, %	11	1		2.5			8			5	11	4	1			
Corynebacterium, %	6	2	1	5.0	1		6		1	12	18	3	8			
Micrococcus, %	1			7.5			20			5	38	15	2	2		
Yeasts, %				2.5						1	2	55	67	92		1
Miscellaneous, %	12	2	13.5	37.5						10	10	7	9	2		2

a view of the Radiation Laboratory at the Low Temperature Research Station with samples of irradiated foods being inspected by Hannan (right) and Shepperd (left). Figure 9.16 gives microbiological data obtained at that laboratory on irradiated raw minced chicken meat.

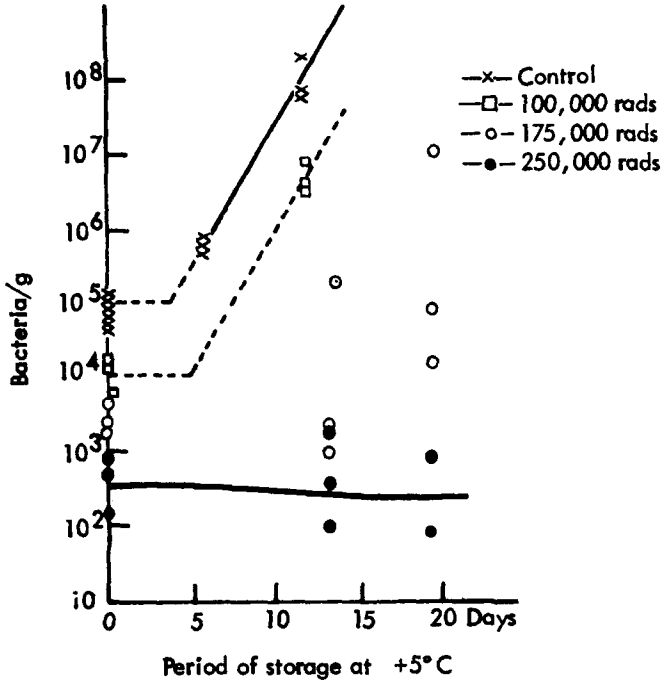


Fig. 9.16—The effect of storage at 5°C on the viable count of control and irradiated samples of minced chicken meat.[110]

Lags in the rise of bacterial populations as a result of low dosages of irradiation are similar to those for beef. The noteworthy difference is that for 0.25 Mrad, there is little or no increase in population over extended periods of time. Hannan reports that although some changes in flavor, odor, and color of the irradiated chicken were detectable at this dosage, the change in flavor of the cooked chicken was, at the most, very slight.[109]

Ingram and Thornley[110] reported that the flora on control eviscerated whole chickens consisted almost entirely of a mixture of nonpigmented *Pseudomonas* strains, *Achromobacter* and a mixture of coliform bacteria. After radiopasteurization the population consisted almost entirely of *Achromobacter*. The ultimate spoilage flora in minced chicken breast given a dose of 0.25 Mrad and stored at 5°C were found to be microbacteria and faecal streptococii. The presence of the latter gives some doubt as to the safety of radiopasteurizing chicken in the minced form.

Eviscerated whole chickens were given a dose of 0.8 Mrad and stored in loose film bags at 1 to 3°C. In this case the ultimate spoilage organisms were a mixture of bacteria of types commonly found on chilled meats. Storage life was increased from about 10 days for controls to about 50 days for radiopasteurized fowl. It was concluded that this process was safe providing the chickens were kept free to an atmosphere of air and at chill temperatures.[110]

ODOR AND FLAVOR PROBLEMS OF HIGH-DOSAGE IRRADIATION OF MEAT

FRESH MEAT

Observations at the Fission Products Laboratory, the University of Michigan, indicated that irradiated raw beef given no special pretreatment (such as cooking, freezing, or irradiating in evacuated cans) posessed a detectable flavor change if the radiation dose exceeded 0.10 Mrep. However, cooked beef could be given between 1.0 and 1.2 Mrep without affecting

texture or flavor. At dosages higher than 1.2 Mrep the lean meat became softer and stringier, whereas the fat, although still white, was almost completely liquefied. The fat in the cans of control beef was distributed throughout the can.[106]

Huber et al.[45] reported that holding raw beef frozen at subzero temperatures before irradiation, followed by storage at room temperature after irradiation, gave a product ranking almost as high as the control (see Table 9.18).

Table 9.18—ELECTRON IRRADIATED (1.5 Mrep) RAW BEEF[45]

Prestorage		Evaluation		
Time, hr	Temp., °C	Odor	Flavor	Color
24	3	2.5	2.8	1.5
	−15	3.0	3.5	1.5
72	3	3.0	2.8	2.8
	−15	4.8	4.5	4.5
	−5	5.0	4.0	4.5
	−180	4.5	4.5	4.5
96	3	3.8	3.0	3.5
	−15	4.8	4.3	4.5

* Average of taste panel of five members. Controls rated as five.

CANNED MEATS

Kirn et al.,[47] of Swift & Company reported extensive data on a wide variety of meats given either 1.45- or 2.00-Mrep radiation doses using the Capacitron and stored for as long as 1800 days at 7°C. These authors state that in general irradiated meats tend to recover their color and flavor on holding under oxygen-free conditions. This recovery occurred in 10 to 30 days; additional storage did not further improve the quality. All beef products (raw, cooked, cured, or salted) developed a bitter flavor in addition to burnt-like flavors usually attributed to irradiation. However, chicken, lamb, pork, and bacon were not bitter and were considered by the taste panel to be the products with the best flavor. A summary of the data is given in Table 9.19.

In the scores given in this table, ratings of 9 and 10 represent excellent, 7 and 8 desirable, 5 and 6 acceptable, 3 and 4 poor, 1 and 2 very poor or repulsive.[47]

The texture of the meats was good except for beef liver, which showed extensive shrinkage. Red pigments of all canned raw meats showed initial radiation damage but all products recovered except beef liver. Cooked beef, originally brown in color, turned bright pink. It was concluded however, that meats given some heat processing prior to irradiation were most satisfactory under long storage.

Experiments on storage of irradiated canned foods at higher temperatures were conducted by Pratt and Ecklund at the American Can Company.[48] Meats were irradiation-sterilized and stored at 40, 70, and 98°F and examined at periods up to 3 years. Vegetables were irradiated with sterilizing dosages, stored at 40 and 70°F, and examined at periods of up to 1 year. The results are summarized below.[48]

Ground Beef

An attractive raw red color survived irradiation, but a noticeable off-flavor and odor developed. This was somewhat reduced by irradiation in the frozen state, but was unaffected by pasteurization, addition of sodium ascorbate, or frying. Some liquid formation occured after 9 months storage at 70 or 98°F and an additional "liver-flavor" developed, but these were both reduced by heat pasteurization to 160°F after irradiation. Hard white particles of tyrosine also separated on the top of the loaf after this length of storage. The development of these particles was delayed by adding sodium ascorbate. The ground beef was not readily made into patties for frying after 5 months at 98°F and the attractive color had faded somewhat after 1 year at this temperature. After 3 years at this temperature all the cans had produced gas.

Table 9.19—RESULTS OF TESTS FOR ODOR AND FLAVOR ON IRRADIATED MEATS STORED AT REFRIGERATOR TEMPERATURES[47]

Test No.	Product*	Appearance	Aroma	Flavor	Taste produced by irradiation
1	Beef steaks with salt				
	Control	7.5	8.5	9.3	Bitter,
	+ 1 day	2	7.5	5.0	metallic
	+ 1800 days	8	2	2.75	
2	Beef steaks without salt				
	Control	7.5	8.5	9.0	Bitter,
	+ 1 day	1.8	7.5	7.5	metallic
	+ 1800 days	5	2	2.5	
3	Hamburger, raw				
	Control	8.5	9.0	9.0	
	+ 1 day	3.3	2.5	4.0	
	+ 1800 days			3.5	
4	Hamburger, broiled				
	Control		4.0	5.2	Extremely
	+ 1 day		3.6	3.2	bitter
	+ 30 days		4.0	5.0	
	+ 1800 days	7.0	5.0	3.5	
5	Hamburger + deoxgenase and glucose				
	Control		7.5	7.3	Bitter
	+ 1 day	2	5.0	4.8	after-taste
	+ 1800 days	5.4		5.4	
6	Hamburger + 0.1% monosodium glutamate				
		9.0	9.2	9.3	Bitter
	+ 1 day	6.8	4.3	2.0	
	+ 1800 days	8.0		2.2	
7	Hamburger + 1% succinic acid				
	Control	9.0	9.2	9.3	
	+ 1 day	9.0	9.2		
	+ 1800 days	6.8		7.0	
8	Hamburger + 0.25% sodium ascorbate and citric acid				
	Control	8.5	9.0	9.0	Very re-
	+ 1 day	2.5	3.0	5.0	pulsive
	+ 1800 days	6.4		2.5	
9	Beef, roast				
	Control	8.0	8.8	9.6	Irradiated,
	+ 1 day	8.0	6.0	6.4	scorched
	+ 10 days		7.4	5.8	
	+ 21 days		5.4	4.4	
	+ 60 days		4.8	4.0	
	+ 1800 days	8.0	8.0	4.2	
10	Veal, roast				
	Control		8.0	8.6	Slightly
	+ 2 days		5.4	4.0	irradiated
	+ 11 days		4.4	4.0	
	+ 30 days		5.8	4.4	
	+ 1800 days	8.0	4.5	5.0	
11	Pork, roast				
	Control	8.6	9.4	9.4	Very slightly
	+ 1 day	8.6	4.0	2.6	irradiated
	+ 12 days	8.0	5.4	4.0	
	+ 31 days		7.0	7.6	
	+ 1800 days	8.3	7.0	6.7	

Table 9.19—(Continued)

Test No.	Product*	Appearance	Aroma	Flavor	Taste produced by irradiation
12	Lamb, roast				
	Control		9.0	8.8	Strong
	+ 2 days		6.8	2.6	lamb flavor
	+ 11 days		4.4	4.0	
	+ 30 days		7.0	6.6	
	+ 1800 days	8.0	5.5	5.3	
13	Chicken, roast				
	Control		9.6	9.0	Irradiated
	+ 1 day		5.0	2.0	or scorched
	+ 10 days	3	4.4	5.0	
	+ 1800 days	8	6.0	5.2	
14	Liver, beef				
	Control	8	10	10	Repulsive,
	+ 2 days		8.0	7	bitter
	+ 17 days		8.0	7.4	
	+ 1800 days	6	44	3	
15	Pork sausage, raw				
	Control	6	8.8	6.4	
	+ 2 days	6	8.4	6.4	
	+ 15 days	8	8.0	8.0	
	+ 26 days	8.6	7.0	6.6	
	+ 1800 days	1	6.0	6.5	
16	Pork sausage, cooked				
	Control	9.6	9	8.6	
	+ 1 day	8.6	5.6	4.8	
	+ 10 days		9.0	5.6	
	+ 20 days		8.6	8.0	
	+ 1800 days	5	9.0	6.5	
17	Corned beef hash				
	Control		9.0	9.0	Bitter
	+ 2 days		6.0	6.0	after-taste
	+ 1800 days	7		3.3	
18	Ham				
	Control	10	10	10	Very bitter,
	+ 1 day	2	2	2	metallic,
	+ 13 days		7.0	4.4	irradiated
	+ 1800 days	7		3.8	
19	Bacon				
	+ 26 days	8	5.3	3.6	Slightly
	+ 1800 days	7	6	5.5	irradiated
20	Cured pork shoulder				
	Control	10	8.4	9.0	Bitter
	+ 1 day		2.6	2.0	
	+ 15 days		3.6	4.0	
	+ 36 days		5.0	5.0	
	+ 1800 days	8	6.0	3.7	
21	Franks				
	Control	10	10	10	Oxidized or
	+ 1 day	10	2	2	irradiated
	+ 1800 days	5.3	8.0	4.2	

*The control was not irradiated. The other samples were irradiated and stored for approximately the additional days indicated.

Beef Tenderloin

Observations similar to those for ground beef were made but no tyrosin particles appeared on heat pasteurized samples.

Pork Luncheon Meat

This was heat pasteurized at 160°F after irradiation. Off-flavor and odor were produced and a pinkish liquid fat separated. Brown surface discoloration appeared after 1 month at 98°F or 2 months at 70°F, becoming worse with continued storage.

Cured Ham

The ham showed no change in flavor on storage and the color faded only on extended storage.

MEAT FATS

The development of rancidity in fats may limit the radiation dosage to raw meat.[29,46] Brownell et al.,[46] reported data given in Table 9.20 on limited tests on irradiated pork, beef, bacon, and lard fats. The increase in peroxide value of pork fat stored at 4°C is shown in Fig. 9.17.

The control samples of pork fat stored at 4°C became highly rancid after 22 days. Irradiated samples maintained a characteristic taste of tallow for a much longer period of time before developing rancidity. This probably resulted from a decrease in the original concentration of microorganisms by radiation, which depressed the hydrolytic type of rancidity. Tests of irradiated samples did not show objectionable flavor or taste. Samples tasted like tallow and had a strong tallow odor.[46]

Additional data on the irradiation of fats is given by Hannan[29] and by Mead.[49]

PROGRAM OF THE OFFICE OF THE QUARTERMASTER GENERAL, (U. S. ARMY) ON FOOD IRRADIATION

Extensive programs of research and development in the field of irradiated foods are being conducted by the Office of the Quartermaster General, U. S. Army. Some of these studies—such as those on the wholesomeness of irradiated foods and those on sprout inhibition in irradiated potatoes—are described in this chapter. Also brief descriptions of the Food Irradiation Reactor (FIR) and the High Intensity Food Irradiator (HIFI) which were considered for the Army Ionizing Radiation Center[111,112] are given in Chaps. 4 and 10. Table 9.21 is presented to provide a more complete understanding of the scope of this program. This table lists the Department of Army research contracts on the radiation preservation of food[111] as of May 1956.

One of the critical problems in using ionizing radiation, particularly high dosages, to process foods is the development of "off-flavors" foreign to these foods when prepared by conventional methods. Personnel of the Quartermaster Corps have reported on a survey of 80 foods and state that "quite a few of the irradiated foods show considerable promise."[111] Irradiated (3 Mrep) pork loin was reported to show no noticeable change in appearance and taste after 9 months storage at room temperature. Irradiated sliced bacon with a salt content comparable to the civilian product, rather than the high salt content now used in Army bacon, showed good taste acceptance after 6 months storage at room temperature. The irradiated product in frozen storage remained satisfactory three times as long as the frozen control bacon. Irradiated pork sausage was also reported to have good quality.[111,112]

Studies on fish and fish products were also made; shrimps and oysters in particular showed promise. Raw oysters were given 3-Mrep doses of gamma radiation, were stored for 5 weeks at 70°F, and were found to be very tasty either raw or fried. Such food products with long storage life are not marketed at present.[111,112]

Favorable results were also reported for poultry.[111,112] Both fresh and precooked chicken given 3-Mrep doses of gamma radiation and stored for 4 months at room temperature produced

Table 9.20—EFFECT OF GAMMA RADIATION ON SOME ANIMAL FATS[46]

Fat sample	Dose, Mrep	Peroxide value, milliequivalents O_2/kg of fat	
		Control	Irradiated
Pork			
No. 1	1.83	0	48.54
No. 2	2.07	0	58.75
Beef			
No. 1	1.83	0	26.62
No. 2	1.83	0	31.97
Bacon fat	2.07	0	34.21
Lard	2.07	5.01	15.04

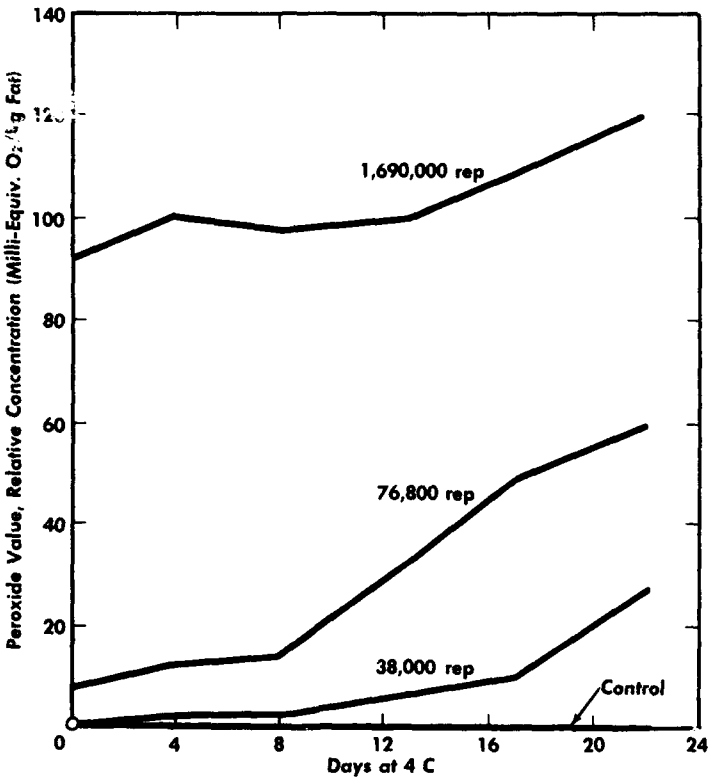


Fig. 9.17—Increase in peroxide value of irradiated pork fat stored at 4°C.[46]

Table 9.21—DEPARTMENT OF ARMY RESEARCH CONTRACTS ON THE RADIATION PRESERVATION OF FOOD[111] AS OF MAY 1956

Organization	City and state	Title
Alabama Polytechnic Institute	Auburn, Ala.	Study of the nutritive and long-range effects of irradiated foods in animals with special reference to the chemical and nutrition changes in vitamin cholines and related nutrients
American Meat Institute Foundation	Chicago, Ill.	Chemistry of color, flavor and odor changes in irradiating meat
		Radiation sensitivity of meat spoilage microorganisms
Armour Research Foundation of Illinois	Chicago, Ill.	Irradiation of lipid protein systems
Columbia University*	New York, N. Y.	Studies on nutritive and biochemical effects of radiation
		Biochemical activity in radiation-sterilized materials
		Effects of ionizing radiation on the development of off-odor and flavor in protein
Cornell University	Ithaca, N. Y.	Effect of ionizing radiation on the nutritive value of foods as determined by growth reproduction and lactation study with dogs
		Effects of radiations on structure of fruits and vegetables
Florida State University	Tallahassee, Fla.	Effect of gamma radiation on odor, color and vitamins of meat
General Electric Co.	Milwaukee, Wis.	Study of chemical changes produced in irradiated proteins and amino acids
		Loan of 1-million-volt electron generator with operator
Hormel Institute (University of Minnesota)	Austin, Minn.	Changes produced in lipid materials by high-energy radiations
High Voltage Engineering Co.	*Cambridge, Mass.*	*Rental agreement* for electron accelerator and operator
Howard University	Washington, D. C.	Chemistry of taste
Iowa Agriculture Experiment Station	Ames, Iowa	Chemical changes in protein of sterilized meat
John Hopkins University	Baltimore, Md.	Effects of high levels of ionizing radiations on animal tissues
Laboratory of Vitamin Technology, Inc.	Chicago, Ill.	The use of additives in eliminating the undesirable effects of ionizing radiations
Massachusetts Institute of Technology	Cambridge, Mass.	Development of a method of radiation sterilization of foods without adverse flavor and chemical changes
Midwest Research Institute	Kansas City, Mo.	Study of chemical additives for foods to prevent damage during radiation preservation
National Canners Association	Washington, D. C.	Study of the destruction of food poisoning organisms in canned foods subjected to ionizing radiations
North American Aviation, Inc.	Los Angeles, Calif.	Logistic and economic feasibility study on radiation sterilization of food
Ohio State University Research Foundation	Columbus, Ohio	Chemical and organoleptic changes in carbohydrates and proteins produced by radiation sterilization
Oregon State College	Corvallis, Oreg.	Short- and long-term survival and breeding capacity of rats fed high levels of food stuffs sterilized by ionizing radiation
Oregon State College	Corvallis, Oreg.	A correlation in radiation-sterilized foods of the chemical changes associated with the characteristic flavor development
		Flavors of food sterilized by combining conventional processing with ionizing radiations
		Increasing the acceptance of irradiated meat and meat products by selected treatment before, during, and following irradiation

Table 9.21—(Continued)

Organization	City and state	Title
Pennsylvania State University	University Park, Pa.	Odor and flavor problems in radiation sterilization of foods
Southern Research Institute	Birmingham, Ala.	Study of flavor and chemical changes in food during sterilization by radiation
Southwest Research Institute	San Antonio, Tex.	Detection of radiation induced free radicals by paramagnetic resonance
Stanford Research Institute	Menlo Park, Calif.	Flavor changes induced by radiation sterilization
		Comparative study of the effects of high and low intensity in radiation in food sterilization; irradiation services
Syracuse University	Syracuse, N. Y.	The chemical enhancement of bacterial radiosensitivity in the radiation sterilization of foods
Texas Agriculture Experiment Station (Texas A & M)	College Station Tex.	Long-range investigations of the nutritive properties of irradiated foods
USDA (for work at Agricultural Marketing Service)	Beltsville, Md.	Control of postharvest diseases of fruits and vegetables by radiation treatment
U. S. Testing Co., Inc.	Hoboken, N. J.	Psychometric evaluation of selected irradiated food items
University of California	Los Angeles, Calif.	Study of the mechanism extent and means of prevention of the impairment of the safety and nutritive value of foodstuffs resulting from the initiation of a damaging chain reaction by exposure of essential unsatisfactory fatty acids in foods to the ionizing radiation required for the sterilization of foods
University of California, Food Technology Department	Berkeley, Calif.	Effects of ionizing radiations on caretenoid stability
University of California	Berkeley, Calif.	Relationships of radiation induced fat oxidation and flavor, color, and vitamin changes in meat
		Effects of ionizing radiations on the nutritive and safety characteristics of foodstuffs
University of Chicago	Chicago, Ill.	Effects of irradiation on *Cl. botulinum* toxin subjected to ultracentrifugation
University of Colorado	Boulder, Colo.	The nutritional adequacy and probable toxicity of foods sterilized by ionizing radiation
University of Illinois	Urbana, Ill.	To study the biological value of the protein of foods sterilized by irradiation as compared to the biological value of the protein of foods conventionally sterilized
		Isolation and identification of the stale flavor components which develop in whole dry milk (as a result of irradiation)
University of Illinois, board of trustees	Urbana, Ill.	Studies on radiation sterilization of sliced apples
University of Massachusetts	Amherst, Mass.	Effects of irradiation on pigmented foods used in rations for the Armed Forces
University of Michigan	Ann Arbor, Mich.	Chick feeding studies using gamma irradiated food
University of Michigan	Ann Arbor, Mich.	Gamma ray sprout inhibition of potatoes
		Use of combined heat and radiation for sterilization of foods
University of Rochester	Rochester, N. Y.	The effect of ionizing radiation on the digestion and protein of fat *in vivo*
University of Wisconsin	Madison, Wis.	Inhibition of enzymatic activity in irradiated foodstuff

Table 9.21 — (Continued)

Organization	City and state	Title
Utah State Agriculture Experiment Station	Logan, Utah	Studies on radiation pasteurization and on radiation sterilization of fruit and vegetable products
Wisconsin Alumni Research Foundation	Madison, Wis.	The possible carcinogenicity of irradiated foods Using animal feeding studies on wholesomeness of irradiated foods

highly acceptable products when prepared for the table. The cooked chicken was reported to have a considerably better texture and appearance than commerical canned whole chicken.[111,112]

Figure 9.18 shows photographs of uncooked chicken irradiated at 3 Mrep and stored for 5 weeks at 70°F and similar chicken after cooking.[111]

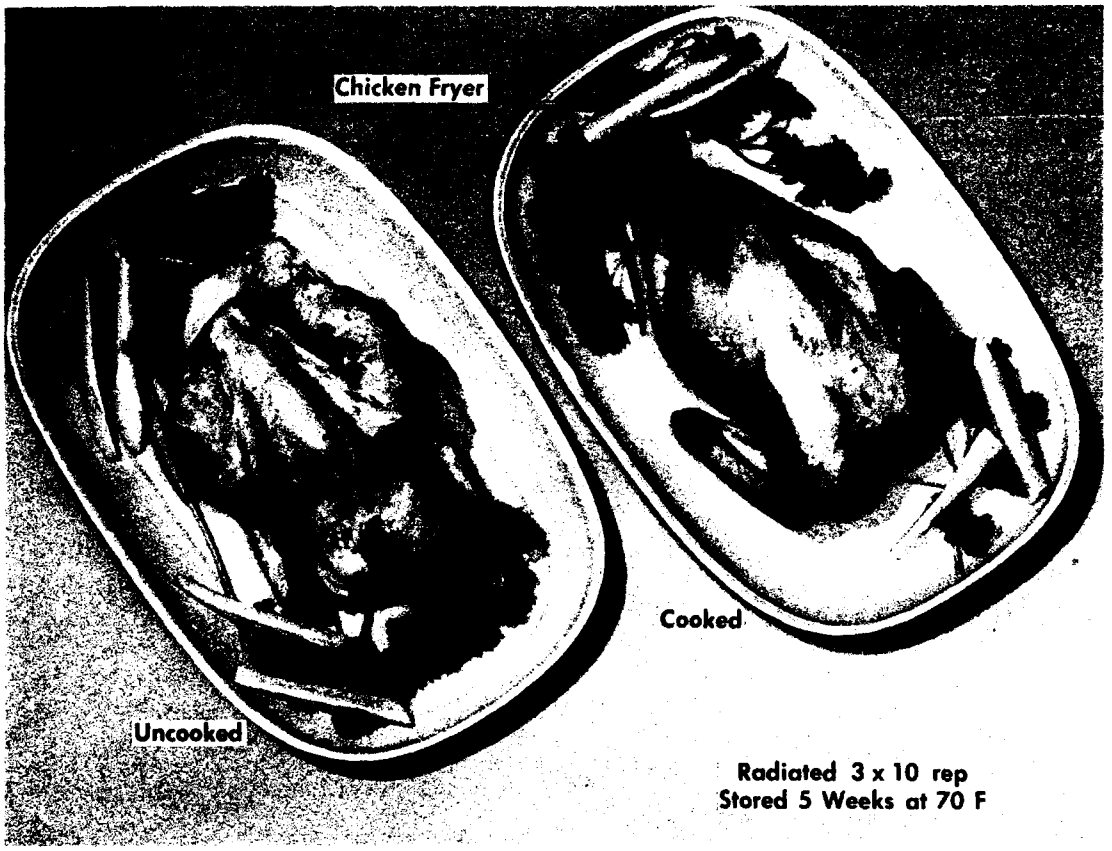


Fig. 9.18 — Chicken fryer. Left: After gamma irradiation and storage. Right: Sample cooked following storage. (Courtesy of Quartermaster Food and Container Institute, U. S. Army)[111]

In recent studies the flavor and odor problems have been explored by investigations of the radiation chemistry of foods. The effect of radiation on various constituents of food is described in Chap. 8. In 1958 Morgan summarized the progress in this direction.[113]

Irradiation of beef has been shown to increase the total carbonyl content.[114] Carbonyls may have a role in reacting with sulfur-containing degradation products produced in meat. Reactions have been postulated and speculations have been made regarding the cause of off-flavors in radiosterilized foods.[115,116] However, Bellamy[117] comments that even though con-

siderable progress has been made in fractionating irradiated meats for off-flavor components there is no general agreement on which of the many isolated products, compounds and types of compounds are chiefly responsible for irradiation induced odors.

EFFECT OF GAMMA IRRADIATION ON THE POTENCY OF PHARMACEUTICAL PRODUCTS

Pharmaceutical products used for parenteral (by injection) therapy must be sterile. Sterilization with heat is usually convenient with products that are not heat-labile. Heat-sensitive products can usually be sterilized by microfiltration and are packaged under sterile conditions. Sterilization of the finished package would assure utmost safety of the product. Sterilization by ionizing radiation offers a number of interesting possibilities.

Several antibiotics were irradiated and checked for potency by Controulis et al.[50] In this study the anhydrous antibiotics were irradiated with a dose of 1.9 Mrep. Following irradiation, the samples (along with control samples) were dissolved in sterile distilled water in appropriate concentrations and cylinder assay tests were carried out for potency of the various antibiotics, using *Staphylococcus aureus* as the test organism. Results are presented in Table 9.22.

Table 9.22—THE EFFECT OF GAMMA RADIATION ON THE POTENCY OF SEVERAL ANTIBIOTICS[50]
[Expressed as diameters (mm) of sterile halo in cylinder test]

	Days after irradiation						
	3 days		6 days		90 days		
Antibiotic	Irrad.	control	Irrad.	control	Irrad.	control	Physical appearance*
K-penicillin "G"	37†	37	30	32	37	37	No color change; no apparent change in solubility
Streptomycin HCl	24	24	18	18	13	13	Slight change in color of treated powder to gray
Aureomycin	31	31	25	20	23	24	No color change; no apparent change in solubility
Chloromycetin	23	23	14	21	16	16	Slight change in color of treated crystals to gray
Terramycin	29	24	17	20	21	21	No color change; no apparent change in solubility

* Immediately after removal from the Co^{60} source following 2.04 -Mrep exposure to gamma radiation.
† Figures represent zones of inhibition against *Staphylococcus aureus* in the cylinder assay test of solutions prepared from treated and untreated powders.

The data in Table 9.22 indicate no appreciable over-all differences in potency between the irradiated and control samples of the several antibiotics. With the exception of penicillin, which appeared to retain its potency while in solution for 90 days, both the irradiated and the control solutions showed losses in potency during storage. The footnote under the table indicates the physical appearances of the powders or crystals soon after irradiation. A slight change in color, from white to gray, was noted in the irradiated samples of streptomycin and chloromycetin. However, there did not appear to be any differences in the solubilities of these samples in comparison with the control samples.

Table 9.23 gives some additional results reported by Controulis et al.[50] on the gamma-ray sterilization of a variety of pharmaceutical products and Table 9.24 is a similar table reported by Huber[51] using electron sterilization.

All the pharmaceuticals listed in Table 9.23 were contaminated before irradiation and were found to be sterile after receiving an irradiation dose of 2 Mrep. Had the solution been

Table 9.23—RESULTS OF Co^{60} GAMMA-RADIATION STERILIZATION OF VARIOUS PHARMACEUTICALS[50]

Sample No. and product	Tests	Potency of uncontaminated samples, irradiation dose, Mrep		
		Control	0.085	2
Calcium gluconate	Activity	OK	OK	OK
Ascorbic acid	Activity, mg/ml	275.5	283.3	273.0
Theelin aq. suspension	Activity, mg/ml	2.70	2.21	2.19
Pitocin	Activity	100%	50%	0.02%
Diptheria-tetanus-pertussin combination	Agglutination tetanus combining power, unit/ml	0.8	0.8	0.8
Tetanus antitoxin	Activity, unit/ml	1900	1900	1500
Antuitrin growth*	Activity			

*Trademark registered by Parke, Davis & Company for anterior pituitary gland fraction.

Table 9.24—DATA ON STERILIZATION OF PHARMACEUTICALS WITH ELECTRONS FROM A CAPACITRON[51]

Pharmaceutical	Impulses/μ sec	Contamination	Potency		Container
			Untreated	Treated	
Thiamine HCl	2–4	Unknown	4.6 mg/cm^3	4.4 mg/cm^3	Glass
Protein Hydrolysate	2	GNB, GPC			Aluminum
Penicillin-Na	3	*B. subtilis* fungi spores	220,000 units	220,000 units	Glass
Streptomycin-H_2SO_4	4	*B. subtilis*	100,000 units	100,000 units	Glass
Testosterone	4	*B. subtilis*	100%	100%	Glass
Prolactane	4		100%	110%	Glass
Pituitary hormone	2	Unknown	3800*	4000*	Plastic
Hyaluronidase	2	Unknown	100%	97%	Plastic
Clarase	2	Unknown	100%	87%	Aluminum
Trypsin	2	Unknown	100%	100%	Aluminum
Yeast	2–4	*B. subtilis*			Plastic
Riboflavin			0.07 mg/g	0.07 mg/g	
Pyridoxine			0.04 mg/g	0.04 mg/g	
Pantothenic acid			0.10 mg/g	0.10 mg/g	
Niacin			0.35 mg/g	0.35 mg/g	

*International units.

more concentrated, or had other pharmaceuticals been used, a larger radiation dose might have been required for sterility because of the "protective effect" described in Chap. 8 in the discussion on microorganisms. Pitocin a pituitary gland hormone preparation, is an example of a pharmaceutical whose potency is drastically affected by irradiation.

Additional information on radiation preservation of food is offered in Refs. 52 to 118.

COMMERCIAL RADIOSTERILIZATION OF SURGICAL SUTURES

In 1959 Artandi and Van Winkel[118] described the commercial sterilization of surgical "catgut" sutures at the Ethicon Division of Johnson and Johnson Co. using an electron beam from a linear accelerator. Gamma radiation was not selected because deep penetration was not required for the packaged sutures which are of small size. The minimum killing dose was established for over 150 different species of microorganisms. A dose of 2.5 Mrads was selected which is 40 per cent above the minimum required to kill the most resistant organism. It was found that sutures of the catgut type can be given a 5-Mrad dose before the properties are reduced to those of heat-sterilized sutures.

The substitution of radiation for heat results in stronger sutures. Other advantages are a better package of aluminum foil which is more simple to manufacture than the sealed glass packages used for heat-sterilized sutures. Also use of radiation permits continuous processing whereas heat sterilization is a batch operation. This commercial use of radiation is a milestone in the application of this new process.

WHOLESOMENESS OF IRRADIATED FOODS AND FOOD CONSTITUENTS

The lethal effect of gamma radiation on microorganisms offers considerable promise for processing perishable foods to increase storage life. But, since gamma radiation also induces some chemical changes in foods, it must be established that they are still wholesome before they are eaten. There is some vitamin loss in irradiated foods, as in other processing methods such as canning. It is necessary to prove that a diet consisting mostly of irradiated food can, in addition to having no toxic effects, support normal growth and maintain health over long periods.

PROCEDURES RECOMMENDED BY FOOD AND DRUG ADMINISTRATION

Lehman and coworkers of the Food and Drug Administration staff have outlined the principles and methods required for investigating wholesomeness of irradiated food.[119–120] These authorities recommend that the studies be directed along two major lines of approach: potential toxicity and nutritional adequacy. Such considerations as freedom from radioactivity, carcinogenicity, and antigenicity must be included in toxicity studies on irradiated foods. Chemical and physical tests should precede animal tests to provide possible clues which may enable investigators to plan animal experiments with greater intelligence and insight. For disclosing toxic effects, the animals should be challenged with a relative excess of the substances under consideration; for nutritional adequacy the challenge should be made by reducing the supplement to a level where the inadequacy is intensified. In addition to the experiments with normal, healthy animals, supplemental investigations should be undertaken with animals given noxious substances which induce a state of stress and thereby increase the susceptibility to toxic products. Because different species of animals and even various strains of the same species frequently differ in sensitivity, several species are required.

In all toxicity studies, it must be kept in mind that irradiation-induced changes in the food are extremely subtle. It is desirable, therefore, to include in the experimental plan a measure of cellular metabolism as a supplement to the usual gross observations of growth, reproduction, etc. Such an approach might be achieved through the determination of enzyme concentration in the tissues of the animals fed the experimental diets.

POSSIBILITY OF INDUCED RADIOACTIVITY

The question of radioactivity induced in elements present in foods is relevant to the problem of wholesomeness. Gamma radiation from Co^{60}, or pure fission products, is not believed

to produce detectable nuclear activation except in the element indium. To investigate this, experiments were performed by Meinke and Brownell[8,130] in which 25 food elements, listed below, were subjected to doses of Co^{60} gamma radiation up to 100 Mrep.

Aluminum	Iron	Potassium
Boron	Iodine	Silicon
Calcium	Lead	Silver
Carbon	Magnesium	Sodium
Chromium	Manganese	Sulfur
Chlorine	Molybdenum	Tin
Cobalt	Nickel	Zinc
Copper	Oxygen	
Fluorine	Phosphorus	

Cobalt-60 emits two gamma photons of 1.17- and 1.33-Mev energy. As expected, the gamma activation of indium was found, but the instrument (a scintillation well-counter) detected no activity above background even when the 25 elements received high doses. These elements were chosen on the basis of complete mineral analysis of beef. Although these studies were not considered complete, the conclusion was that radioactivity in foods induced by exposure to gamma radiation of about 1-Mev energy is not a problem.[8,130]

Hannan[29] has compiled from literature the threshold energy levels for inducing radioactivity by gamma rays. Table 9.25 lists various isotopes of elements in the order of decreasing abundance in biological materials.

Table 9.25—THRESHOLD ENERGY LEVELS FOR THE PRODUCTION OF INDUCED RADIOACTIVITY IN VARIOUS ELEMENTS* BY IRRADIATION WITH HIGH ENERGY ELECTROMAGNETIC RADIATION[29]

Element	Type of nuclear change	Threshold energy, Mev	Half-life of product	Refs.
C^{12}	γ, n	18.7	21 m	143
O^{16}	γ, n	16.3	2.1 m	143
N^{14}	γ, n	10.65	10 m	145
P^{31}	γ, n	12.35	25 m	145
K^{39}	γ, n	13.2	7.5 s	145
S^{32}	γ, n	14.8	3.2 s	145
Ca^{40}	γ, n	15.9	1 s	145
Fe^{54}	γ, n	13.8	8.9 m	145
Mg^{24}	γ, n	16.2	11.6 s	145
Mg^{25}	γ, p	11.5	14.8 h	145
Mg^{26}	γ, p	14.0	62 s	145
Cu^{63}	γ, n	10.9	10 m	145
Cu^{65}	γ, n	10.2	12.8 h	145
I^{127}	γ, n	9.3	13 d	145
Br^{81}	γ, n	10.7	6.4 m	145
Al^{27}	γ, n	14.0	7 s	145
Si^{28}	γ, n	16.8	5 s	145
Li^{7}	γ, p	9.8	0.85 s	145
Be^{9}	γ, n	1.67	very short	145
H^{2}	γ, n	2.2		145

*The elements are listed very approximately in order of their abundance in biological material.

The gamma energies required to induce activity is, in most cases, much greater than the gamma energy of fission products or isotope sources. The last two elements produce a γ-n reaction with low energy gamma rays, but the product Be^{8} has a very short half-life and deuterium produces nonradioactive hydrogen.

597082 O-61—22

Since neutrons are produced in γ-n reactions, they can in turn induce nuclear reactions in other atoms. It has been estimated that a neutron flux of 10^5 to 10^6 neutrons/cm^2-sec for periods up to 30 min may be permitted without producing appreciable activation. Based on abundance of various elements listed in Table 9.25 and their capture cross-section, the possibility of producing significant neutron flux is considered quite remote.

The use of accelerated electrons for food irradiation requires attention to e,n reactions, and γ-n reactions, the gamma rays (bremsstrahlung) being generated by the passage of the electron beam through the sample.

Ovadia et al.[122] have reported a study of this problem, using selected samples irradiated with a high-intensity 25-Mev electron beam from a linear accelerator. The air dose resulting from the radioactivity of processed foods was evalulated for typical irradiation schedules, and the storage time required for the dose rate to fall below permissible levels was evaluated. These values apply to the processing of organic substances which are composed mainly of carbon, nitrogen, and oxygen. It was concluded that only short-lived activities of carbon and chlorine are objectionable. However, with an adequate storage time, this activity can be reduced to an insignificant level.[122]

POSSIBILITY OF TOXICITY

The irradiation-induced reactions associated with various constituents of food have been described. Some of the compounds resulting from such reactions might possibly present problems of toxicity of irradiated food. The questions of toxicity can be explored by means of animal feeding studies. Many such studies have been made and others are also currently in process.

Short-term feeding experiments with rats have been reported by Brownell[106,132,133] and others. The Office of The Surgeon General, United States Army, and its contractors have undertaken short-term studies with animals and also human volunteers. The results of some of these studies have been reported by Reed, Kraybill, and Witt,[123-126] Teply and Kline[127,128] and Benham et al.[129]

The first long-term feeding experiments with electron-irradiated raw meat fed to albino rats were reported by Poling et al., of Swift and Company.[131] Similar experiments at The University of Michigan using gamma-irradiated cooked meat were reported by Brownell et al.[8,106,133] Various other laboratories are currently engaged in long-term feeding studies and some of these are listed in Table 9.28. In addition to using the albino rat as an experimental animal, long-term studies are being conducted with dogs and monkeys.[117]

Short-term Experiments with Rats

Short-term animal feeding studies were undertaken by Brownell et al.,[106,132,133] to determine the minimum irradiation dose required to produce acute toxicity in diets—if, indeed, radiation could cause toxicity. Albino rats were fed a diet consisting of 50 per cent canned cooked meat. The diet was prepared in two parts, protein-carbohydrates and vitamins. The first part consisted of canned meat, casein, corn starch, and alpha cellulose. The second part included the vitamins, either in pure form or as yeast or liver extracts. Water content was increased to 44 per cent. Because water was present, it was possible to study the indirect effect of gamma radiation on food as a result of free radical formation in water. In the first short-term experiment, the irradiation dose was 2 Mrep. One group of rats was fed the nonirradiated diet. The second group was fed the diet in which only the protein and carbohydrate portion was irradiated and to which the nonirradiated vitamin supplement was added. The third (control) group was fed an entirely irradiated diet. All three groups were allowed to eat as much as, and when, they wanted. After 2 months it was observed that, whether the diet was all irradiated, partially irradiated, or nonirradiated, the growth of all rats, male and female, was essentially the same.

The same procedure was followed in another short-term experiment, but the irradiation dose was raised to 20 Mrep. Animals fed on these diets showed little difference during the first 3 weeks. After 7 weeks, growth of males and females was about the same for both the control and the partially irradiated diets. The entirely irradiated diet supported very poor

growth,[106,133] as shown in Fig. 9.19. However, when animals from this group had diet supplemented by the nonirradiated vitamin mixture, they recovered dramatically, as shown in Fig. 9.19. They mated and had normal offspring.[106,133]

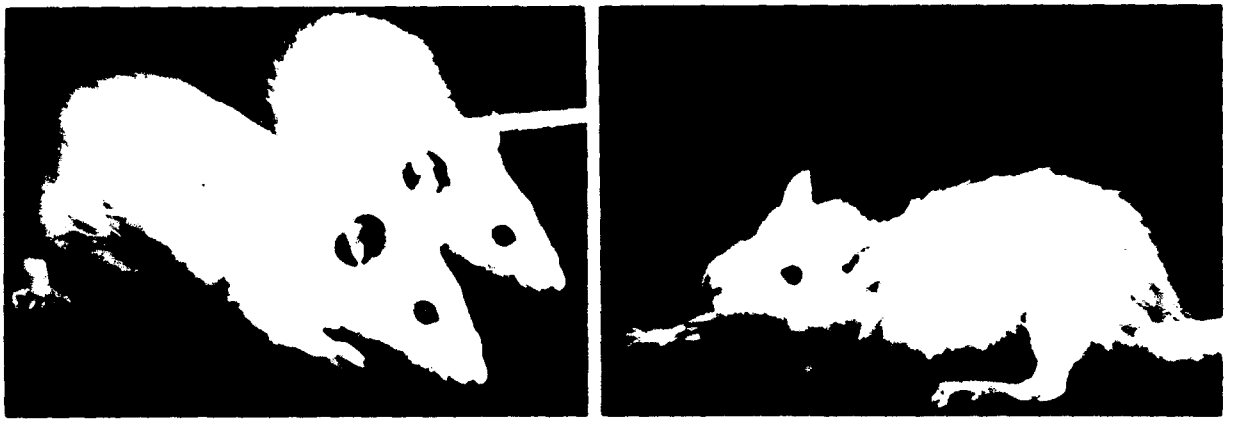


Fig. 9.19—Three rats that were fed a completely irradiated diet (at 20 Mrep) for 10 weeks; the two on the left after being fed for three weeks on the same diet supplemented with a water-soluble vitamin premix; the one on the right before receiving the special diet to correct thiamin deficiency.[106,133]

From this experiment it was concluded that there was vitamin loss at a 20-Mrep dose, but that no acute toxicity was produced in the food because there was normal growth on an irradiated diet supplemented with nonirradiated vitamins.

In a third short-term experiment, the irradiation dose was raised to 45 Mrep in an attempt to find a threshold of acute toxicity.[8] Because diets receiving this much irradiation become quite unacceptable, even to the rats, it was necessary to force-feed the animals by tube. It was also evident from almost identical weight curves, shown in Fig. 9.20, that there is no acute toxicity observed when diets are irradiated with 45 Mrep and fed to rats for a period of a month. These short-term experiments indicate that while vitamin destruction occurs with

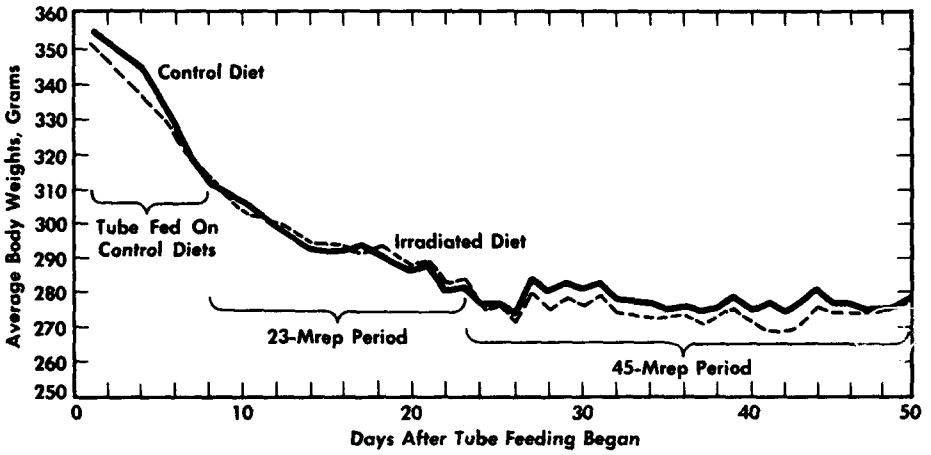


Fig. 9.20—Average weights of albino rats tube-fed on a control diet and a diet receiving a radiation dose of 45 Mrep.[8,133]

Table 9.26—SUMMARY OF REPRODUCTION PERFORMANCE IN LONG-TERM EXPERIMENT[133]

	1st breeding of parent gener.		2nd breeding of parent gener.		1st breeding of 1st filial gener.		2nd breeding of 1st filial gener.		1st breeding of 2nd filial gener.		2nd breeding of 2nd filial gener.	
	Con. group	Exp. group	Con. group	Exp. group	Con. group	Exp. group	Con. group	Exp. group	Con. group	Exp. group	Con. group	Exp. group
No. females bred	20	20	20	20	20	20	20	20	20	20	20	20
No. males used	20	20	20	20	12	12	12	12	10	10	10	10
No. females sterile*	2	5	2	6	2	1	3	2	1	0	0	0
No. males sterile†	8	9	10	5	4	3	3	3	0	2	2	2
No. males not proven‡	2	3	2	2	0	0	2	2	0	0	0	0
No. females conceiving first week	8	3	5	4	6	5	10	12	12	12	10	8
No. females conceiving second week	1	3	3	4	6	6	3	4	6	4	5	6
No. females conceiving third week	3	4	4	3	3	7	3	0	1	2	3	1
No. females conceiving fourth week	0	0	3	2	0	0	1	0	0	1	1	1
No. females conceiving after fourth week	4	5	1	1	3	1	0	2	0	1	1	2
No. females resorbing fetuses	2	0	2	0	0	0	0	0	0	0	0	0
Total No. litters born	16	15	16	14	18	19	17	18	19	20	20	18
Total No. litters born dead	1	1	1	0	2	0	1	1	0	0	0	0
Total No. litters born alive, not surviving weaning	1	1	1	3	0	1	1	0**	3	3	6	2
Total No. pups born	157	119	145	115	184	195	173	188	195	204	161	163
Total No. pups born dead	5	3	17	11	22	9	17	13	3	0	3	4
% pups born dead	3.18	2.52	11.72	9.56	11.95	4.62	9.83	6.91	1.5	0	1.87	2.45
No. pups born alive per female bred	7.6	5.8	6.4	5.2	8.1	8.3	7.8	8.8	9.6	10.2	7.9	8.8‡
Total No. pups born alive, not surviving weaning	25	21	30	23	17	24	21	14**	64	76	88	59
Average No. pups born per litter	9.8	7.9	9.1	8.2	10.22	10.26	10.18	10.44	10.3	10.2	8.05	9.05
Average No. pups per litter at 5 days	8	6.6	6.1	5.6	8.4	9.3	8.5	9.2	8.7	8.4	7.1	7.5
Average No. pups per litter at 21 days	8	6.6	6.1	5.6	8.0	8.6	7.6	8.7**	6.7	6.4	3.5	5.6
Total No. young disposed of¶	0	0	0	0	0	0	5	7	3	7	0	0
Average weight of young at 21 days, g	48.6	45.5	57.8	52.7	45.5	47.7	52.9	49.9**	27.1	27.0	‡‡	‡‡
Total No. young reaching weaning§	127	95	98	81	145	162	135	161**	128	128	70	100
Average No. young weaned per female bred	6.43	4.8	4.9	4.1	7.2	8.1	6.5	8.1**	6.4	6.4	3.5	5.9
% pups born alive which survived 21 days	83.5	81.8	76.6	77.9	89.4	87.1	86.5	92.0	66.7	62.7	44.3	62.9

* Mated six times unsuccessfully.

† Mated unsuccessfully with at least one female which later became pregnant by another male.

‡ Mated only with pregnant or sterile females.

¶ For purposes of reducing litters to ten after birth.

§ Does not allow for animals that were disposed that might have survived to weaning age.

** Data do not include those of pups from females conceiving after 4th week.

†† Two females on experimental diet died during breeding. They were not counted in computing averages.

‡‡ The young were noticeably low on weight at weaning. They were often left with their mothers after weaning date in an attempt to enable them to grow at the optimum rate; therefore, we do not have any accurate weaning-weight data on these animals.

doses of gamma radiation that might be used for food processing, animals fed diets irradiated at levels perhaps 8 to 10 times greater than the highest doses for processing foods, exhibit no toxic effects.

Long-term Experiments with Rats

Brownell et al., reported (Refs. 8, 106, 132, and 133) the first long-term feeding experiments to study growth, reproduction, hematology, and pathology of male and female albino rats fed a gamma-irradiated diet throughout their lives. The diets were basically the same as in the short-term studies described, except that in the irradiated diet, vitamin and fat supplements were nonirradiated; the bulk of the diet (88.4 per cent) containing the protein, carbohydrate, and water received a 4-Mrep gamma dose. From parents on each diet, three successive generations of rats were born and raised. Rats in each generation were bred twice. Weanlings from the second breeding were selected to breed the successive generation. Data on growth and reproduction were obtained on these offspring as well as on the parent animals.

In all four generations there was no appreciable variation in weight between the rats fed the irradiated diet and those fed the control (nonirradiated) diet. The increase in weight in each generation showed the same trend. The entire experiment covered over 100 weeks.

Table 9.26 presents the data from the first and second breedings of the parent, first fillial, and second fillial generation rats. The data reveals no consistent superiority of animals fed the nonirradiated diet over those fed the irradiated diet.

In the same study, it is reported that on the basis of observations with respect to incidence of disease, efficiency of food utilization, and hematological and pathological examinations, no significant differences were found to occur. Table 9.27 summarizes the hematological studies.[133]

Table 9.27—AVERAGE RESULTS OF HEMATOLOGICAL STUDIES OF PARENT RATS* IN LONG-TERM EXPERIMENT[133]

Group	Hemoglobin, g/100 cm^3	Hematocrit, %	Reticulocytes, %	White count, No./mm^3	Eosinophil count, No./mm^3	Differential count: Lymphocytes, %	Differential count: Polymorphonuclear leukocytes, %
Control males	14.9	47.4	2.9	10,760	119	80.0	18.1
Control females	14.5	45.5	3.0	9,550	120.0	81.0	17.3
Exptl. males	15.0	47.7	2.5	13,390	113.0	83.7	14.7
Exptl. females	14.8	45.9	1.7	11,260	128.0	81.7	16.3

* Five to seven test animals were used for each test period.

In the report originating at Swift and Company,[131] on the study of animals fed with air electron-irradiated diet, similar conclusions were drawn. A total of 2685 albino rats was observed over three generations. The experimental animals were fed with ground beef (60 per cent of diet) that was given an electron-radiation dose of 2 Mrep. After a preliminary finding of decrease in male fertility, the diet was supplemented with vitamin E to correct the deficiency.

Other Short-term Feeding Experiments

Several irradiated diets other than those employed in the University of Michigan experiments have been used elsewhere. Some contained egg powder, milk, peaches, ham, and cereals.[134] Kraybill et al.,[134] report data on growth obtained with the last four foods using gamma doses of 3 and 6 Mrep. They also reported studies on ground beef, sliced bacon, haddock fillet, green beans, whole kernel corn, sliced beets, frozen strawberries, bread, military cereal bar, and powdered whole milk.[134]

In these studies the diet consisted of 65 per cent (dry weight) of nutritionally adequate basal ration and 35 per cent (dry weight) of the food to be tested, plus a nonirradiated vitamin supplement. Parallel experiments were conducted with animals fed nonirradiated control diets. Experimental animals ate and grew well on all diets at radiation levels studied (3 and 6 Mrep). Histopathological examination of representative tissue and also gross observations indicated that all animals raised on all diets grew and developed satisfactorily and maintained good health.[134]

Long-term Experiments with Chickens

In addition to experiments on mammals, two-year studies with 183 parent chickens and 3 subsequent generations were conducted at the University of Michigan as part of the Surgeon General's program.[135] Chickens were kept for 13 months on a wet-mash diet that had received a dose of 3 Mrep of gamma radiation. The diet consisted of nutritionally complete commercial poultry mashes. Four different mashes were fed at appropriate stages as the fowl developed from chicks to breeding pullets. The basic food substances in these mashes are in human diet in other forms. The mash was mixed with water equal to 84 per cent to its dry weight, and vitamin supplements were added after irradiation.[135]

Factors studied were: mean weekly body weight, efficiency of food utilization, egg production and percentage of hens laying, fertility of eggs following artificial insemination, hatchability and embryonic mortality, growth of chicks hatched, blood cell counts, and gross pathology and histopathology of tissues removed from birds dead or killed.

A slight lag in the growth rate was observed for the experimental groups, as compared to the control groups.[135] This was similar to observations made during feeding experiments with rats and mice in which diets of very different nature were used.

No difference was apparent between egg-laying of the experimental and control groups. Tests on the eggs of both groups showed them about equally fertile and hatchable. Blood count data revealed no significant difference between the control and experimental fowl. Pathological studies of the two groups had similar results. The only apparent effects were a slightly reduced growth rate and a slight delay in reaching maximum hatchability for pullets on the irradiated diet. The authors concluded that the evidence indicated no chronic or subacute toxicity, or gross nutritional losses other than vitamin losses.[135]

Long-term Experiments with Dogs and Monkeys

Several other studies currently being made involve the use of dogs and monkeys as test animals and a variety of foods as listed in Table 9.28. Foods were irradiated at 2.79 and 5.79 Mrads, and stored for a minimum of three months. A control diet containing unirradiated food was included in all tests.

The results obtained, as yet, show no apparent manifestation of toxicity, nor abnormalities in blood haemaglobin content, red and white blood cell counts, or differential counts.[26]

POSSIBLE CARCINOGENICITY OF IRRADIATED FOODS

In an interim paper, Teply and Kline reported experiments on wholesomeness and possible carcinogenicity studies conducted at Wisconsin Alumni Research Foundation.[136] The irradiated food included apricots, asparagus, Brussels sprouts, cabbage, cauliflower, celery, cherries, chicken, corned beef, cranberries, crackers, gelatin dessert powder, macaroni, mushrooms, nut roll, pears, peas, pork sausages, sweet and white potatoes, pound cake, salmon, shrimp, and tuna. The foods were irradiated with 3- and 6-Mrep doses. Control foods were not irradiated. Short-term feeding studies were conducted on rats with these foods mixed with 35 per cent basal ration. There was no significant difference between rats fed control and irradiated foods except in one experiment where a decreased growth rate was observed due to the effect of radiation on the sucrose which forms 90 per cent of gelatin desert powder.

Experiments to test possible carcinogenicity are extremely difficult. These are the only experiments designed specifically to test this possible effect of irradiation. Groups of mice were subjected to experiments embodying the following general principles:

1. Testing of food materials irradiated with substerilizing, sterilizing, and above-sterilizing doses.
2. Uses of both beta and gamma radiation.
3. Feeding, skin painting, and subcutaneous injection experiments.
4. Emphasis on, but not restricted to, sterol fractions.
5. Use of various strains of mice.
6. Use of known carcinogens in some of the tests.

The authors concluded that there was no clear indication of the production of carcinogens by irradiation of the food materials studied.[136]

Table 9.28—PARTICIPATING RESEARCH INSTITUTIONS IN SURGEON GENERAL'S PROGRAM OF IRRADIATED FOODS[111]

Institution	Research
Columbia University	Vitamin E studies
Alabama Polytechnic Institute	Micronutrients
University of Illinois	Micronutrients
Texas A & M College	Longevity, reproduction, and lactation of rats
University of Michigan	Long-term feeding, rats and chickens
Oregon State College	Long-term feeding of rats with pork and organ meats
University of California at Los Angeles	Feeding studies of rats with irradiated fats
Wisconsin Alumni Research Foundation	Carcinogenicity, mice
Armour Research Foundation	Short-term feeding, rats
Cornell University	Long-term feeding, dogs
University of Rochester	Digestibility studies, dogs
Johns Hopkins University (McCollum Pratt Institute)	Feeding studies on rats with degradation products of irradiated foods
University of Colorado, Medical Nutrition Laboratory	Long-term feeding, rats Short-term feeding, humans
Hazelton Laboratories	Long-term feeding of rats with corn and tuna
University of Miami	Long-term feeding of rats with beef stew. Long-term feeding of dogs with beef stew
Vanderbilt University	Long-term feeding of rats with beef. Long-term feeding of beagles with boned chicken, ground beef, or pineapple jam
University of Syracuse	Long-term feeding of rats with chicken stew and cabbage
Virginia Polytechnic Institute	Long-term feeding of dogs with shrimp
Georgia Coastal Experimental Station	Long-term feeding of beagles with bacon
Massachusetts Institute of Technology	Long-term feeding of dogs with dried whole eggs
Industrial Bio-test Laboratories	Carcinogenicity studies

FEEDING IRRADIATED FOODS TO HUMAN BEINGS

Short-term feeding of irradiated foods to human volunteers was studied[111] at the Medical Nutrition Laboratory, Fitzsimmons Army Hospital, Denver, Colo., from June 1955 until December 1958. Four studies, each involving 9 or 10 subjects, were conducted during 1955 and 1956. Each study lasted about 36 days. During the first 15 days of each study, half the subjects ate nonirradiated food and the others irradiated food. A rest period of about 6 days followed, after which the groups were reversed so that each subject ate both diets. At no time were the volunteers told which diet was irradiated.[111]

Irradiated foods used in these studies were first checked for absence of toxicity by short-term feeding experiments with laboratory animals. The foods were then checked for palatability by taste panel tests at the Quartermaster Corps Food Container Institute. Table 9.29 lists foods tested in the four studies. The selection of irradiated food was increased with each successive study.[111]

Table 9.29—IRRADIATED FOODS USED IN FEEDING STUDIES WITH HUMAN VOLUNTEERS[111]

Study No.	Total calories in diet provided by irradiated food, %	Foods tested		
1	35	Bacon	Peaches	Cereal bar
		Beef, ground	Strawberries	Bread
		Haddock	Green beans	Powdered milk
		Ham, ground	Beets	
		All items above plus:		
2	65	Pork sausage	Pound cake	Shrimps
		Macaroni	Green peas	Dried pears
		All items above plus:		
3	82	Corned beef	Celery	Crackers
		Carrots	Brussel sprouts	Tuna
		Mushrooms		Frankfurters
		Potatoes, white	Melon balls	Dessert powder
			Cherries	Rice
		Cauliflower	Salmon	Pound cake
		Cabbage	Lima beans	Macaroni
		Asparagus	Peanut butter	Bread
		Beef, ground	Powdered milk	
		Pork sausage	Pears	
		Bacon		
		All items above plus:		
4	100	Sweet potatoes	Corn	Strawberry jam
		Peas	Dried apricots	Sugar
		Nut roll	Tenderloin steaks	Ham steaks

Table 9.30—MEDICAL PROCEDURES FOLLOWED FOR HUMAN TEST SUBJECTS[111]

General physical examination	Weekly during studies
Chest X ray	Before each study
Liver function test	Weekly during studies
Kidney function test	End of each period
Blood counts	Twice weekly
Electrocardiograms	End of each period
Urinalysis	End of each period
Dental examinations	Before and after each study
Eye examinations	Before and after each study
Body weight	Daily during studies
Blood pressure	Twice daily during studies
Temperature, pulse, respiration	Twice daily during studies

Throughout the studies, subjects were constantly tested for possible toxic effects. Tests included frequent physical examinations, blood counts, studies of liver and kidney function, electrocardiograms, basal metabolic rates, and X-ray studies. Extensive energy-balance studies of the diets and analyses of the excreta were made. A summary of the medical procedures for the human test subjects is given in Table 9.30. One of the subjects is shown undergoing a basal metabolism test in Fig. 9.21.[111]



Fig. 9.21—Collection of air expired from lungs of test subjects for basal metabolism test. (Courtesy of Medical Nutrition Laboratory, Office of the Surgeon General)[111]

These completed tests showed no significant difference in the general health of the subjects and no evidence of toxic effects for any of the irradiated foods. In a few foods small differences were found in the caloric value per gram of wet food between irradiated and control foods. The significance of these differences has not been determined. Since the first study with human volunteers was completed, the volunteers have been examined at intervals of 3 months but no evidence of residual toxic effects from irradiated foods has been observed.

GENERAL COMMENTS ON WHOLESOMENESS OF IRRADIATED FOOD

Short-term studies with rats indicated that, as compared to changes in palatability and vitamin content, acute toxicity is an unimportant aspect of food irradiated at levels much higher than those used commercial pasteurizing or sterilizing with gamma radiation. Results of long-term feeding and breeding studies indicate no chronic or accumulative effects on rats from nearly 2 years of diet in which all but the vitamin and mineral supplements, and part of the fat supplement, received a 4-Mrep dose of gamma radiation.[133]

Animals fed the irradiated diet grew, reproduced, and lactated the same as those on the nonirradiated diets. No significant differences were detected between the two groups in hematology, pathology, or in the gross observations.[133]

The U. S. Army is extensively studying food processing with ionizing radiation. The chief object is to improve the quality of preserved rations used by the Armed Forces. As part of this program, the Division of Research and Development, Office of the Surgeon General, Department of the U. S. Army, is investigating wholesomeness of irradiated foods. The primary object is to determine whether irradiated foods may be fed safely to human beings. These studies are being conducted at the Medical Nutrition Laboratory, Fitzsimmons Army Hospital, and at a number of universities and privately sponsored research institutes. The various laboratories were listed in Table 9.28.

The broad program of study listed in Table 9.28 is designed to prove the safety of irradiated and preserved foods. Both potential toxicity and nutritional adequacy of irradiated foods are being explored. Possible subtle effects of irradiated foods and obscure toxicity are being checked by means of long-term feeding of experimental animals. Possible carcinogenicity is being tested on rats and mice.[111]

STATUS OF WHOLESOMENESS OF IRRADIATED FOOD AS OF 1960

In August 1959 the Chicago Midway Laboratory of the University of Chicago reported on a special experiment to determine the possibility of induced radioactivity in beef[137] irradiated with gammas from Co^{60}. Four pounds of beef were given a 10-Mrad dose in a 55-kilocurie Co^{60} source. The conclusion was that "There is no measurable difference between the irradiated and nonirradiated samples, using the most reliable techniques and equipment available anywhere in this country."[137]

At the end of the summer of 1959 the decision was made to cancel construction of the $7.5 million food irradiation center of the U. S. Army. The issue was confused by the release of partial information on incompleted studies of contractors of the Surgeon General's Office, U. S. Army. Some of the information picked up by the press was that irradiated food caused: (1) bleeding in rats, (2) blinding of rats, (3) sterility in dogs, and (4) enlargement of the heart auricle in mice.[138]

Kraybill, a recognized authority on the wholesomeness of irradiated food reviewed each of these observations (January 1960) and pointed out that "There is no evidence that there is any toxicity in irradiated foods for either man or animals. The false conclusion that there is such evidence has arisen from misleading information published by the press relative to biological response of experimental animals fed with irradiated foods."[139]

He pointed out that the bleeding observed[140] in some experiments with rats was due to a deficiency of vitamin K as a result of destruction of the vitamin in the diet and a failure of the animal to synthesize the vitamin (with rat generally is able to synthesize vitamin K). Diets containing cooked meats, heated fats, or soybean protein will also produce a vitamin K deficiency resulting in bleeding if a supplement of the vitamin is not added to the diet. Addition of foods containing vitamin K or vitamin D will prevent bleeding (hemorrhagic syndrome). Thus, this is a problem of proper diet and not a problem of toxicity.[139]

In the case of blindness in one research study[141] it was noted with a particular strain of rats that some of the offspring of both the control animals and those on the irradiated diets were born blind. The incidence of blindness was low in both groups and repeated experiments showed slightly lower incidence in animals on the irradiated diets. Congenital blindness has been observed with this strain of rats in other unrelated feeding experiments. Kraybill concludes that "the incidence of blindness is not associated with the irradiation of food."[139]

In the case of sterility in dogs only a few female dogs were involved in the observations. Also these limited observations are not consistent with those of other investigators who fed irradiated foods with supplements to dogs. Burns et al.[135] observed a loss in fertility in chickens when the oil-soluble vitamin supplement was omitted from the irradiated diet. Fertility was restored by returning the Vitamin D and E additions to the diet. If a loss in fertility is proven to exist in dogs fed an irradiated diet (shown by a significant number of animals), it is believed to be a vitamin deficiency problem (as in the case of bleeding from vitamin K deficiency) and not a problem of toxicity.[139]

In the case of enlargement of the heart auricle in mice it was observed in 1958 that over a period of 3 months some animals on the irradiated diet developed rupture on the auricle. In later studies the auricle rupture could not be duplicated with any strain of mice tested regardless of whether they were fed irradiated or nonirradiated diet. Also the omission or supplementation of vitamins in the diet failed to explain the earlier observations. If the irradiation of the diet had been responsible, it should have been possible to demonstrate the effect in repeat analogous experiments. Since this was not observed it can not be concluded that the enlargement of the heart auricle observed over a short period was caused by irradiation of the diet. Instead it seems more likely that some accidental unknown factor entered into the experiment. Possibly some toxic material accidently entered the diet during this period. Kraybill comments "It is quite evident, however, that no toxicological property can be associated or related to the feeding of irradiated foods. In addition such observations (on heart defect) have not been confirmed in the feeding of a broad spectrum of irradiated foods to hundreds of mice in the work reported by other investigators."[139]

The establishment of the wholesomeness of irradiated food requires the performance of many negative type experiments with many animals and many foods. If the irradiated food is as wholesome as the control food the results will be negative and no significant differences will be observed between the two groups of animals. However, if a difference is observed it must be explored and the reason for the difference established. To date no observations have been made that establish any lack of wholesomeness in irradiated foods. There is some destruction of vitamins which increases with dosage, but the vitamin loss is usually less than that in heat-processed food.

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Chapter 10

POSSIBLE PRACTICAL APPLICATIONS OF THE BIOLOGICAL EFFECTS OF GAMMA RADIATION

The uses of trace amounts of radioisotopes in research was described in Chap. 6. In Chaps. 4 and 5 the uses of larger quantities of radioisotopes in various industrial applications and in chemistry were discussed. Chapters 8 and 9 were devoted to descriptions of the effects of ionizing radiations on various biological chemicals, simple organisms, plants, and plant and animal products. However, possible commercial applications of these effects were only briefly mentioned.

This chapter deals with some selected possibilities in the commercial application of some biological effects of gamma radiation described in Chaps. 8 and 9. The selected applications include the control of insect populations, irradiation of food as a public health measure, radiopasteurization as a new method of processing fresh meats, high radiopasteurization as a new method of processing a variety of foods, irradiation of potatoes to inhibit sprouting, and radiosterilization of foods, surgical items, and medical supplies.

CONTROL OF INSECT POPULATIONS

The following paragraphs describe the use of nuclear radiation in eradicating or controlling various types of insects. Applications of radiation for such purposes are presently limited to the direct use of radiation on insects and to the irradiation of nonedible agricultural products. Therefore, these applications will be described first. If approval is obtained from the Food and Drug Administration for the irradiation of foods, a number of additional applications will be possible.

SCREWWORM *(CALLITROGA HOMINIVORAX)*.

One of the most successful biological uses of nuclear radiations has been the classic case of eradicating the screwworm on the Island of Curacao by releasing male flies made sterile by gamma radiation.[†] The screwworm is the larva of a fly shown in the lower right corner of Fig. 10.1; it is about three times the size of the common housefly.

This fly is a native pest in southwestern United States where stockmen spend millions of dollars annually on range riders to examine and treat infected herds. The fly did not exist in the Southeast until 1933 when it was introduced by infected animals brought from the Southwest. Since then Knipling[1] in 1956 estimated that this pest has cost the livestock industry in Florida, Alabama, Georgia, and the Carolinas at least $240,000,000.

Each female of the species lays about 200 eggs in cuts and other wounds in cattle or game. The eggs hatch to maggots (the larval stage) that destroy additional healthy tissue and produce an enlarged festering wound that attracts more flies. If the animal is not treated it will be killed by the insect. Figure 10.1 also shows a young steer with a severe screwworm infestation that started on the ear as a result of wounds from tick bites.[1] An adult steer can be killed in 10 days if untreated.[2]

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The mature larvae (screwworms) drop to the ground where they pupate in the same way as the larvae of the Mexican fruit fly described in Chap. 8. The pupa state lasts about 10 days and then adult flies emerge to repeat the cycle. The flies may migrate 25 miles or more before laying eggs.



Fig. 10.1—Steer with severe screwworm infestation.[1] Insert: adult female screwworm; normal size is about $\frac{5}{8}$ in.

In attacking the problem of screwworm eradication, it was found in laboratory tests that a dose of 3 krep caused complete sterility in male flies but left them with normal sexual behavior.[1] Experimental mating of a number of flies in cages showed that the female mated only once and, if this mating was with a sterile male, the eggs laid were sterile. Also, the caged matings showed that the percentage of sterile eggs was almost directly proportional to the percentage of sterile males in the cage. This shows the possibility of decreasing the population of this insect if enough sterile males can be released in a given area. Table 10.1 shows the mathematical result of releasing, at four different periods, a number of sterile males equal to twice the original natural population of males. Theoretically such a procedure would eliminate the population.[1]

The Curacao Experiment

To prove the method, it was necessary to attempt eradication in an isolated area of limited size. The Island of Curacao has an area of about 170 square miles and had a screwworm population, per square mile, as high as any area in Florida. A cooperative agreement was reached with the Netherlands Antilles Government to attempt eradication on this island.

Table 10.1—THEORETICAL POPULATION DECLINE WHEN STERILE MALES (SCREWWORMS) ARE ADDED TO A NATURAL POPULATION[1]

Natural population	Sterile males released	Ratio of sterile to fertile males	Population decline (theoretical maximum)
1,000,000	2,000,000	2:1	333,333
333,000	2,000,000	6:1	46,619
46,619	2,000,000	42:1	1,107
1,107	2,000,000	18,000:1	< 1

The experiment began in March 1954. Flies were raised in a laboratory in Florida and, while in the pupa stage, were exposed to 5 to 7.5 krep of gamma radiation. Sterile flies of both sexes emerged and were flown to Curacao and released by air at a weekly rate of 400 sterile males per square mile—three to four times the natural population of males. To test the results, goats were placed in 11 different animal pens located at various places on the island and were examined daily for screwworm eggs. The eggs found were allowed to mature in the laboratory to discover what proportion would produce larvae. The results of the experiment are summarized in Table 10.2.[1]

Table 10.2—SUMMARY OF CURACAO SCREWWORM EXPERIMENT[1]

	Number of egg masses		
Weeks	Fertile	Sterile	Egg masses sterile, %
1	15	34	69
2	17	38	69
3	17	36	68
4	10	37	79
5	7	42	86
6	3	23	88
7	0	10	100
8	0	12	100
9	0	0	

Stockmen on Curacao reported no incidence of cases of screwworm in livestock after the ninth week, and a check 18 months later showed no evidence of the pest on the island.[1]

Eradication of the Screwworm in Southeastern United States

This dramatic demonstration of the eradication of the screwworm on Curacao gave great promise of a means for eradication elsewhere. However, such a project is a tremendous undertaking. The area in Florida in which screwworms normally overwinter covers about 50,000 square miles. Releasing 400 sterile males per square mile per week entailed rearing, sterilizing, and releasing about 40,000,000 flies per week.

In 1960 Jefferson[2] described how the screwworm fly was eradicated in southeastern United States. In 1957 the screwworm infestation of cattle reached a peak of 80 to 85 per cent of all cattle in the state of Florida. Losses from such a high degree of infestation were estimated to $10 million per year in Florida alone and another $10 million in other southeastern States.[2]

To explore the feasibility of using the sterile male technique, a pilot campaign was conducted in 1957 over a 2000 square mile area in Florida. A 70 per cent reduction in screwworm incidence was obtained. To fund a major compaign the State of Florida appropriated $3 million dollars. Additional but lesser sums were allocated by the States of Alabama, Georgia, South Carolina, and Mississippi, and the Federal government appropriated $6.76 million for eradication of the pest.[2]

To undertake the campaign a sterile fly factory was created at Sebring, Florida, in the heart of the Florida ranching area. A World War II hangar for B-17 planes was used to house the fly factory. About $425,000 was required to convert the hangar, $106,000 for Co^{60}, radiation shields, control and handling facilities in the radiation room, $478,000 for other equipment for growing and handling the insects, and the balance was used for operation of the factory.[2]

The fly factory was designed to produce 100 million sterile flies per week. A colony of 3 million fertile flies was required to supply fertile eggs for this production rate. The hangar was divided into 2 floor levels and the second floor was used for hatching eggs and growing the larvae. After the eggs hatched the larvae consumed large quantities of food during their growth. Weekly requirements for food were 40 tons of ground meat, 4500 gal of beef blood, 65 gal of plasma, 35 gal of honey, and 9600 gal of water.[2]

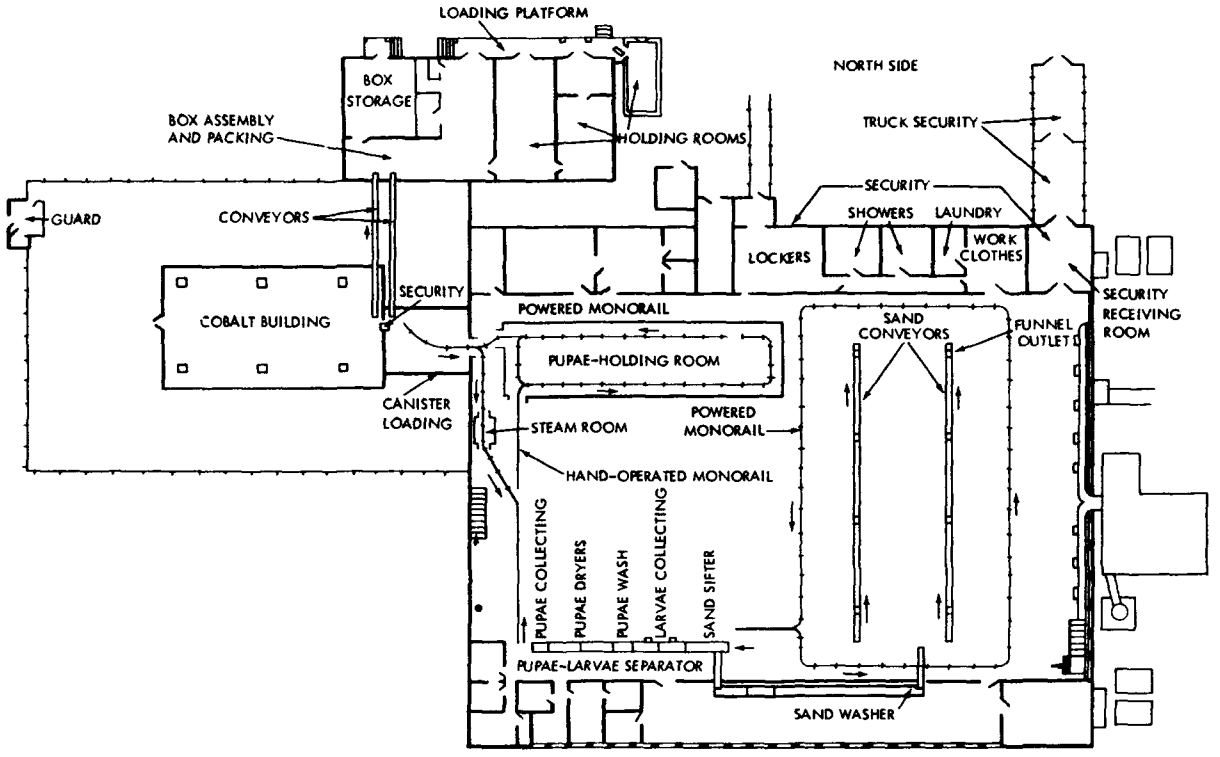


Fig. 10.2—Ground floor of screwworm fly factory.[2]

The mature larvae were dropped by gravity to sand beds in the pupae-holding room shown in the center of Fig. 10.2. This illustration is a plan view of the ground floor where the pupae developed into sterile flies.

In the pupae-holding room the larvae went into pupation in about 8 hr and were held there an additional 5.5 days at 80°F and 95 per cent relative humidity to mature. Two days before adult flies emerged the pupae were exposed to a dose of 8000 r in Co^{60} irradiators located in the cobalt building (see Fig. 10.2). Six irradiators were used with about 600 curies of Co^{60} in each. The sources consisted of cobalt strips 2 by $\frac{1}{8}$ by 13 in. clad with stainless steel. The strips were arranged in a cylindrical pattern with a diameter of about 7 in. The dose rate was about 800 r/min and exposure times were from 12 to 14 min. The sources were contained in the centers of cylindrical lead "pigs" used for shielding. To obtain efficient production of the

sterile pupae automatic operation was used. About 18,000 pupae were loaded into canisters using volume measurement and the canisters were moved by automatic controls into the radiation chambers, held for the required exposure, and then returned to the loading position.[2]

After the sterile flies emerged from the irradiated pupae the flies were loaded into boxes. Planes carrying 1000 boxes of flies were flown over prearranged patterns of terrain at an elevation of about 1500 ft. The boxes were released and opened by automatic ejectors so as to distribute 100 to 800 sterile flies[2] per square mile.

As a result of this campaign the screwworm fly was eliminated in Southeastern United States in 16 months. A total of 3 billion sterile flies were released, and the total cost was about $10 million—half the annual cost caused by this pest at the peak of its population. On Nov. 14, 1959, the fly factory was shut down since its objective had been completed several months ahead of schedule.[2]

In addition to eradicating the screwworm fly, this method might also be used to control the Mediterranean fruit fly, which has been a problem in Hawaii and in Florida, or the Mexican fruit fly described in Chap. 8.

One of the most dangerous insects known is the dreaded tsetse fly, which has made about 4½ million sq miles of Africa virtually uninhabitable. Studies have been made under the East Africa Tsetse Research Organization of Uganda and the London School of Tropical Medicine and Hygiene. Pupae of the tsetse fly were irradiated with gamma radiation at Harwell, England. Although the effort required is great, the sterile male technique offers for the first time the possibility not only of controlling but of eradicating some of man's most dangerous insect enemies.

RAISING THE QUARANTINE ON NONEDIBLE AGRICULTURAL IMPORTS AND EXPORTS

Quarantines are placed on many imported and exported agricultural products to prevent the spread of insect pests, parasites, and pathogenic organisms from one country to another. Both edible and nonedible agricultural products are involved but only the nonedible products can be processed with radiation until the Food and Drug Administration approves the irradiation of foods. As to the nonedible products, Byerly, of the U. S. Department of Agriculture, has stated "If experimental results establish the minimum dosage required to assure disruption of the life cycle of the insects involved, and if the contemplated treatment will assure a minimum rate which will equal or exceed the minimum dosage required for insect control, we (U. S. Department of Agriculture) will be pleased to recommend to regulatory officials that irradiation will be satisfactory from the standpoint of effectiveness against insects."[3]

In many cases the quarantines may be removed by fumigating the product with a poisonous gas so as to destroy the undesirable insect or other organism. Regulatory officials inspect this operation to assure that sufficient poison and time are used to kill all offenders.

To obtain good penetration in bulk products vacuum fumigation may be required. In this process the items to be treated are loaded into a steel chamber designed for vacuum service. When the chamber is filled with the product to be treated the doors are closed and locked pressure tight, and the vacuum pumps exhaust the air from the chamber and pores of bulk products such as cotton, tobacco, and burlap bagging. A controlled amount of poisonous gas such as HCN or methyl bromide then is admitted to the chamber.

Air is admitted after the gas is in the chamber which partially reduces the vacuum and forces the gas into the evacuated pores of the product. The product is kept in the chamber in contact with the gas usually for two or more hours to allow sufficient time for the poison to diffuse throughout the interstices of the product and to kill the organisms. The mixture of air and poisonous gas is then pumped out of the chamber. The chamber may be flushed with air one or more times with evacuation before each air application to wash as much of the gas out of the product as possible.

The process of vacuum fumigation has a number of disadvantages: The chamber must be pressure tight without openings for conveyor mechanisms. As a result the chamber usually must be loaded manually using fork lift trucks which adds an appreciable labor cost to the process. The use of poisonous gas is a hazard which is another disadavantage. Nevertheless, over 50 ports of entry to the United States have vacuum fumigation facilities. A few of the larger

ports such as New York City and New Orleans have facilities capable of handling from 500 to over 1000 bales per day of imported cotton. Reasons for the use of gamma radiation in place of vacuum fumigation are:

(1) Experiments in a number of laboratories in this country and abroad have shown that low dosages of gamma radiation (5,000 to 50,000 rads depending upon species and metamorphic stage) can be 100 per cent effective in breaking the life cycle of a variety of insects and parasites.
(2) Treatment with gamma radiation can be more positive than chemical fumigation because the penetration of radiation is instantaneous whereas a variable diffusion time is involved in chemical fumigation which may result in insufficient treatment in zones of low diffusivity.
(3) In the egg and pupa stages of insects the respiration rates are very low. For this reason and because eggs may be deposited in protected areas, destruction by fumigation is difficult but is readily accomplished by gamma irradiation.
(4) Certain fumigants may leave residual contamination of poisons but gamma irradiation leaves no residues and produces no induced radioactivity in agricultural products.
(5) Gamma radiation chambers can be designed for continuous operation whereas batch operation and considerable labor are required for loading and unloading vacuum fumigators.

STERILIZATION OF ANIMAL PRODUCTS FOR CONTROL OF DISEASE

Animal products such as meats, hides, bristles, bone, and hair may carry pathogenic organisms causing diseases such as anthrax and foot and mouth disease, if the products are obtained from regions where such diseases are not under control. Fresh meat from Argentina is not admitted to the United States for this reason. In the past outbreaks of anthrax in the United States have been traced to shaving brushes containing unsterilized hog bristles imported from China. Now such items must be sterilized before shipment to the United States or at the port of entry. Heat sterilization is effective with most of these products and for this reason there is no quarantine on imported cooked meats. Heat may produce undesirable changes in the products and for this reason chemicals are used instead of heat to sterilize hides and hair. Radiation can be used as a third method of sterilization.

During the summer of 1959, a 150,000-curie Co^{60} source was installed at Dandenong (near Melbourne) Australia to sterilize raw goat hair for the control of anthrax.[4] The hair is used in making carpets at the Westminster Carpet Co. at Dandenong. In the past the hair was sterilized by formaldehyde fumigation. It has been estimated that the use of gamma radiation in place of formaldehyde fumigation will produce a saving of 25 per cent in the cost of sterilization.[4]

In 1960 it was reported[5] that the capacity of the facility was to be increased from 150,000 to 500,000 curies. In addition to sterilization of goat hair the unit will be used to sterilize pharmaceuticals. This is the first commercial gamma radiation facility in the free world (Russia has built a large capacity potato irradicator) and is another milestone in the industrial use of radiation.

CONTROL OF PINK BOLLWORM IN IMPORTED COTTON

As yet radiation, has not been used commercially in the United States for the removal of quarantines. This application appears to be very promising for many products because of the high cost of vacuum fumigation. In case of nonedible products the process could be used now because approval by the Food and Drug Administration is not required and also no problems of off-flavor are involved. This application of radiation appears particularly promising in the case of imported cotton.

Over 500 tons of African cotton are imported daily to the United States. The USA is one of the few cotton-producing countries in the world without a pink bollworm problem. Since imported cotton has this infestation, every bale which enters the country is required by law to be sterilized at the port of entry. Fumigation with cyanide or methyl bromide is the method presently used to comply with the law.

Cyanide fumigation has a moderate cost but is dangerous. Methyl bromide is safer but costs $0.72 per pound and the price is increasing. Both fumigants leave residual contamination of poisons which introduces additional hazards. Vacuum fumigation of cotton is a batch process and about 8 hours is required per batch.

Two criteria must be met in a process using radiation to break the life cycle of the pink bollworms found in cotton bales: (1) The dose must be high enough to kill all the bollworms and (2) the dose must be low enough to assure that no damage is done to the cotton itself.

Specific data on the effect of gamma radiation on the pink bollworm are not available at this time. However, extensive work is now being conducted at the Department of Agriculture's Brownsville Laboratories. Based on results obtained with other insects (see Chap. 8) it is believed that a dose of 20,000 rad would break the life cycle of the bollworm.

The maximum permissible radiation dose for the cotton was determined using the data of Pan, Proctor, and Goldblith at MIT,[6] and checks well with the work by Teszler and Rutherford at North Carolina State.[7] They showed that the quality of the cotton was actually improved by dose rates of 100,000 rep or less. Some of their data are reproduced here in Table 10.3.

Table 10.3—EFFECT OF GAMMA RADIATION ON PHYSICAL PROPERTIES OF COTTON[6]

Dose, Mrep	Tensile strength, g	Toughness, g-in.	Natural strength curve, %
Control	3905	966	100
0.05	4040	991	103
0.10	3990	981	102
1.00	3480	746	89
2.00	3040		77

The data of Table 10.3 indicate a 3 per cent strength increase at a dose of 50,000 rep, a 2 per cent increase at 100,000 rep, and a reduction of strength at higher dosages. The initial increase in strength is caused by the release of points of residual internal stress. The radiation has the same effect on cotton that annealing does to a stressed metal.

Proctor et al., also found that gamma radiation sterilized the cotton, and showed that a "general increase in mildew resistance was noticed in those samples irradiated over those samples unirradiated."[6]

Thus, a gamma dose between 20,000 and 100,000 rep is believed to result in: (1) the breaking of the life cycle of the pink bollworm, (2) an increase in cotton fibre strength up to 3 per cent, and (3) an increase in the mildew resistance of the cotton.

Design for a Cotton-Bale Irradiator

Fleming and Walsh[8] have considered a possible design for a gamma irradiator to process bales of imported cotton. Figure 10.3 shows a plan view of their proposed irradiation chamber.

In their suggested design bales of cotton are loaded on a mechanical conveyor at A with the long sides of the bales parallel to the conveyor flow. The bales are moved to B where they are switched into two streams, one on each side of the source. When a bale arrives at point C or C′, a ram pushes the bale into the irradiation position, D. When the sides next to the source have received an adequate dose of radiation, the bale is mechanically turned over and a new side pushed into position. After all four sides have been irradiated, the bale is moved by the conveyor to the unloading point E. The two ends of the bale are irradiated by source rods which extend beyond the bale.[8]

The design gives a high efficiency in utilization of gamma radiation. End losses are reduced by placing bales on both sides of the source, each only 3 in. away. The source rods are positioned so that the entire surface of the bale receives nearly the same dosage.

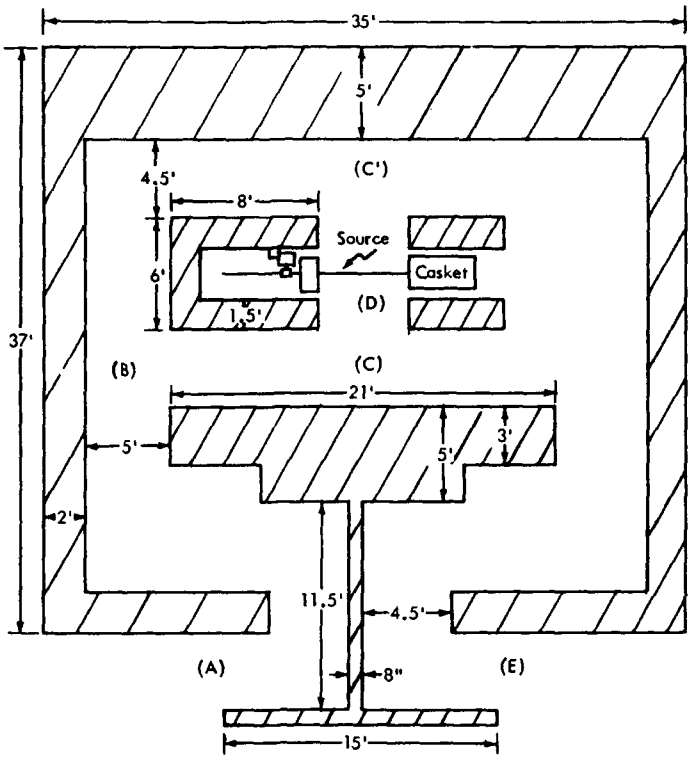


Fig. 10.3—Plan view of proposed gamma irradiator for imported cotton.[8]

Table 10.4—ESTIMATE OF COSTS FOR FUMIGATION OF COTTON (150 BALES PER DAY, 300 DAYS PER YEAR)[8]

Wages and salaries	$21,480 per year
Methyl bromide costs	64,800
Labor overhead	3,220
Operation and maintenance	2,000
Total	$91,500 per year

Table 10.5—ESTIMATE OF COSTS[8] FOR IRRADIATION OF COTTON (150 BALES PER DAY, 300 DAYS PER YEAR)

Source	$168,784
Radiation chamber and accessories	48,760
Source casket	5,000
Shipping expense	500
Total	$224,044
Operating costs	
Plant amortization	$ 53,000
Labor costs	16,250
Plant overhead and maintenance	2,000
	$ 71,250
Less: Amortized source salvage	8,750
Total	$ 62,500 per year
Cost per bale $1.39	
Cost per 100 lb $0.28	

The source consists of 3-ft-long Co^{60} rods of equal strength arranged to form a plane. The bales are placed only 3 in. from the source, parallel to the source plane so as to approach as close to the source as possible and yet limit the surface dose to less than 100,000 rep while delivering 20,000 rep at the required depth in the cotton.

The pink bollworm appears in the cotton bales only as larvae and location is limited to the outer inch or two around the bale. As a safety factor, the design is based upon delivering a 20,000-rep dose to a depth of 3 in. in the bale. A source activity of approximately 100,000 curies was calculated as adequate for the irradiation of 150 bales per day.[8]

For flexibility of operation, the source was designed so that the dose at the ends of the bale is higher than that at the center. In this way the facility can accommodate bales of slightly larger dimensions.

Fumigation Costs Compared to Irradiation Costs for Cotton Bales

Methyl bromide is the most popular fumigant for cotton. Hydrogen cyanide fumigation is cheaper but the additional dangers involved make it undesirable. Fleming and Walsh[8] assumed a labor force of 4 men plus a supervisor and estimated the operating costs for a vacuum fumigation facility using methyl bromide. The Department of Agriculture has recommended a dose of 4 lb of methyl bromide (at a cost of $0.72 per pound) per 1000 lb of cotton. Table 10.4 is a summary of the estimated fumigation costs.

This estimated cost of $91,500 per year corresponds to a fumigation cost of $0.41 per 100 lb of cotton. This includes only operating costs and does not consider amortization of the fumigation plant investments.

The cost of processing cotton bales by radiation (both with Co^{60} and with Cs^{137}) were estimated by Fleming and Walsh[8] and compared to the operating costs associated with fumigation, using a basis of 150 bales per day and 300 operating days per year. The interest rate used in amortizing the capital cost was 6 per cent. The Co^{60} facility investment costs were written off in five years (about one half-life). Estimates of capital costs and operating costs for the cotton bale irradiator are given in Table 10.5.

A comparison of Cs^{137} and Co^{60} for this cotton irradiation facility was made on the basis of pay-off period, using the $91,500 yearly operating cost for fumigation. It was found that the Co^{60} facility would pay for itself in 3.7 years whereas the Cs^{137} facility would require 6.1 years as a pay-off period. Thus at present isotope costs, Co^{60} is the most economical source material.

A Co^{60} radiation facility, used for 5 years without source replacement, can process cotton bales at $0.28/100 lb, while the present operating cost for fumigation is $0.41.

Fleming and Walsh[8] concluded that a radiation facility for imported cotton bales would be superior to the present fumigation methods. The estimated per-bale cost is over 30 per cent less with radiation than with fumigation. Also, fumigation is slower and inherently more dangerous, whereas the radiation can be safe and can slightly improve the quality of the cotton.[8]

IRRADIATION OF TOBACCO AND OTHER NONEDIBLE PRODUCTS

Raw tobacco is purchased from growers and brokers and then usually is stored for periods up to 3 years for curing. During the curing period a mild fermentation occurs which mellows the tobacco and improves its quality. A number of insects may attack tobacco during the curing period but the two major pests are the tobacco moth[9,11] and the cigarette beetle.[10,11]

Since colonial days the tobacco industry has used wooden hogsheads for handling and curing tobacco. The standard hogshead in use today is 4 ft in diameter, 4 ft long, and holds about 1000 lb of compacted tobacco leaves. The adult stages of the insect pests mentioned previously enter the hogsheads through the crevices between the wooden staves and deposit eggs that develop into larvae that infest the tobacco. Before the hogsheads are placed in storage and before they are exported vacuum fumigation is used to kill the insects. During storage in tobacco warehouse the outside of the hogsheads are sprayed with insecticides at periodic time intervals to prevent insect infestation while the tobacco is being cured.

Boxed cigars may be kept in retail stores for some time before sale and use. Vacuum fumigation of boxed cigars is often used to kill any insect eggs that might have escaped previous treatments.

There is no U. S. Law requiring vacuum fumigation of imported or exported tobacco or tobacco products as in the case of imported African cotton. However, as a matter of practice and quality control essentially all exported tobacco in hogsheads is vacuum fumigated. One of the larger vacuum fumigation facilities specializes in treating exported tobacco and can handle over 500 hogsheads per day. The cost of fumigation to the operator (using HCN) is about $1.50 per hogshead. About half of this cost is that of labor required for loading and unloading the batch-operated vacuum fumigators.

As in the case of imported cotton, it is believed that gamma irradiation could be used to advantage in place of vacuum fumigation. Doses of over 1 Mrad have little or no effect on the quality of smoking tobacco. Thus, the much smaller dose required to control insect infestation would not lower the quality of the product. Insect control could be more positive and costs could be less.

Doses higher than required for insect control might have application also. Sometimes foreign microorganisms on the tobacco interfere with the desired fermentation and cure. A radiopasteurization dose could be used followed by inocculation with the desired culture so as to produce better control of the cure.

Gamma radiation will destroy some carthogenic compounds. The possibility exists that a large dose of radiation might be used to destroy compounds in tobacco that may produce carcinogens in tobacco smoke. This possible application may prove to be unfeasible but might be worthy of investigation.

In addition to cotton and tobacco other nonedible agricultural products are fumigated for control of insects pests and parasites. Broomcorn imported for use in making the common broom must be fumigated for control of the corn borer. Scrap burlap bags that have been used abroad to ship potatoes and other root crops are imported for use in making low cost roofing felts and cheap carpet backing. These bags must be treated to destroy nematodes that have become a serious problem in overseas areas. In many parts of Europe the nematode infestation of the soil is such a problem that potatoes can be grown on the soil only once in 5 years. Rotation with other crops not so favorable to nematode growth is used between potato-growing years. The nematode problem exists in some parts of the United States. Long Island, N. Y., was once a well-known potato producing area but the golden nematode established itself in this area and has ruined the soil for potato production. Fassuliotis and Sparrow[12] have reported that a radiation dose of 20,000 rads will break the cycle of the golden nematode. If radiation cost could be drastically reduced it might be feasible to treat the top 12 in. of soil.

Ports of entry having a gamma irradiator might use it for treating imported or exported tobacco, imported broomcorn and imported burlap bagging. If approval is obtained from the Food and Drug Administration for irradiation of foods, a wide variety of fruits (especially tropical fruits) and vegetables might be treated in the same facilities.

INSECT CONTROL IN EDIBLE PRODUCTS

Many more quarantines are placed on edible than on nonedible agricultural products. Soft skinned fresh fruits grown in areas having the Mediterranean fruit fly, the Mexican fruit fly, the melon fly, and a host of other pests are barred from importation into the United States unless vacuum fumigated or steam treated so as to destroy the culprits. The areas involved include Puerto Rico, Hawaii, Mexico, southern Europe, Africa, South America and southern Asia or just about every area growing tropical fruits outside the United States.

Pineapples have such a tough skin that the insect pests mentioned do not attack the fruit which fortunately removes this valuable fresh fruit from quarantine. However, every mango (one of the most delicious of tropical fruits) imported from Puerto Rico must be vacuum fumigated. Mangoes that sell retail for 3 cents each in Puerto Rico may bring 10 times that price in New York City. Part of the price increase is due to the cost of vacuum fumigation. Here again radiation might be used to advantage.

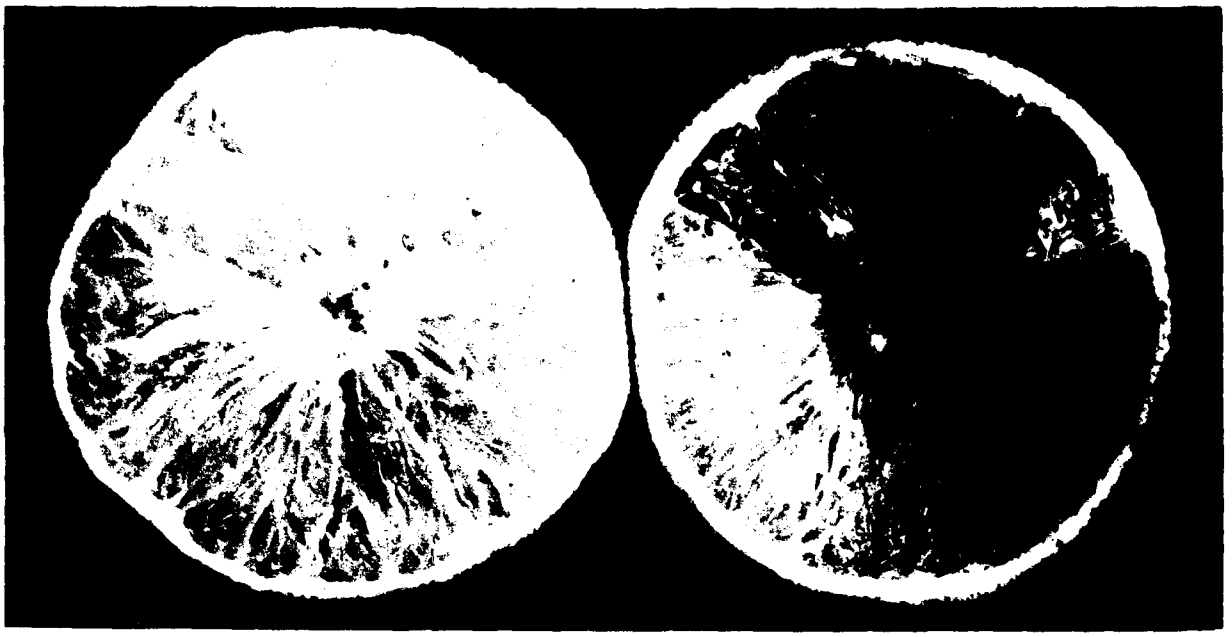


Fig. 10.4—Right: Half section of nonirradiated grapefruit showing secondary damage caused by larvae of Mexican fruit fly. Left: Half section of undamaged irradiated grapefruit.[13]

As mentioned previously (see Chap. 8), only a low dose of radiation (5,000 to 50,000 rads) is required to break the life cycle of most insects and parasites infesting agricultural products. This subradiopasteurizating dose produces no flavor changes and usually only slight texture or color changes in most fruits. Almost no detectable chemical changes are produced except for a possible reduction in ascorbic acid content in some fruits and vegetables. Thus, this very mild irradiation treatment should be more acceptable to the Food and Drug Administration than the much higher (100 to 1000 times) dose necessary for radiosterilization.

CONTROL OF FRUIT FLIES

The possibility of eradicating the Mediterranean fruit fly in Florida by the sterile male technique has been mentioned. Such a program would, however, be expensive and would require a great effort of organization. In the meantime, citrus fruit shipped from areas infested with the fly must be steamed to kill the eggs and prevent spread of the area of infestation. Steam treatment lowers the quality of the fruit.

In the case of the Mexican fruit fly, the use of the sterile male technique would not be successful in Texas unless it was also used in Mexico because the fly migrates from Mexico to Texas. This fly is almost identical to the Mediterranean fruit fly in appearance and habits. An adult Mexican fruit fly was shown in Chap. 8 (see Fig. 8.24).

Figure 10.4 shows a half section of an irradiated grapefruit on the left and a half section of a nonirradiated control fruit on the right. Note the secondary damage in the control fruit resulting from destruction caused by the larvae of the Mexican fruit fly.[13]

DESIGN FOR A MOBILE PRODUCE IRRADIATOR

Brownell et al.[14] have proposed the use of a railway mobile gamma-radiation facility that could be moved to various citrus shipping areas in Florida and in Texas to sterilize fruit shipped from infested areas. The low dosage required (10 krep or less) produces no undesirable effects in the fruit (See section in Chap. 8, Effect of Ionizing Radiation on Some Insects and Parasites). The proposed railway gamma-irradiation facility is shown in perspective in Fig. 10.5.

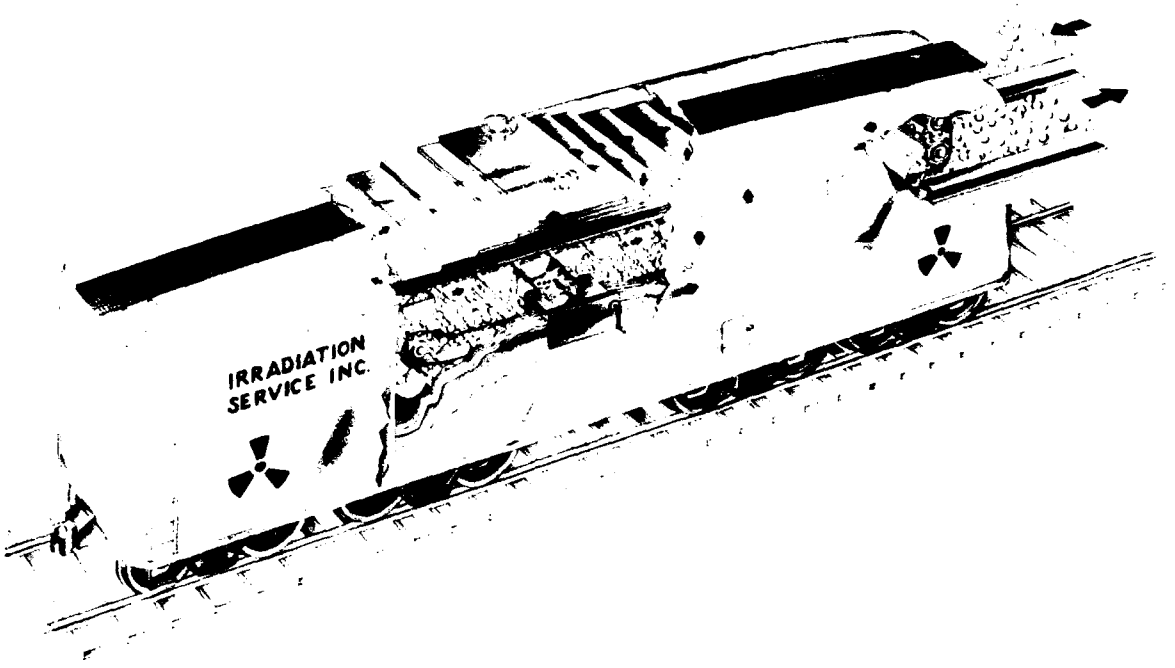


Fig. 10.5—Perspective view of railway mobile irradiation facility.[14]

The use of a mobile unit can be more economical than a built-in irradiation facility when the material to be irradiated is seasonal in nature. Rather than remaining idle for many months between seasons, a mobile facility might be moved to Idaho or Maine to irradiate potatoes; to Washington, Oregon, or Michigan to irradiate apples; to Kansas or Minnesota to irradiate grain.

CONTROL OF INSECTS IN FLOUR, GRAIN, AND MEAL

Insect infestation results in a tremendous annual loss of stored flour, grain, and cereal products. The granary weevil *(Sitophilus granarius)* is probably the most damaging insect to grain; and the confused flour beetle *(Tribolium confusum)* is the most damaging to flour and prepared cereal products.

The bean weevil *(Acanthoscelides obtectus)* the rice weevil *(Sitophilus oryzae)* and the lesser grain borer *(Rhizopertha dominica)* as well as many other insects[15] listed in Table 10.6 also contribute to the estimated loss of $100,000 annually in the United States of flour, grains, and cereal products as a result of infestation.[16]

The mobile unit shown in the figure is designed to use spent MTR fuel elements. The shielding material consists of multiple layers of hot-rolled mild-steel plate 2 in. thick. The same steel shield is used to carry the carload, eliminating the structural undercarriage in the car. The car shown is estimated to have a total weight of 173 tons. Ten fuel elements cooled for 40 days are used and are kept in two water-cooled lead pigs weighing about 4 tons each and located on either side of the car.

The total cost of the facility is estimated to be $93,400 which includes the cost of railway car. The source is rented at $240 per element per year. The annual operating costs are estimated at $114,300.

The unit if used for food irradiation can process 22,500 tons per year using a dose of 20,000 rep and operating 4160 hours per year. This amounts to $5.10 per ton of material processed.

The confused flour beetle is the insect most frequently found in flour after it leaves the mill. The upper half of Fig. 10.6 shows the appearance of the beetle on an enlarged scale. The adult beetle normally is about $\frac{1}{7}$ in. long.[15] Usually both the insects and the eggs can be screened out of the flour but the eggs are more difficult to remove, particularly from coarser

Table 10.6—SOME INSECT PESTS THAT INFEST FLOUR, GRAIN, AND CEREAL PRODUCTS[16]

Grain weevils
- Granary weevil
- Rice or black weevil
- Broad-nosed grain weevil
- Coffee bean weevil

Grain borers
- Lesser grain borer
- Larger grain borer

Grain moths
- Angoumois grain moth
- European grain moth
- Pink cornworm
- Rice moth

Flour moths
- Indian-meal moth
- Mediterranean flour moth
- Meal moth

Grain and flour beetles
- Cadelle
- Saw-toothed grain beetle
- Square-necked grain beetle
- Foreign grain beetle
- Mexican grain beetle
- Siamese grain beetle
- Flat grain beetle
- Rusty grain beetle
- Confused flour beetle
- Red flour beetle
- Black flour beetle
- Long-headed flour beetle
- Broad-horned flour beetle
- Slender-horned flour beetle
- Small-eyed flour beetle
- Depressed black flour beetle

Eggs of flour and grain insects

Mealworms
- Yellow mealworm
- Dark mealworm
- Lesser mealworm
- Black fungus beetle
- Red-horned grain beetle

Dermestid beetles
- Black carpet beetle
- Larger cabinet beetle
- Varied carpet beetle

Spider beetles
- Hairy spider beetle
- White-marked spider beetle
- Brown spider beetle
- Other spider beetles

Miscellaneous beetles
- Two-banded fungus beetle
- Hairy fungus beetle
- Corn sap beetle
- Cigarette beetle
- Drug-store beetle
- Catorama beetle

Others
- Booklice, or psocids
- Silverfish
- Cockroaches
- Flour or grain mites

products such as corn meal. Under favorable conditions, these eggs may develop into adult beetles in 30 days. The female beetle may live for a year or more, producing 400 to 500 eggs during that period. The short time required to develop from egg to adult may result in the production of many generations of insects and complete infestation of stored flour.

The lower half of Fig. 10.6 is a photograph of eggs of the confused flour beetle and of some other flour and grain insects resting on a piece of cloth. The average layman seldom sees these eggs because of their small size (some as small as $\frac{1}{150}$ of an inch). The eggs are white in color and are coated with an adhesive substance that causes particles of flour to stick to them.

Figure 10.7 shows wheat artificially infested with the granary weevil. The left photograph shows wheat given a 100-krep dose of electrons and stored for 43 days at room temperature. The right photograph shows the damaged untreated wheat.[17]

The great advantage of gamma radiation over chemical fumigation is the effect of gamma radiation on the eggs. Chemical fumigation may be quite effective on larval or adult insects, but is not very effective on insect eggs. Thus, unless the fumigation is repeated to destroy insects hatched from eggs subsequent to the initial fumigation, freedom from insect infestation is not assured. Gamma radiation sterilizes eggs as well as adult insects.

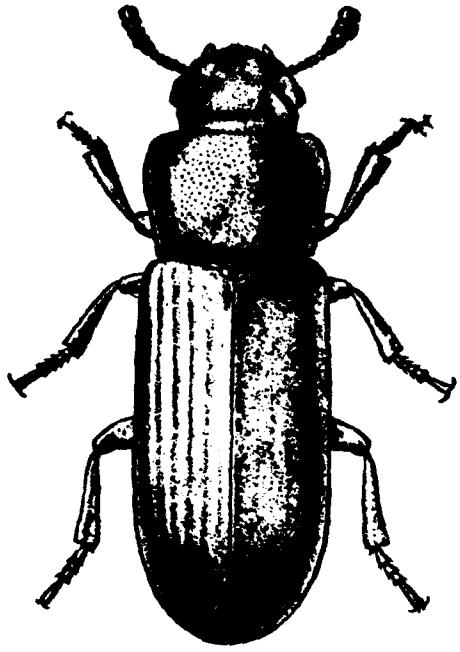




Fig. 10.6—Upper: The confused flour beetle, about $^1/_7$ in. long. Lower: Eggs (about 0.015 in. in diameter) of flour and grain insects. a. Broadhorned flour beetle. b. Cadelle. c. Mediterranean flour moth. d. Confused flour beetle.

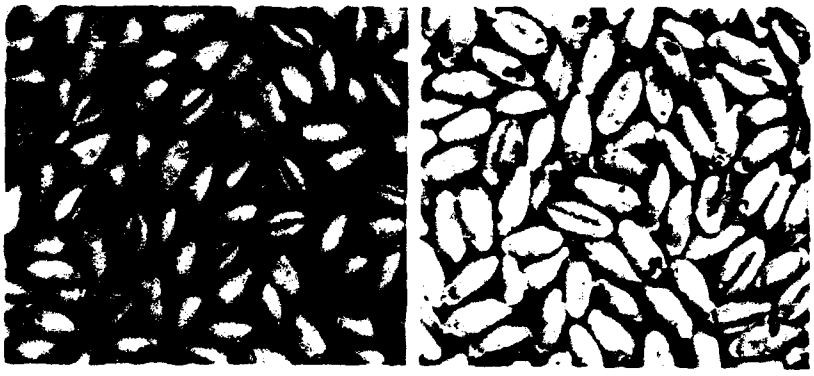


Fig. 10.7 — Wheat artificially infested with granary weevil. Left: Given 10-krep dose of electrons and stored 43 days at room temperature. Right: Untreated. (Courtesy of Michigan State University)[17]

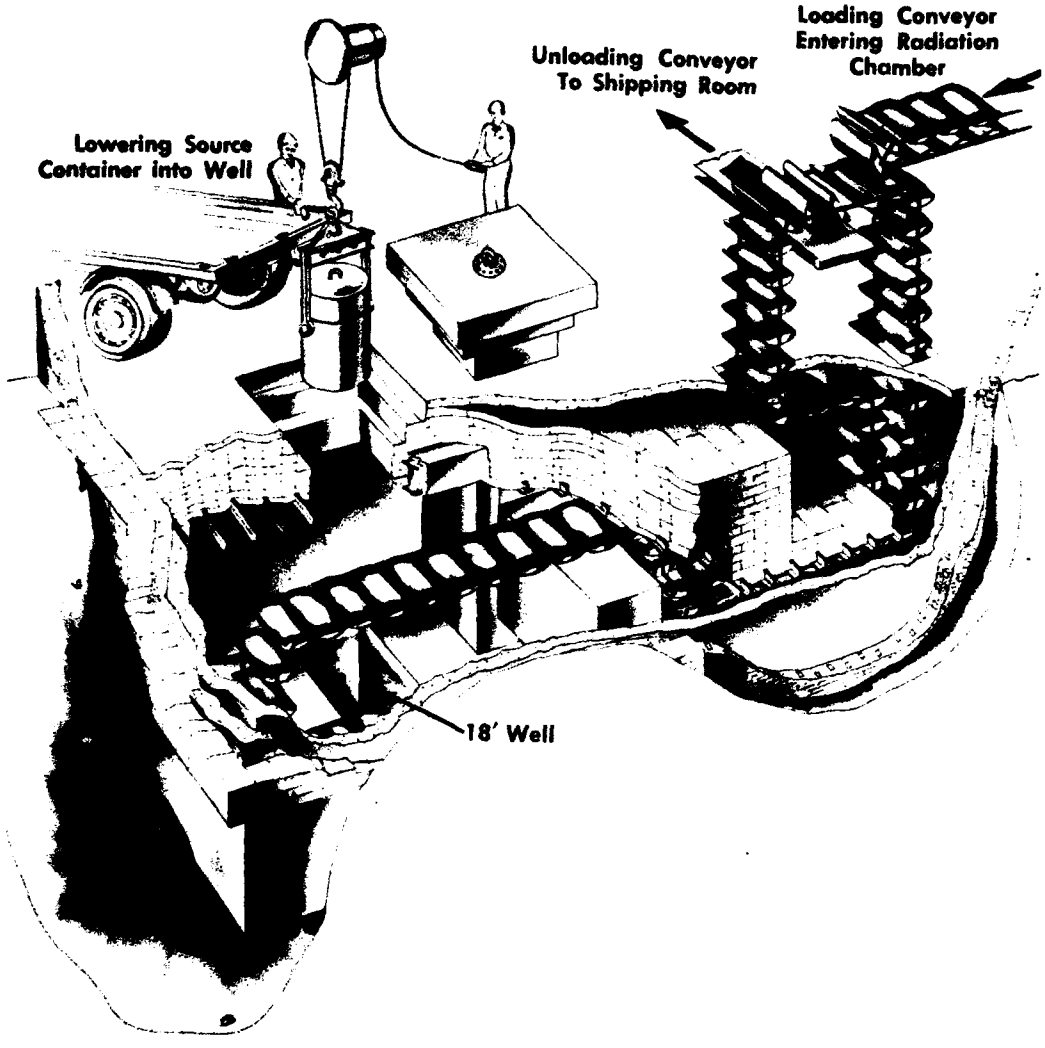


Fig. 10.8 — Perspective view of flour irradiation facility.[18–20]

IRRADIATOR FOR 100-LB BAGS OF GRAIN, FLOUR, OR MEAL

A preliminary design was presented by Brownell et al.[18–20] for an irradiation facility for treating 100-lb bags of grain, flour, or meal to control insect infestation. Figure 10.8 shows a perspective view of this facility.

This irradiator was designed to use spent fuel elements for gamma radiation. The used fuel elements are stored under water in an 18-ft well. A bucket conveyor is used to transport material to be irradiated through the radiation chamber.

Experiments[21–22] have shown that a dose of 10 krep will prevent reproduction of the confused flour beetle, but the design was based on a dose of 25 krep to provide a safety factor. Experience should lead to the reduction of this dosage possibly to 10 krep or less with a corresponding decrease in irradiation cost.

In previous designs by Brownell et al., the initial investment for the irradiation facility was a major item in the consideration of economic feasibility. In the design presented in Fig. 10.8, an attempt was made to minimize capital investment by a number of economies such as locating the radiation facility in the corner of the basement floor of an existing building. Existing walls and exterior earth are used for part of the shielding, and a large part of the concrete used for shielding inside the building consists of commercial 4- by 8- by 16-in. concrete building slabs stacked without mortar for further economy.

A labyrinthine entrance was specified in preliminary designs by Brownell et al., for the irradiation facilities for pork, prepackaged meats, and potatoes. However, in the design for grain, flour, or meal, economy was achieved by using a concrete stepped plug in the roof. This opening is designed as the point of ingress and egress both for personnel and for the shipping container used for the radioactive source consisting of reactor spent-fuel elements.

Although the design was prepared primarily for irradiating flour in 100-lb bags, similiar-sized bags of meal, mash, and a variety of other cereal products might be irradiated in such a facility. The conveyor buckets could also handle bulk grains or bulk flour, although simpler conveyor systems would suffice for the handling of bulk products. The estimated $38,320 for the radiation facility, including conveyor, crane, etc., is believed to be about the minimum cost for a commercial radiation facility of this design.[18,19]

GRAVITY-FLOW GRAIN IRRADIATOR

A design for a gravity-flow irradiator has been suggested by the Curtiss-Wright Corporation[23] for processing grain with gamma radiation. The original design was based on the use of Co^{60} rods but may be modified for use of "Zirnob" ($Zr^{95}-Nb^{95}$). The design is shown in Fig. 10.9

The main feature of the design is the arrangement of Co^{60} rods in a matrix with grain flowing continuously by gravity within the matrix. This arrangement provides a more uniform dose to the grain than that which would be provided by a single Co^{60} rod and grain flowing around it, or a circle of rods and a single cylinder of grain. Also, this system permits maximum absorption of the gamma radiation in the grain. The control of the dosage is obtained by an adjustable orifice at the bottom of the radiation chamber.[23] The estimated cost of irradiation is $0.01 per bushel for 24 hr a day operation of the described equipment based on a price of $2 per curie for Co^{60}.

To reduce the price of irradiation $Zr^{95}-Nb^{95}$ (equilibrium mixture, effective half-life 65 days) might be used. Aebersold has discussed the advantages of this low-capital cost gamma source.[24] Seasonal demands, at the time of crop harvesting, for such radiation facility, may not permit efficient use of Co^{60} (half-life 5 years). If zirconium–niobium is used, it can be written off after a few months use. This low-cost gamma source can become available in large amounts from the waste of production reactors as a result of a fission yield of 6.4 per cent and easy separation from fission product wastes.

Fanhoe et al.[25,26] studied production lines in representative American food handling and processing plants to determine the optimal location for the iradiation source. Figure 10.11 shows their conclusions for a grain irradiator. This study was based on the assumption that a minimum of plant space, capital investment, or product price increase should be available or

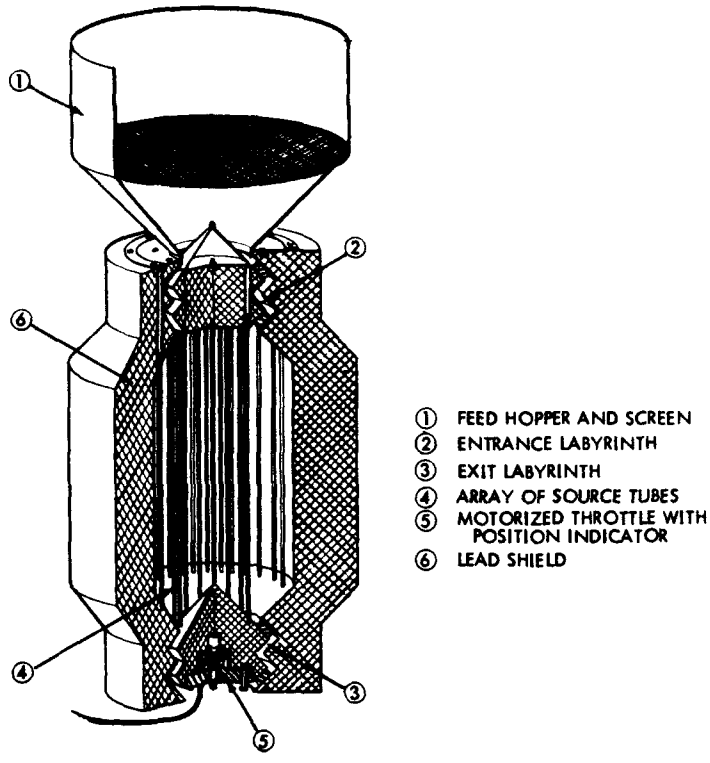


Fig. 10.9—Sectional view of gravity-flow irradiator: 15,000-curie Co^{60} gamma ray source, capacity 120 bu/hr, dia., 42 in.; wt, 14 tons. (Courtesy Curtiss-Wright Corp.)[23]

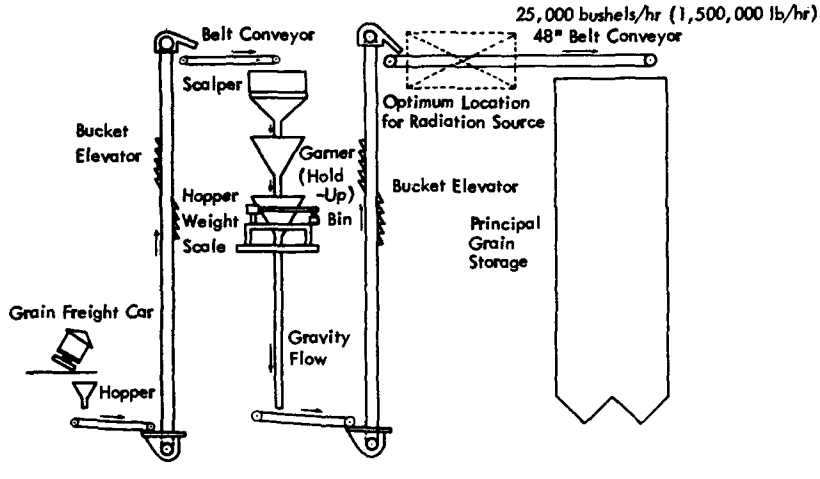


Fig. 10.10—Flow diagram for grain elevator operations.[25,67]

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Table 10.7—HUMAN DISEASES ACQUIRED THROUGH FOOD[31]

Causative organism	Form in which ingested by man	Disease produced in man	Natural host	Incidence and geographic distribution
Trichinella spiralis	Cysts in raw or undercooked pork	Trichinosis	Hog (rat, man, carnivorous animals)	Cosmopolitan. In 16% human autopsies in U. S. (p. 370).* In U. S., 1.5% of all hogs are infected (p. 370)*; 27.8 million people†
Clonorchis sinensis	Metacercariae in freshwater fish	Clonorchiasis (Chinese liver fluke)	Man, dog, cat, hog (snails and fish are intermediate hosts)	Japan, China, Formosa, Korea, French Indo-China. "Disease is equally prevalent in poor and the prosperous, in illiterate as well as the educated," because of custom of eating raw fish (p. 650)*; 19 million people†
Ascaris lumbricoides	Ova in raw vegetables (principal mode of transmission is infected soil)	Ascariasis (intestinal obstruction in children, pneumonia)	Man, hog	Cosmopolitan. In surveys of southern U. S., 1 to 60% in children (p. 447).* In Shantung, China, 23 to 44% in children; 644.4 million people†
Fasciolopsis buski	Metacercariae on raw fruit or edible water plants	Fasciolopsiasis (intestinal fluke)	Man, hog (snail is intermediate host)	Central and South China 10 million people†
Taenia saginata	Cysticerci in meat	Beef tapeworm	Man (cattle are intermediate hosts)	Cosmopolitan. 0.38% of persons infected in Peiping, China. Incidence in cattle: U. S. 0.37%, Holland 3.3 to 5.6%, S. Africa 5% (p. 578)*; 38.9 million people†

Taenia solium	Cysticerci in pork	Pork tapeworm; cysticercosis in tissues	Man (hog is intermediate host)	Cosmopolitan. Incidence of tapeworm in man: Russia 0.2 to 1.5%; incidence of cysticercosis in man: 2.9% Mexico (p. 575).* Incidence in hogs: S. Africa 25%, Mexico 4.3%; 2.5 million people†
Diphyllo bothrium latum	Plerocercoid larva in fresh-water fish	Fish Tapeworm anemia resembling pernicious anemia	Man, dog, cat (fish is intermediate host)	N. America, Europe, Tropical Africa, N. Asia, S. Asia. In Finnish Army 14.5% (p. 548).* About 20% of pickerel and pike in Lake Winnipeg and Lake Manitoba are infected (p.549)*; 10.4 million people†
Paragonimus westermani	Crabs or crayfish containing metacercariae	Paragonomiasis (Oriental lung fluke)	Man, dog, cat, many other animals (snail and crab or crayfish are intermediate hosts)	Japan, Korea, Manchuria, Formosa, China, other countries of Orient; Africa, S. America. Adult parasites have been found in mammals in Canada and many states of U. S.; 3.2 million people†
Fasciola hepatica	Encysted cercaria in water cress, lettuce	Fascioliasis (sheep liver fluke)	Man, sheep (snails are intermediate hosts)	Cosmopolitan. In sheep-raising areas of world. Incidence in Estonia 57% in sheep, 72% in cattle; in Transcaucasia, 48% in sheep; in southern Texas, 21 to 32% of rabbits (reservoir hosts for new infections) (p. 629)*; 100,000 people†

*Page numbers have reference to Belding, D. L: Textbook of Clinical Parasitology, 2nd Ed., New York: Appleton-Century-Croft, 1139 pp.

†Stoll, N. R.: "This Wormy World," The Journal of Parasitology, 1947, Vol. 33, p. 15.

tolerated. However, because of the heavy shielding required, a better location for the irradiator might be at or below grade level directly beneath the hopper weight scale (see Fig. 10.10). Placing the irradiator below grade level would permit the use of earth for some of the shielding, and gravity flow could be used from the hopper weight scale through the irradiator. To prevent reinfestation of the treated grain the conveyor, elevator, and storage bin should be constructed so as to be "insect proof" after receiving irradiated grain.

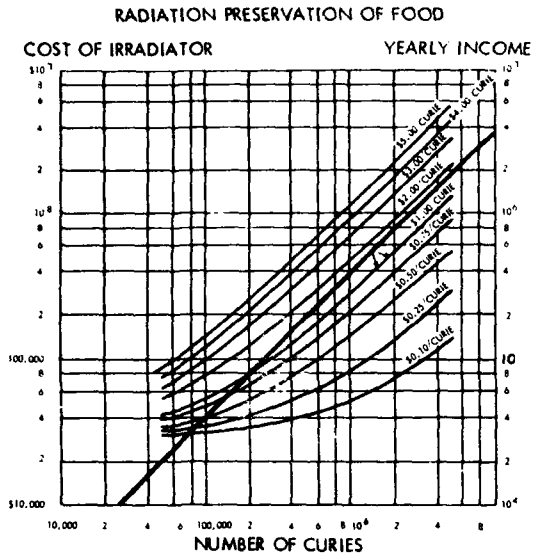


Fig. 10.11 — Economics of grain irradiation[26,46] with Co^{60}.

The economics of irradiation of grain with Co^{60} have been analyzed by Chamberlain.[46] Assuming that the equipment operates 100 days per year and that the irradiation charge is 1 cent per bushel he reported potential income as shown by the heavy straight line in Fig. 10.11. The other lines represent the cost of the irradiator. The vertical distance between the two lines gives the potential annual profit.

IRRADIATION OF FOODS AS A PUBLIC HEALTH MEASURE

The success of heat pasteurization of milk in minimizing the transmission of disease from dairy cows to human beings is well established. In addition to the purely commercial reasons for gamma ray pasteurization of fresh meats and seafoods, such a process has merit from the public health standpoint.

Even though most meats and seafoods are usually fully cooked prior to ingestion, pasteurizing these goods by radiation could be an important additional safety measure for public health. The high incidence of trichina cysts found during autopsies indicates that pork is not always cooked sufficiently to prevent transmitting infection.[27]

Hull lists 91 diseases that are transmitted from animals to man.[28] Of these, 35 are considered as being possibly transmitted to man by the handling of meat or seafoods or by their ingestion when not fully cooked. These diseases are of varying public health importance in different parts of the world. Some of the important diseases of man acquired through food were described by Gomberg et al.[31] and are listed in Table 10.7.

Beef is a meat that is quite often eaten in the rare or undercooked state. Therefore, some consideration should be given to diseases that can be transmitted by ingestion of undercooked beef. Cattle are subject to a wide variety of diseases that may be transmitted to man. These include diseases such as tuberculosis, anthrax, brucellosis, salmonellosis, and listerellosis; infections produced by animal parasites such as tapeworm; and infections produced by fungi and by protozoa.

TRICHINOSIS

Generally, trichinae will have been killed by cooking. Too often, however, live trichinae will be ingested and it is no wonder, therefore, that in surveys of autopsies in the United States

of over 11,000 human bodies, an average of 16 per cent have been found to harbor trichinae. This figure is low; for when more thorough examinations are made, the incidence at autopsy has been found to be as high as 36 per cent. It would be a conservative estimate to state that approximately 25 per cent of the general population of this country develop trichinous infection during their life-time. Fortunately the infection is subclinical in the majority of cases. It is estimated that at least 5 per cent of those who become infected show signs of illness but that only about 1 per cent of all who are infected become sufficiently ill to confine them to bed with clinical trichinosis. The mortality rate from clinical trichinosis (patients confined to bed) in the United States is 5 to 6 per cent. It has been said that "trichinosis is one of the major health problems in the United States" and that "the United States has the greatest problem of trichinosis of any country in the world."[29,30]

Experimental tests by Gomberg and Gould with gamma radiation from Co^{60} and from reactor fuel slugs have shown that a low dose of gamma radiation, about 15 to 20 krep, is ample to break the cycle of trichinosis.[31] Results have recently been supported by Taylor and Parfitt.[32]

Design for a Pork Irradiator

A gamma irradiation facility was designed to irradiate whole, slaughtered hogs for the control of trichinosis.[33-35] Figure 10.12 shows a perspective view of the preliminary design for the commercial irradiation facility. Figure 10.13 is a plan view of the same facility.[33-35]

The source consists of a plaque 6 ft by 5 ft by 0.4 in., and uses 1.5 megacuries of Cs^{137}. Concrete shielding is used for the radiation chamber and the source is lowered in a water well with a liner fabricated of stainless steel and with dimensions of 9 ft by $4\frac{1}{2}$ ft by 24 ft.

A dose of 30,000 rep can be delivered to the surface of 2000 hogs per day. The cost is estimated to be 23 cents per hog, assuming a cost of 30 cents per curie for Cs^{137}.

BEEF TAPEWORM

An infection caused by eating undercooked beef is beef tapeworm. Hull states that "beef tapeworm infection is widely distributed in this country and elsewhere, and, furthermore shows no evidence of diminishing in the human population."[28] The larvae of the beef tapeworm develop in the striated muscle of the steer (and larvae of the pork tapeworm in pork) after the animal (cattle or hog, respectively) has ingested eggs of the specific tapeworm. Man becomes infected by eating the undercooked flesh containing the larvae.

Taylor and Parfitt[32] have recently reported on the use of radiation to control parasites transmitted to man through fresh meat. They comment that "The continued presence of parasitic worms even among the elite of Western civilization shows that the hygiene of our modern cities and homes has reached only a relatively advanced postition. Three parasites are transmitted through meat, two tapeworms, *Cysticercus bovis* and *Echinococcus granulosus*, and a small roundworm, *Trichinella spiralis*, and are still of frequent occurrence in Great Britain, in common with other developed countries." Of the three *C. bovis* is most common in England and *T. spiralis* least common. The incidence of human infestation with these parasites has reached a low level in England but present methods of control will not bring about eradication or significant reduction in the present level of incidence.[32]

Cysticercus bovis is transmitted to man as a result of eating raw or rare beef containing cysts of this parasite in much the same way that *T. spiralis* is transmitted to man by eating raw or undercooked pork. Cattle may become infested as a result of grazing on grass contaminated with droppings of birds such as gulls that scavenge sewage. Use of human excrement as a fertilizer for cattle forage can also cause infestation of cattle.

Sheep are a reservoir for *E. granulosis*. Feeding raw slaughter house waste to dogs transmits the parasite to the dog which in turn can transmit the tapeworm to man.

The large tapewrom of *C. bovis* known as *Taenia saginata* (see Table 10.7) will not mature in any host but man which greatly limits research studies. Because of this difficulty studies were made on a closely related tapeworm, *Cysticercus pisiformis* which is found in the rabbit and which matures in the dog. Doses of 10 to 50 krad of gamma radiation were used. Ninety per cent of the worms were rendered sterile by the 10 krad dose. Higher doses produced ste-

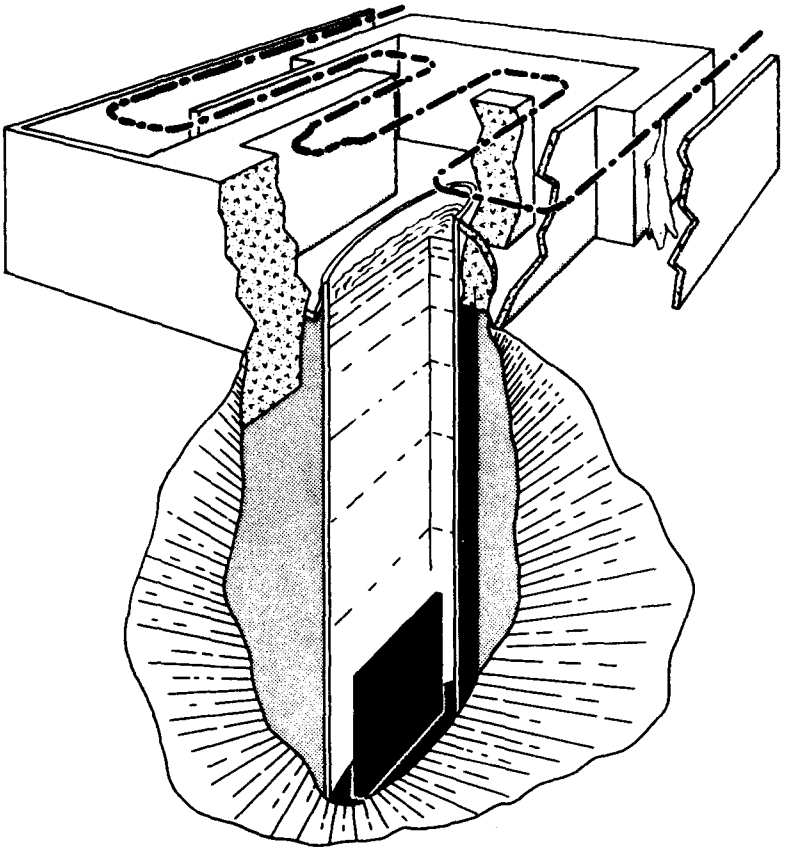


Fig. 10.12—The large planar source is shown in its storage position at the bottom of the shielding well in a pool of water. In use, the source is raised to a position between the two conveyor rails (shown by the dashed lines) which carry the hog carcasses.[33–35]

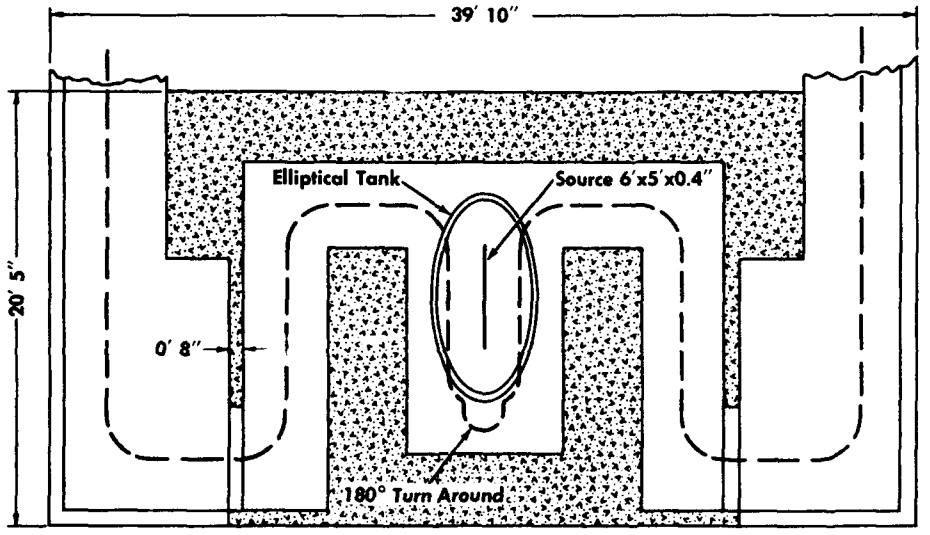


Fig. 10.13—Plan view of pork irradiation chamber.[33–35]

rility and also greatly stunted the growth of the worm. It was concluded that gamma radiation is effective in preventing normal growth and reproduction of *C. bovis* as well as *T. spiralis*.[32]

Taylor and Parfitt[32] commented that if subradiopasteurization of fresh meat is used in combination with greater centralization of slaughter of beef and pork, both *C. bovis* and *T. spiralis* might be eradicated. Eradication of *E. granulosus* will be a greater problem since it is transmitted by "man's best friend," the dog, rather than directly by meat.[32]

TUBERCULOSIS

Man is susceptible to the bovine strain of the tubercle bacilli. A survey by Price[28] showed that 11.7 per cent of 1202 cases of tuberculosis in the United States were bovine. Hull[28] states that "from a public health point of view, tuberculosis cattle constitute the most important animal source for tubercle bacilli that are virulent for man." However, the vigor with which the veterinary profession in the United States has attacked the problem of tuberculosis in cattle has nearly eliminated this disease in cattle in the United States. Out of over 14 million cattle slaughtered and inspected in 1945, less than 6000 showed lesions considered tubercular. Recent tuberculin tests of large groups of cattle also showed that the average incidence of tuberculosis in the cattle of this country is less than 1 per cent as compared to over 30 per cent estimated for Great Britain, France, and other European countries.[28]

Limited tests by Lawrence[36] showed that a radiation dose of 240 krep resulted in no growth of a human (Olsen) strain of tubercle organisms, but that a dose of 80 krep was insufficient to prevent growth. Thus, while the transmission of tuberculosis from fresh or undercooked beef is probably a minor consideration, a pasteurizing radiation dose between 80 and 240 krep might be useful in minimizing such transmission.

BRUCELLOSIS

Also known as Bang's disease or undulant fever, brucellosis is second only to tuberculosis in seriousness as a disease of cattle. It is prevalent in 42 states both in dairy and range cattle. Infections in Connecticut have run as high as 90 per cent of the herds, but 10 to 30 per cent infection has been more typical for herds in most states.[28] The prevalence of this disease in cattle and the fact that man is susceptible makes brucellosis worthy of consideration. The number of cases in man reported by state health officers has been increasing and has averaged around 3000 to 4000 cases per year.[28] The fact that the incidence of the disease is much higher among packing house workers than in the nation as a whole demonstrates the transmission of the disease by contact. A large number of human cases, however, result from human consumption of unpasteurized milk and milk products.

ORNITHOSIS

A disease of domestic fowl and identical to psittacosis in parrots, ornithosis is a disease that has caused some alarm. In the United States the disease is widespread in pigeons, who transmit it to chickens, turkeys, and ducks. The disease may be transmitted to man by the handling of infected dressed fowl.

Prelinimary tests by Lawrence[36] indicated that the virus that causes this disease is quite susceptible to gamma radiation. Doses of 240 krep inactivated the virus, but doses of 80 krep did not. This subject should also be given additional study.

INFECTIONS FROM SEAFOOD

A number of infections can also be transmitted by fish and other seafoods. Fish tapeworm infection in man is quite common. Pasteurization of seafoods with gamma radiation could reduce the transmission of such infections to man.

RADIOPASTEURIZATION OF FRESH MEATS, POULTRY, EGGS, AND SEAFOODS

The radiopasteurization of meat has many problems requiring further study. These should be investigated, since the new tool of ionizing radiation offers a new opportunity to reduce disease transmitted by meat.

The effect of radiopasteurization on meat was described in Chap. 9. It was concluded that for ground raw pork and ground raw beef, refrigeration shelf life can be extended to as much as two weeks,[41] with a dose of 80 krep.

Hannan and Shepherd[42] have reported similar conclusions on irradiated raw chicken. Radiation doses of 250 krad gave extended control over spoilage for several weeks[42] without producing excessive organoleptic changes (see Chap. 9).

There appears to be little objective for the irradiation of whole eggs. The shell of any egg is not very susceptible to bacterial attack and fresh eggs usually contain no bacteria. However, egg products such as frozen or dried whole egg magma or egg yolk may require pasteurization. Heat can be used but the process must be very closely controlled because of heat sensitivity of the products. Heat pasteurization has the additional disadvantage of necessitating the thawing of frozen products.

The problem of pasteurizing egg products is particularly important in Great Britain where appreciable quantities of these products are imported from Australia, New Zealand, and China. These imports (particularly Chinese) are often contaminated with microorganisms such as *Salmonella, S. paratyphi, S. typhimurium, S. senftenberg, S. thompson,* and *S. pullorum.* Several outbreaks of paratyphoid fever from *S. paratyphi* and food poisoning from *Salmonella* have occurred in Great Britain which have been traced to egg products.[37]

Brookes, Hannan, and Hobbs have studied the use of a radiopasteurizing dose of about 0.5 Mrep of ionizing radiation to treat eggs contaminated with Salmonellae and other microorganisms.[37] Hannan showed that these products could be radiopasteurized so as to be free of contamination. Cakes produced from radiopasteurized frozen eggs and nonirradiated frozen eggs show no loss in cake quality insofar as cake volume is concerned.[37] Other quality characteristics of cakes made from radiopasteurized eggs were closely similar to those of the controls.

Radiopasteurization, besides being a possible public health measure, would also have many commercial advantages. Brownell et al., [38-40] proposed a new method of wholesaling fresh meat based on radiopasteurization. This new method, proposed for consideration by packing houses and retailers, consists of preparing packaged standard cuts of fresh meat, such as packaged fresh ground meat, packaged cut-up chicken, and the like, in retail-size portions at the packing house rather than at the retail meat market. The packaged meat would be radiopasteurized at the packing house by means of a relatively small dose of gamma radiation prior to shipping to the retailer. The feasibility of this method depends upon the extension of the refrigerator shelf like of fresh meat by radiation pasteurization. Recent trends in retailing meats and the economics of meat processing suggest the desirability of handling meats in this way.

The meat departments of many supermarkets have found that customers show a preference for purchasing cuts of meat (cut-up chicken, ground meat, and other meat products) that are already weighed and packaged. This method of merchandising is popular with most costomers, probably because it avoids the necessity of their having to wait to be served by a butcher and permits them to inspect and select various cuts at their leisure. This practice also permits the cutting and prepackaging of meat in advance of the busiest market hours and results in more efficient use of the meat cutter's time. This method of retailing meat is popular with both the retailer and the customer and is becoming more or less standard practice in the larger meat markets.

One disadvantage of prepackaging fresh meat is the need for prompt sale of the cut-up meat to prevent loss by spoilage. This problem has necessitated the limiting of the practice to the larger meat markets that have a rapid turnover. Cut-up chicken and ground fresh meat can be kept for only a few days even under refrigeration, whereas the uncut carcasses can be kept under refrigeration much longer. The uncut flesh of an animal that was in good health at the time of slaughter is relatively sterile but when the flesh is cut the surface becomes contaminated with a variety of microorganisms from the air and from the hands and implements of the butcher. This contamination can become quite extensive in the case of ground meat since meat trimmings with a high surface contamination are frequently used, and quite often the meat grinder itself is not free of contamination. The introduction of a large number of microorganisms into an excellent growth medium accounts for the short shelf-life of refrigerated ground

fresh meats. Rapid spoilage practically prohibits the preparation of ground meat at the packing houses unless it is to be frozen.

The proposed method of handling cut-up, prepackaged, and radiopasteurized meat consists of the usual slaughtering procedures through chilling. Chilling prior to cutting is desirable to render the meat firm for easier cutting and also because there is less growth of microorganisms if the meat is cut while chilled and kept at refrigerator temperature until used by the consumer. Instead of shipping refrigerated quarter and half carcasses of beef, pork, and lamb from the packing house to the retailer, it is proposed that the chilled carcasses be cut, prepackaged, weighed, packed in cartons, and pasteurized by gamma radiation at the packing house and then shipped under refrigeration to the retailer. The advantages to the packing house, the retailer, and the consumer are enumerated in Table 10.8

DESIGN FOR RADIOPASTEURIZATION FACILITY FOR FRESH MEAT

A preliminary design has been described for a possible commercial gamma irradiation pasteurization facility for fresh meat. The description of this facility and the estimated cost for treating fresh meat are given in Refs. 38 to 40.

Figure 10.14 is a perspective view of the irradiation facility that was designed to radiopasteurize prepackaged fresh meat. In this design the meat would be cut in retail-sized portions and packaged in polyethylene or other flexible packaging materials. These packages of retail size would be loaded into cardboard shipping cartons 8 in. by 1 ½ ft by 2 ft. Each filled carton would weigh approximately 100 lb. A conveyor, containing baskets (each capable of holding 5 cartons of this size), would pass the cartons of meat up and down four times on either side of the radiation source. Since 8 in. of meat was measured to absorb one-half the radiation (one "half-value" thickness), 4 passes 8 in. thick would absorb 50, 25, 12.5, and 6.25 per cent, respectively, in each pass for radiation normal to the surface. Thus, four half-value thicknesses would absorb 93.75 per cent of radiation normal to the surface, provided the absorber was free of voids and discontinuities.

The capacity of the radiation facility shown in Fig. 10.14 was calculated to be 13.4 tons of prepackaged meat per hour based on the use of 1.5 megacuries of Ce^{137} for the source to give a radiopasteurizing dose of 80 krep. This design uses the same quantity of radiocesium (1.5 megacuries) as the pork irradiation facility shown previously in Figs. 10.12 and 10.13 but is 400 per cent more efficient in using the gamma radiation emitted. That is, five times as much pork can be irradiated (using the same dose) because of more efficient absorption.

The estimate of the annual operating cost is given in Table 10.9. This estimate assumes depreciation of the radiation chamber and source over a 10-year period with amortization of the total investment by use of a "sinking" fund. Cesium-137 has a 30-year half life and the source is assumed to have a salvage value of only 50 per cent at the end of the 10-year amortization period even though, through activity replacement, it will be at full strength.

Based on an irradiation dose of 80 krep and an operating schedule of 20 hr per day, 260 days per year, the cost of irradiation was estimated to be $0.0011 (or about 1 mill) per pound.

HIGH RADIOPASTEURIZATION IN PRESERVING FOOD

Brownell and Purohit have reported that a new process of food preservation has been given limited study and appears very promising as a means of increasing the refrigerator storage life of a number of perishable foods.[43,44] In this process, foods such as meats and vegetables are precooked or blanched, respectively, then packaged in retail-sized portions in 2-mil polyethylene bags, heat sealed, and given a 1-Mrep dose of gamma radiation. This radiation dose is not sufficient to sterilize but is sufficient to destroy most vegetative organisms, molds, and yeasts that cause rapid spoilage even under refrigeration. On the other hand, limiting the dose to 1 Mrep avoids some of the undesirable flavor experienced with sterilizing doses of radiation. Preliminary tests have shown successful storage of such items as smoked salmon, smoked chub, cleaned boiled shrimp, green peas, and diced avocados.[43,44]

Table 10.8—ADVANTAGES OF CUTTING AND PREPACKAGING FRESH MEAT AT THE PACKING HOUSE[40]

Advantages to the retailer

1. Fewer butchers, if any, would be required at retail stores. (More would be needed at packing houses, where they could be used more efficiently.)
2. Some retailers might dispense entirely with meat cutting.
3. Handling of bones and meat scraps would be eliminated.
4. Shipping costs would be reduced.
5. Packaging of meat at the store would be avoided.
6. Less flour space would be required for meat processing.
7. Less refrigerator space would be required per pound of meat sold.
8. Total cost of packaged meat could be reduced by mass handling methods at packing houses.
9. Fresh pork would be free from danger of contamination by trichinae.[31]
10. A number of the 80 or more other diseases transmitted from animal to man[28] might be subject to centralized control by radiopasteurization.
11. Loss of meat by spoilage could be reduced.
12. Satisfying consumer preference for prepackaged meats, even in smaller stores, could increase trade.
13. Necessity of weighing could be eliminated or reduced.

Advantages to the packing house

1. Mass-handling methods of cutting could be used at packing houses for greater economy.
2. Mass-handling methods of prepackaging could be used at packing houses.
3. Packing houses could handle bones and meat scraps at a profit more easily than the retailer.
4. Demand for specialty items such as radiopasteurized prepackaged meat could increase the trade of packing houses using this method.
5. Total trade also could be increased by reducing handling costs through mass-handling methods at packing houses.
6. The sale of pork could be increased by eliminating the danger of trichinosis.*
7. Since only salable meats would be shipped, both shipping cost and space, and handling would be reduced (70 lb of cut meat is equivalent to 100 lb of carcass meat).

Advantages to the consumer

1. Of even greater importance to the consumer than to the retailer and packing houses, pasteruization of pork would remove the causative agent of trichinosis and eliminate danger from this disease.
2. There are more than 80 other diseases transmitted from animal to man,[28] including a number transmitted by the handling of raw meat or by the ingestion of undercooked meat. Examples of such diseases are anthrax, ornithosis, salmonellosis, beef tapeworm, brucellosis, etc. A number of these diseases, in addition to trichinosis, possibly could be controlled by gamma irradiation, which would be an important public health consideration.
3. Radiopasteurized meat has a longer refrigerator shelf-life than untreated fresh meat, which would be an advantage to housewives who shop at a supermarket only once or twice a week.
4. The cost of meat could be reduced by savings realized in mass-handling techniques.

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*Certain types of meat products popular in Europe, such as some of the sausages made with uncooked pork, could be prepared form irradiated pork without the danger of trichinosis. Such variety would increase meat consumption. In addition, irradiation of all raw pork, at central plants, could well supplant the method of microscopic inspection of pork for the trichina larval worms in those countries in Europe and South America where such microsopic inspection is required by law.

Table 10.9—ESTIMATED ANNUAL OPERATING COSTS[40] FOR MEAT-IRRADIATION FACILITY USING Cs^{137}

Wages, salaries	
Experienced health physicist (half time)	$ 4,000
Maintenance engineer with health physics training (half time)	3,000
Cleaning and sanitation (part time)	1,000
Supervision and clerical labor (10% of operational labor)	800
Salaries and wages not associated with operation of radiation chamber (50% of direct labor and supervisory costs)	4,400
Total	$ 13,200
Operating costs other than wages and salaries	
Maintaining source activity ($125,000 every 5 years)	$ 25,000
Refrigeration	2,000
Utilities	1,000
Conveying	1,000
Repairs and maintenance of chamber (5% of chamber costs)	4,100
Miscellaneous contingencies	1,500
Total	$ 34,600
Overhead	
Payroll overhead (15% of direct labor and supervision)	$ 1,980
General plant overhead (50% of labor and operation cost)	23,900
General administration overhead (10% of labor and operation cost)	4,530
Total	$ 30,410
Taxes, Interest, and Insurance	
Property taxes (2% of radiation chamber cost)	$ 1,650
Income tax (2½% of total investment)	14,260
Interest (5% of total investment)	28,520
Insurance (1% of total investment)	5,700
Total	$ 50,130
Depreciation and obsolescence	
Radiation facility (8% of chamber cost)	$ 6,600
Radiation source (8% of initial cost minus the salvage value)	19,260
Total	$ 25,860
Total cost for year	$154,200

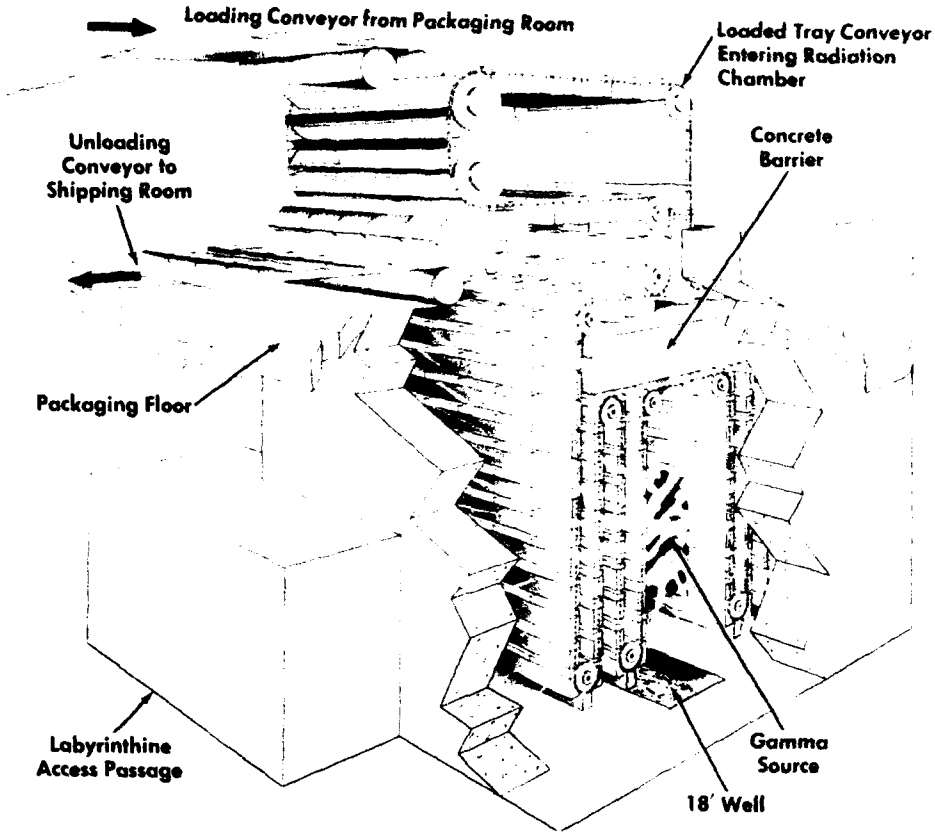


Fig. 10.14—Cutaway view in perspective of radiation chamber for prepackaged meat.[38-40]

The vegetative cells of *staphylococcal*, *salmonella*, and *streptococcal* organisms can be inactivated by dosages used in the process of high radiopasteurization. The primary danger of food poisoning, using dosages in this range, would be that of the spores of *Clostridium botulinum*. The vegetative cells are destroyed and with proper storage conditions, i.e., at refrigerator temperatures, the development of spores of this organism to the vegatative state with the accompanying production of toxin could be prevented. As a precaution, the food items to be processed in this manner should be packaged aerobically, in containers such as polyethylene, which readily transmit oxygen, and thus conditions are provided which are unfavorable to the development of the spores into vegetative cells. As a further precaution, the foods may be given a heat-treatment such as cooking, in the case of seafood and meat, or blanching, in the case of fresh vegetables, prior to irradiation. Such a limited heat-treatment would not destroy all the spores of anaerobes, but when used in combination with radiation treatment, it will provide an additional factor of safety.

DESIGN FOR HIGH-RADIOPASTEURIZING FACILITY

Gamma irradiation facilities were designed[43,44] similar to that shown in Fig. 10.14 but have an estimated capacity of about 2 tons per hour using a 1-Mrep dose. Cost estimates similiar to those described indicated that foods could be irradiated (1 Mrep) at an estimated cost of about 1 cent per pound.

A radiation dose of from 0.8 to 1 Mrep is required for high radiopasteurization. As this is an appreciable radiation dose, radiation costs will be high and a radiation facility that makes efficient use of radiation should be used. For this reason, a multipass conveyor, similar to that considered for the irradiation of prepackaged raw meat (see Fig. 10.14), was proposed. If Cs^{137}

sources were purchased for such a facility, four passes, each containing a half-value thickness of food, are recommended. However, if cooling-reactor fuel elements are used instead of Cs^{137} as a source of radiation, the cost might be reduced. In using reactor fuel elements, two passes are recommended.

A number of reactor fuel elements properly arranged can be used to provide a uniform radiation field in one plane at selected distances from the source. Such a scheme is proposed for the high-radiopasteurization facility shown in Fig. 10.15

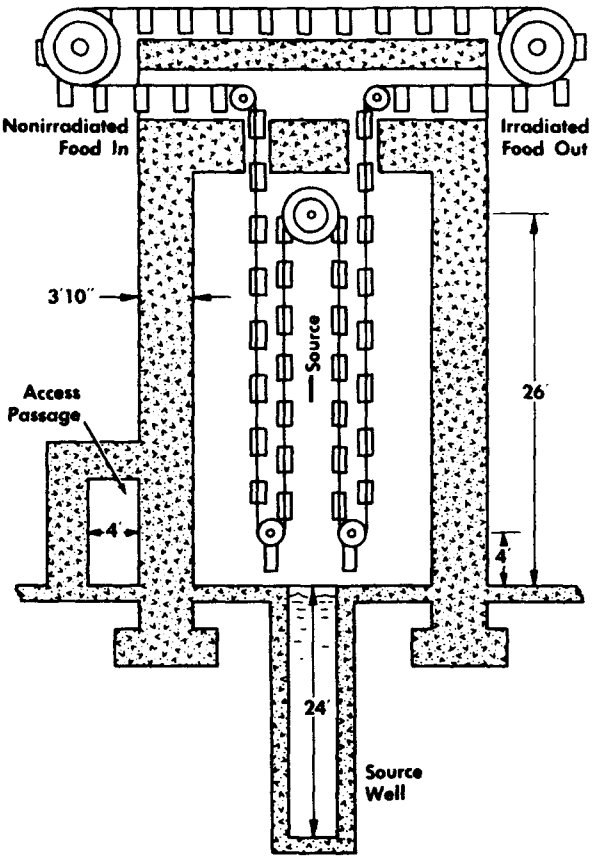


Fig. 10.15 — Elevation view of high-radiopasteurization chamber.[43,44]

Packaged mixed fission products or packaged Cs^{137} separated from fission products might be used as the sources of radiation for a high-radiopasteurization gamma facility. However, such sources are not at present available and will not be available until suitable plants are put into operation to process large quantities of fission-products wastes. On the other hand, fuel elements are being removed constantly from reactors now operating, and the supply will increase as new reactors come into operation. The supply of such reactor fuel elements is limited, but a sufficient number could be made available to establish a number of radiation facilities capable of processing foods on a commercial scale.

It is not proposed that cooling-reactor fuel elements be considered as the answer to the problem of suitable gamma sources for general use in industrial plants, but such fuel elements might fill the need for radiation sources for the immediate future. Reactor fuel elements may be too expensive to use if the high inventory costs of fissionable materials were charged against the fuel elements when used as a source of radiation. However, for reasons relating to the processing techniques, it is the practice in present fuel-processing plants to store these elements under water for several weeks before they are processed. During this period, the intense radiation is dissipated in the water used to shield the cooling elements. Except for limited use in some research experiments, this radiation is at present wasted.

The reactor fuel elements proposed for use in this facility have a very high gamma activity but a very rapid decay rate. It is suggested that each fuel element to be used for a period of about 2 months and then replaced with a new one. If the replacement schedules for different fuel elements are staggered, greater uniformity of radiation flux will be possible. However, it probably will be necessary to make adjustments in the material flow rate through the facility during the period of operation to compensate for the decay of the fuel elements. The irradiation of a large volume of foods necessitates continuous operation involving the use of a conveyor system to pass the foods into the radiation chamber, past the radiation source, and out of the chamber. Efficient operation requires that the total thickness of foods being conveyed through the chamber absorb most of the radiation.

Based on an irradiation dose of 1 Mrep, the capacity of the radiation facility using 20 reactor fuel elements[43,44] as shown in Fig. 10.15 was calculated to be 2.1 tons per hour. The estimated annual operating costs and the estimated unit costs are given in Tables 10.10 and 10.11, respectively.

The commercial development of the process of high gamma radiopasteurization probably would greatly increase the use of refrigeration both in commercial handling of food and in storing foods in the home. If foods could be kept for 2 or 3 months or longer by such a process, the housewife might use an additional refrigerator solely for storage. In this regard, the process would be competing with the use of a deepfreezer, but it would be used for food stored for shorter periods. Foods can be stored at 40°F more cheaply than at deepfreezer temperatures, and the waiting period required for thawing would be eliminated. The added convenience to the housewife of such a process and the savings in prevention of food spoilage would probably justify its use.

Cost estimates given in Table 10.11 indicate that a large-sized plant capable of processing about 2 tons per hour with two 8-hr shifts per day for 260 days per year could irradiate foods with a high-radiopasteurization dose of 1 Mrep at an estimated cost of about 1 cent per pound. This cost is considered to be in the range of commercial feasibility.

The design was based on the use of 20 cooling-reactor fuel elements rented at an estimated rental charge of $5,000 per month, plus handling and shipping charges. It was suggested[43,44] that such a rental charge might be helpful in making nuclear-power reactors more profitable and thereby hasten the day when electric power from nuclear reactors will be able to compete with electric power from conventional fuels.

POTATO IRRADIATION FOR SPROUT INHIBITION

The irradiation of potatoes for preventing sprouting under refrigerator storage without developing undesirable flavor changes has been described in Chap. 9. Certain types, such as Idaho russets, are preferred for baking, French fries, etc. However, they are available only about 8 or 9 months of the year. It usually is not possible to keep them in refrigerated storage for longer periods because of sprouting followed by rapid deterioration. Tests with these and other irradiated potatoes have demonstrated that irradiated mature northern potatoes can be kept in refrigerated storage from one year until the next, thereby making desirable types of mature potatoes available the year around.

DESIGN FOR A POTATO IRRADIATOR

Figure 10.16 shows a perspective view of a design for a potato irradiation facility described by Brownell et al.[45] The radiation source selected consists of two cooling reactor fuel elements. In this facility, potatoes are unloaded onto a vibrating conveyor where they are fed through a "Peck-type" conveyor-elevator. The potatoes are then conveyed in a single pass on either side of the radiation source and are dumped at the high point of the return onto a discharge vibrating trough conveyor. It is estimated that the facility could irradiate about 250 bushels of potatoes an hour using a radiation dosage of 10 krep at a cost of $4.00 per ton.

Irradiation of grains and potatoes has been studied by American Machine and Foundry Company.[46,67] The firm has presented a design of a railway car for processing grain, a semi-

Table 10.10—ESTIMATED ANNUAL COST FOR OPERATING A HIGH-RADIOPASTEURIZATION UNIT USING 20 REACTOR FUEL ELEMENTS[43,44]

1. Total investment	
Estimated cost of radiation chamber	$ 66,000
2. Operation cost	
a. Wages and salaries:	
(1) Two operators with limited health-physics training	$ 10,000
(2) Supervision and clerical labor	2,000
(3) Salaries and wages not associated with operation of radiation chamber	6,000
Total	$ 18,000
b. Other operation costs:	
(1) Shipping costs for 20 reactor fuel elements (every 2 months)	$ 12,000
(2) Handling cost for fuel elements during transfer and installation	10,000
(3) Rental of 20 fuel elements for 12 months ($5,000 per month)	60,000
(4) Repairs and maintenance on chamber (5% of chamber and conveyor costs)	3,300
(5) Miscellaneous	1,000
Total	$ 86,300
3. Overhead	
a. Payroll overhead (15% of wages and salaries)	$ 2,700
b. General plant overhead (50% of wages, salaries and operation)	52,150
c. General administration (10% of cost of labor and operation)	10,430
Total	$ 65,280
4. Taxes, insurance and interest 10% of total investment	$ 6,600
5. Depreciation and obsolescence of radiation chamber $66,000 × 0.08	5,280
Total cost for year	$181,460

Table 10.11—ESTIMATED UNIT COST FOR HIGH-RADIOPASTEURIZATION[43,44]

Dose, Mrep*	Cost, 260 days†		Cost, 100 days‡	
	Dollars/ton	Cents/lb	Dollars/ton	Cents/lb
0.10	2.06	0.1	3.06	0.18
0.25	5.15	0.25	7.65	0.45
0.50	10.30	0.51	15.30	0.90
0.75	15.45	0.76	22.95	1.35
1.00	20.60	1.03	30.6	1.79

*Capacity = 2.1 tons/hr (for 1 Mrep dose).

†260 days/year (16 hr/day)

Capacity per year = $2.1 \times 16 \times 260$ tons = 8.74×10^3 tons.

Therefore, unit cost = 20.6 dollars/ton = 1.03 cents/lb.

‡100 days/year (24 hr/day)

Capacity per year = $2.1 \times 24 \times 100$ tons = 5.04×10^3 tons.

Therefore, unit cost = 35.8 dollars/ton = 1.79 cents/lb.

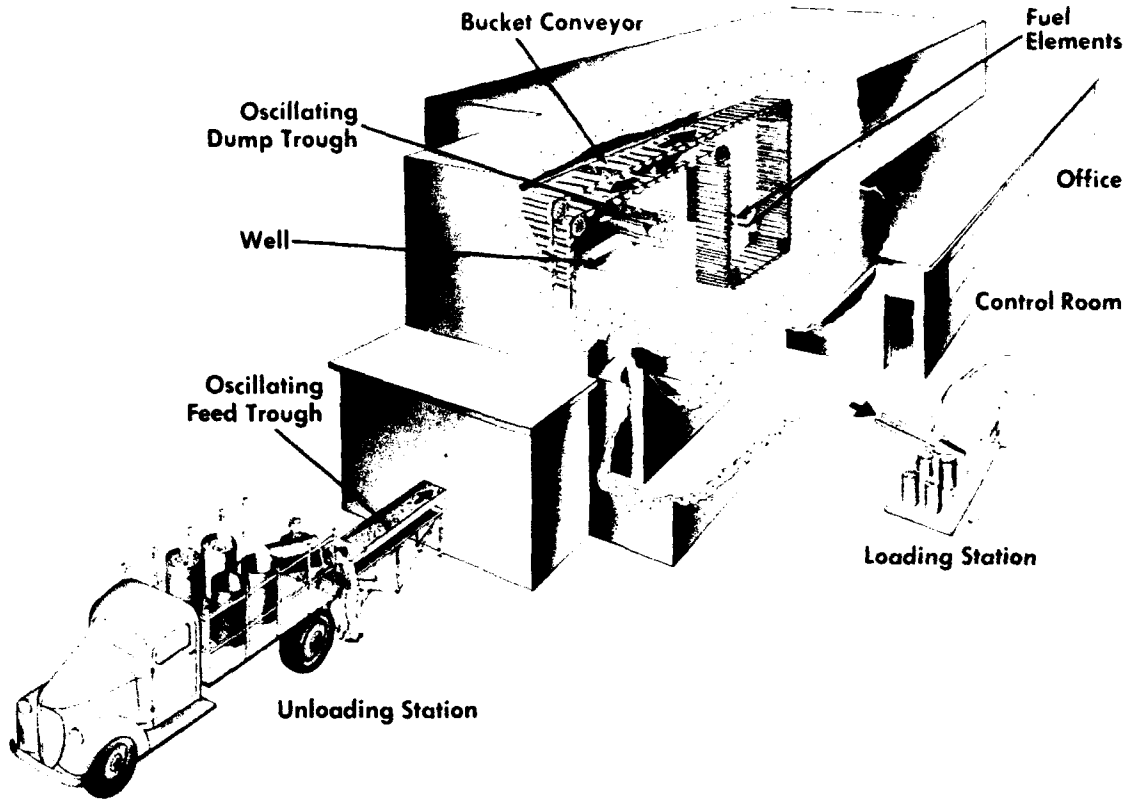


Fig. 10.16—Perspective cutaway view of potato irradiator using cooling reactor fuel elements.[45]

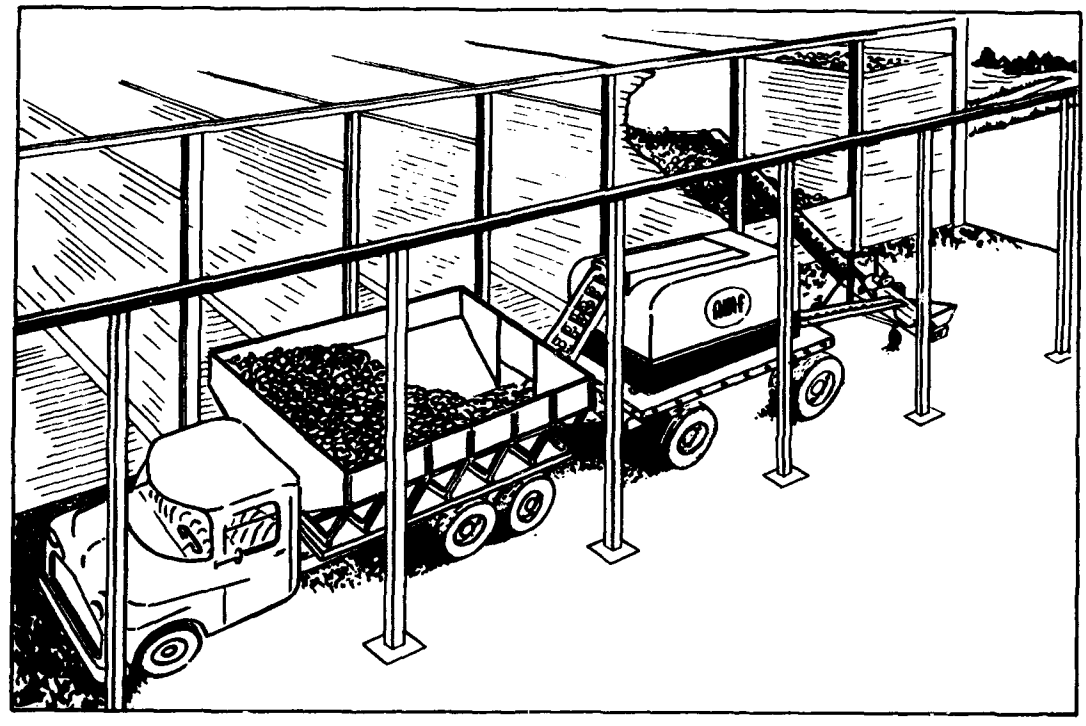


Fig. 10.17—Truck-drawn mobile potato irradiator. (Courtesy American Machine and Foundry Co.)[46,67]

mobile crop irradiator and facility for irradiation of potatoes with Co^{60} gamma radiation. Gravity feed systems are used. A mobile truck-drawn facility is shown in Fig. 10.17.

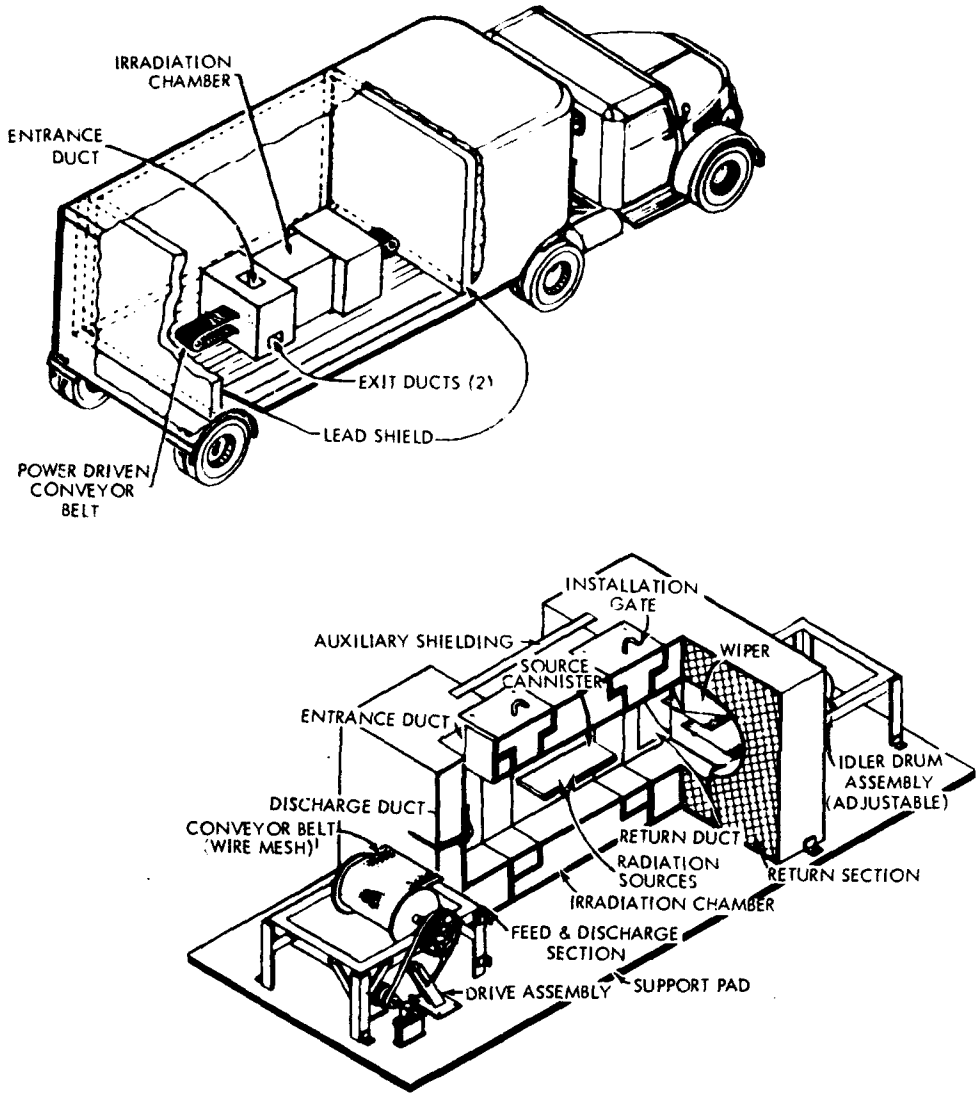


Fig. 10.18—Portable pilot potato irradiator. (Courtesy Brookhaven National Lab.)[47,67]

Kuhl et al.,[47] also have presented a design for a portable pilot irradiator such as shown in Fig. 10.18 (4000 curies–6 flat plaques of Co^{60}) that can provide for the irradiation of 40 to 50 million bushels of potatoes per year out of the total crop of 375 million. Designs are also presented for multimegacurie sources. Kuhl states that handling costs are comparatively low for Co^{60} sources.[48]

RADIOSTERILIZATION OF FOODS

A great many studies are now in process in a number of laboratories using various doses of ionizing radiation for food preservation. The initial studies were in most cases directed toward the use of "sterilizing" doses in the range of 2 to 6 Mrep. Work to date has indicated that such doses of radiation produce undesirable changes in many foods, particularly in the odor and flavor of meats and dairy products. At present, no completely satisfactory method for preventing these undesirable changes is known but by using a variety of special techniques a large number of satisfactory radiosterilized foods have been developed.[17] Radiosterilization would

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permit long-term storage at room temperature and is the untimate goal of much research being conducted in this field. However, it presents more problems than the use of lesser dosages of radiation described previously in this chapter.

The problem of odor and flavor changes in radiation-sterilized foods was discussed in Chap. 9 and will not be discussed again in this chapter. One problem that does exist, which has been considered, is what the lower radiation dose limit should be for sterilization. Original investigators studying irradiated foods in the early 1950's had suggested that, based on data obtained with cultures of microorganisms in pure culture media, doses as low as 2 Mrep might be satisfactory. However, after the protective effect of foods was studied, the sterilizing dose was revised upward to 3 Mrep in the studies supported by the Quartermaster Corps of the U. S. Army. Findings by Anderson et al.[49] of radiation resistant microorganisms that require as much as 6 Mrep for sterilization has introduced new uncertainty as to the minimum radiation dose permissible in the radiosterilization of foods. This problem is now being studied by various contractors in the research program of the Quartermaster Corps of the U. S. Army, and a tentative value of 4.5 Mrad is considered adequate for "commercial" sterilization.

Using gamma radiation doses of 2.6 Mrep, Pratt and Eckland[50] reported that a number of canned foods had been satisfactorily preserved at room temperature for several months. The Quartermaster Corps has also reported on the satisfactory storage of a number of foods given radiation doses of 3 Mrep.[17] Both of these reports were discussed in Chap. 9.

Various size containers have been used in research studies for radiosterilized foods. As gamma radiation is very penetrating, there is no difficulty in giving fairly uniform radiation doses to foods packed in size 10 (about 6 ½-in. diameter) commercial containers, the largest diameter of "tin can" used for canning foods.[51,52]

DESIGNS FOR FOOD RADIOSTERILIZERS

Many studies have been made on the economic feasibility of food irradiation in comparison with present procedures, on the economics of various sources of radiation, and on the optimal locations of the radiation source. Several designs using various radiation sources for processing foods are presented for comparison. Among these are the logistic and economic feasibility study on radiation sterilization of foods by North American Aviation, Inc.,[53] study of representative American food processing plants and optimal location of radiation source by Fanhoe,[25] and various designs and economic studies presented by Kuhl,[48] Manowitz,[54] and Chamberlain.[46]

Jackson[55] has concluded that introducing irradiation for foods should not require large outlays for new equipment as conventional methods of handling, storage, and shipment should be satisfactory. However, some modifications may be necessary such as the conveyor system in the irradiation field. In most instances the radiation can be carried out subsequent to the packaging operation even though at times it may follow cooking or sealing operations, or be combined with heat treatment.

Fanhoe[25] has conducted studies to determine the optimal location for the irradiation source in a canned meat processing plant. As in case of irradiation of grain, this study was based on the assumption that neither substantial equipment nor a great amount of plant space is available and that the food products can stand little in the way of a price increase. The location selected as optimal is shown in Fig. 10.19.

Fanhoe et al.[56] have suggested pilot plant processing schemes using MTR fuel elements. Figure 10.20 shows a sketch of one of these designs. Provisions are made for lowering the fuel assembly into a storage canal when not in use. The food is loaded into tote boxes and can be given a varying amount of dosage by varying the speed of the drive mechanism carrying the tote boxes.

Their calculations based on a set of 23 fuel assemblies with the equivalent of 1 million curies at installation, showed that sterilization for 104 lb per hour production basis would cost $0.077 per pound.

North American Aviation, Inc. in their report on logistics and economic feasibility study on radiation sterilization of foods, have also advanced such designs using MTR fuel elements.[53]

An irradiation facility using Cs^{137} has also been advanced by North American Aviation, Inc.,[53] and is shown in Fig 10.21.

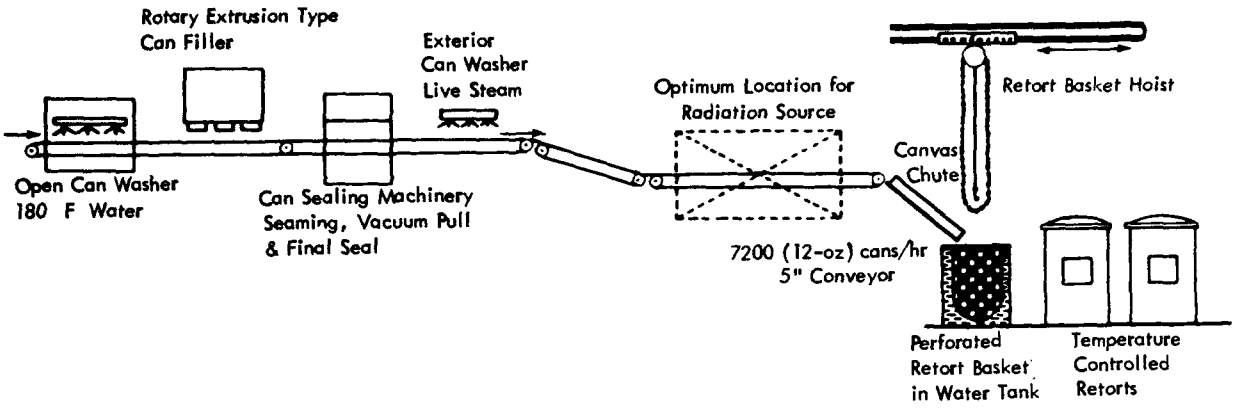


Fig. 10.19—Flow diagram for canned meat processing.[25,67]

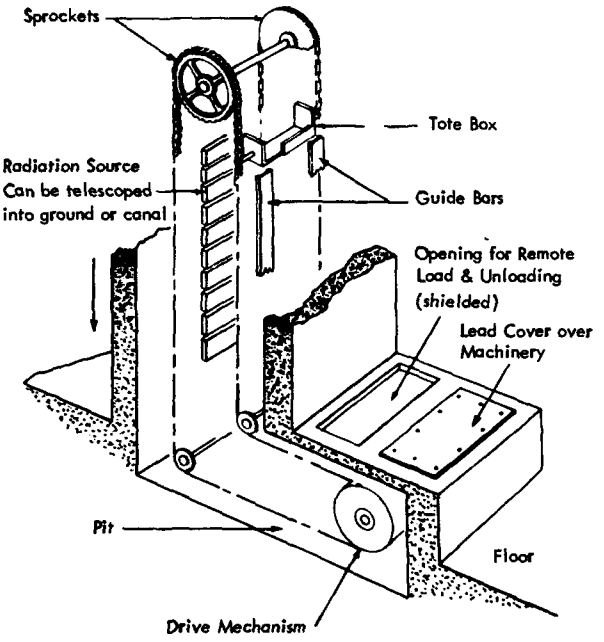


Fig. 10.20—Pilot irradiator using spent fuel elements.[66,67]

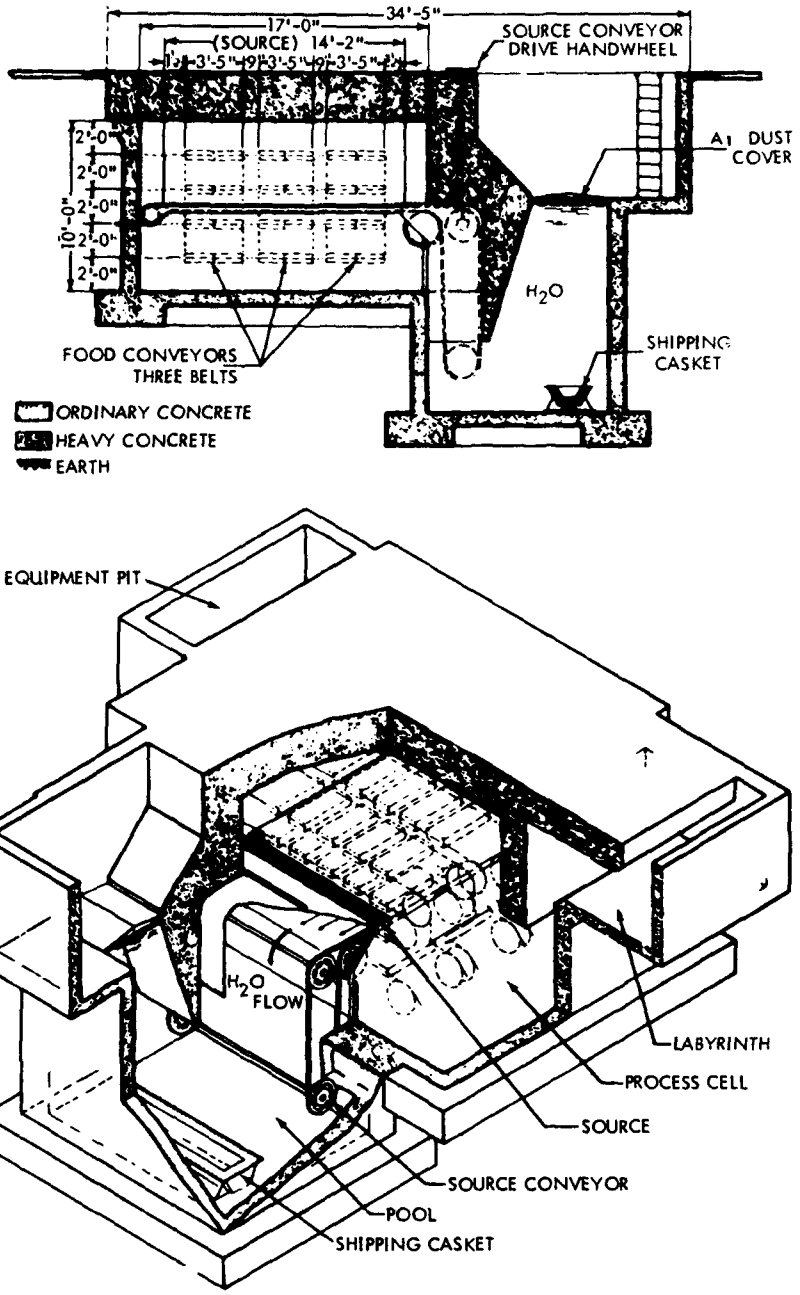


Fig. 10.21—Gamma irradiator using Cs^{137}. (Courtesy North American Aviation)[53,67]

Three conveyor belts which can be operated independently are used. Their economic study reports an estimated capital cost of 2.05 cents per pound for 2.5×10^6 rep and based on 3.31×10^6 lb per year with reactor fuel elements. (The study did not include cesium cost, however.)

Kuhl[48] has proposed a 500,000-curie cobalt source. A schematic diagram of the design is shown in Fig. 10.22.

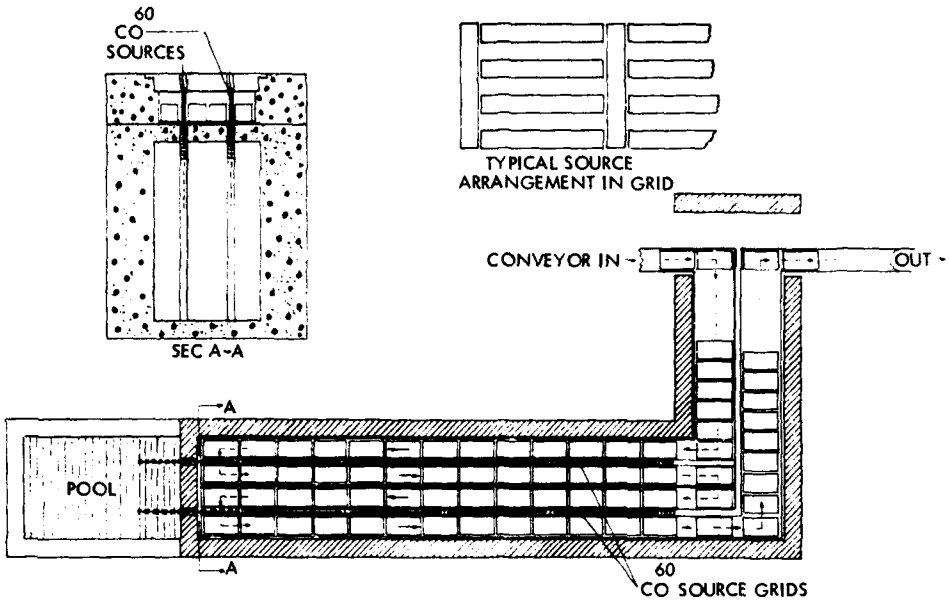


Fig. 10.22—Gamma irradiator using Co^{60}. (Courtesy Brookhaven National Lab.)[48,67]

The radiation source is in the form of thin flat strips encased in thin sheets of stainless steel. In his economic evaluation he also states that the handling charges are comparatively low in a Co^{60} facility. On the basis of 2500 lb of unit density material per hour and 4000 hr per year, it is reported that a dose of 2×10^6 rep would cost $0.0346 per pound. A multimegacurie source of Co^{60}, in the form of rods, has been suggested by Manowitz.[54]

U. S. ARMY IONIZING RADIATION CENTER

The U. S. Army Quartermaster Corps developed plans for the establishment of an ionizing radiation center to be located at Stockton, Calif., with the objective being to undertake early pilot plant operation. The cost of the center[57] was expected to be approximately $7,500,000.

It was originally planned to build a linear-accelerator-conveyor complex, comprised of a 24-mev, 18-kw accelerator and a heterogeneous reactor utilizing an indium sulphate solution as the source of gamma rays (see Chap. 4). However, the plans for the reactor were cancelled in favor of a multimegacurie Co^{60} source. This source was known as High Intensity Food Irradiator (HI-FI). The facility was to be charged initially with approximately 2 million curies of high specific activity Co^{60} which would have been produced in the Atomic Energy Commission's reactors. The cobalt source consisted of 2 vertical plaques with a conveyor containing food passing between (see Fig. 10.23).

In addition, the center was also to have a food processing plant with conventional equipment together with aseptic closing facilities, sharp freezing equipment, etc. These lines had a design capacity of 3000 lb of food products per hour. Figure 10.23 shows a plan of the Ionizing Radiation Center.[58] Late in 1959 plans to build the center were dropped.[59]

RADIOSTERILIZATION OF SURGICAL ITEMS

In addition to use in radiopreservation of foods, large multicurie sources of gamma radiation may be used in the field of medicine for radiosterilization. One such application is the

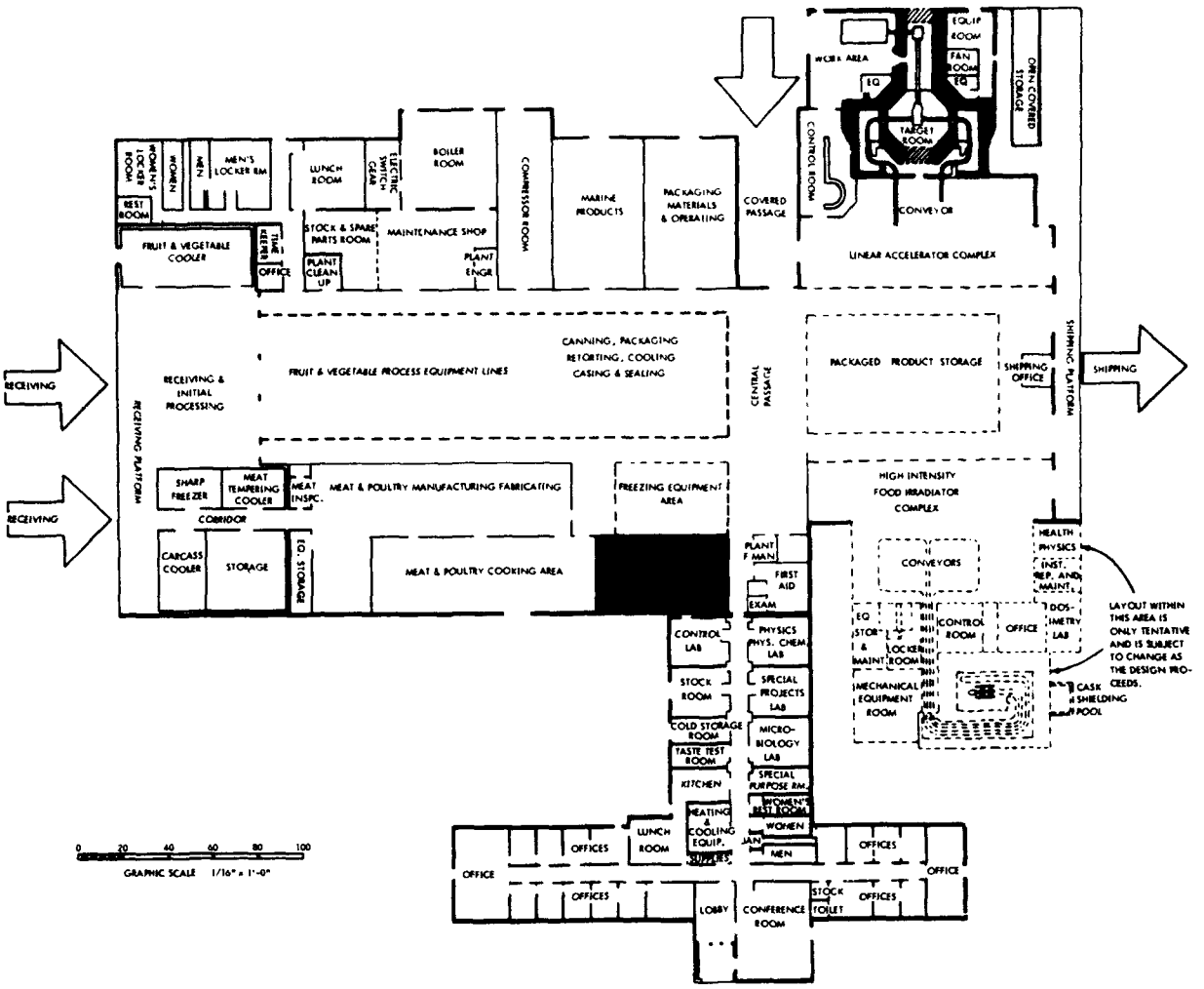


Fig. 10.23—Plan view of proposed U. S. Army Radiation Center showing HI-FI irradiator at lower right.[58]

sterilization of surgical homografts such as human bone-bank bone,[60,61] aorta,[62] and cartilage. Other applications are the sterilization of dressings, sutures, and instruments. Sharp instruments such as knives and scalpels are easily dulled by steam sterilization and at present are sterilized by being kept in strong bactericidal solutions, which is not a completely satisfactory technique. Other special instruments such as lighted probes have insulation that may be short circuited by either steam or solution sterilization. Solution sterilization short-circuited the wires running through probes used in surgery at the University Hospital in Ann Arbor. Therefore, the probes were packaged and heat sealed in polyethylene bags and sterilized with gamma radiation using the 3-kilocurie Co^{60} source in the Fission Products Laboratory at The University of Michigan.

ADVANTAGES OF RADIOSTERILIZATION OF BONE-BANK BONE

As pointed out by De Vries[60,61], two major problems confront the surgeon who makes use of bone-bank bone. One is the difficulty encountered in obtaining the bone and the other is controlling its sterility. A simple method of obtaining and sterilizing bone-bank bone without greatly altering its ability to stimulate new bone formation would certainly inprove the usefulness of bone-bank bone and increase its popularity.

Bone-bank bone, subjected to storage in antiseptics and antibiotics or to boiling, freezing, or freeze-drying, may still harbor living bacteria and viruses or be so altered in its chemical and physical properties that its usefulness as a transplant is impaired. Kreuz has reported that

16.3 per cent of their bone-bank bone, obtained from fresh cadavers, was rejected because of bacterial contamination; and Shutkin reported a case of homologous serum hepatitis following the use of refrigerated bone-bank bone.[61]

Boiling and autoclaving of bone before use, in all probability destroys the bacterial and viral contaminants but has the disadvantage of denaturing the protein constituents and inactivating the tissue enzymes. Freeze-drying bone, although an excellent means of preservation, does not assure the surgeon of a sterile product for it is recognized that many bacteria and viruses, including the common pathogens, are resistant to freezing and freeze-drying. If the surgeon were to assume that all bone obtained from the bone-bank were contaminated, he would be forced to sterilize it by one means or another before use.

Comparative Studies on Bone-bank Bone

De Vries and Maxwell[61] reported a comparative histologic study of bone transplants with special reference to irradiated bone. The rate of incorporation of bone treated with Co^{60} irradiation was compared with that of bone treated by freezing, freeze-drying, boiling and autoclaving. In the study, 5-mm cylinders of bone were taken from the dog ilium, processed, and inserted into various sites in the legs. The animals were killed at definite intervals, and histologic sections were made. The chief objective was the comparison of the rate of incorporation of transplants sterilized by the two methods known to kill viruses and spores, i.e., autoclaving and irradiation. The results were shown[61] in an animal that was killed 26 days after implantation of the processed homografts. The untreated homograft shows active new bone formation along the trabeculae of the bone transplant, as indicated by the photomicrograph shown in Fig. 10.24 (top). The lyophilized (freeze-dried) and irradiated transplant shows new bone formation that is at least as active as that of the untreated specimen. This is indicated in the photomicrograph shown in Fig. 10.24 (middle). Conversely, new bone formation in the autoclaved transplant is relatively indolent, as is indicated in the photomicrograph shown in Fig. 10.24 (bottom). This same pattern was confirmed by similar changes observed in 15 other animals killed at shorter intervals.

These results seem to show a definite trend indicating that lyophilized and irradiated bone is incorporated as rapidly as untreated homografts, and that it is incorporated into the body much more rapidly than autoclaved bone. It is believed that except for autoclaving, irradiation is the only method available for completely sterilizing bone. The tentative conclusion is made that irradiation of bone provides complete sterilization without materially altering its rate of incorporation. The clinical implications of this are important. It appears that a method of completely sterilizing bone now exists for use with homografts that does not reduce the rate of incorporation of the graft.

Use of Radiosterilized Human Bone-bank Bone

Human bone was cut to suitable sizes, placed in pyrex tubes, and lyophilized in the tubes to prevent enzyme degradation; the tubes were then sealed under partial vacuum.[61] The sealed tubes were given gamma-radiation doses of 4 Mrep. This high dosage was used to provide greater assurance of sterility. Lyophilized human aorta has also been sterilized by radiation.[62] Radiosterilized bone has been used in hundreds of human patients at the University of Michigan Hospital with unusual success.[60,61]

DESIGN FOR A GAMMA-RADIOSTERILIZER FOR SURGICAL PRODUCTS.

Brownell et al.[63] describe a design for a high-level gamma source for radiation sterilization in hospitals. The radiation source consists of 36 cylindrical rods, $\frac{1}{4}$ in. in diameter by 12 in. in length, which contain either Co^{60} or the refined waste fission product, Cs^{137}.

The cylindrical container for the source serves a dual purpose. It is used as a shipping container to transport the source from the reactor site and then becomes an integral part of the unit. An annular plug is rigidly attached to the source and seals the opening in the container during shipping and when the source is in the "off" position. This plug stays in position,



Fig. 10.24—Top: Photomicrograph of untreated homograft in a dog 26 days after implantation. Note active new bone formation along area of transplant. Middle: Lyophilized and irradiated homograft of bone in the same animal. New formation is at least as active as that with the untreated transplant. Bottom: Autoclaved homograft in the same animal. New bone formation is relatively indolent. (Courtesy of D. S. Maxwell, M. D., University of Michigan Hospital, Ann Arbor, Mich.)[61]

shielding the source and cannot be moved unless the door is bolted shut. An electric motor opens and closes the door, and another motor moves the source in and out of position.

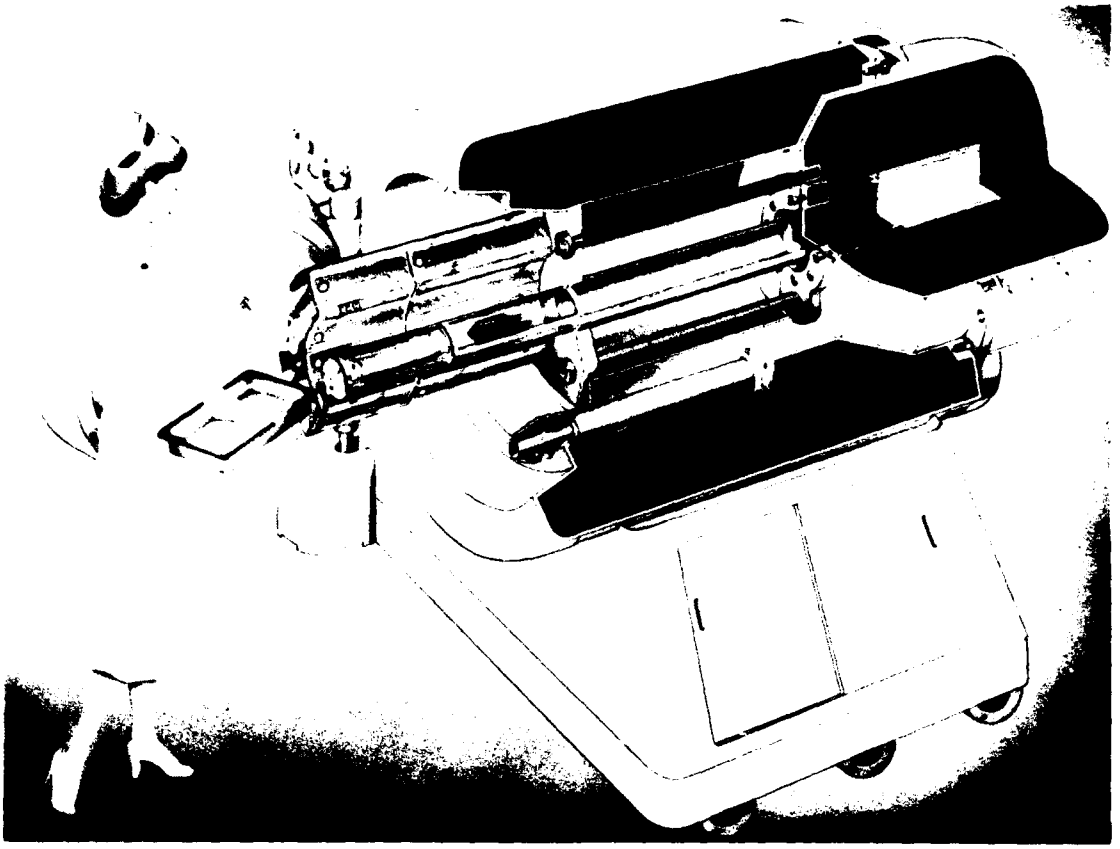


Fig. 10.25— Proposed hospital gamma radiosterilizer for surgical products.[63]

Figure 10.25 shows an operator placing material for irradiation in trays withdrawn from the radiation unit. Materials requiring a uniform dose, such as human bone, are placed in the cylindrical drawer in the center. Materials that do not require so accurate a dose are placed in the larger trays outside the source.

After charging, the door "close" button is pressed; and the motor closed the door, which is then bolted. The source "on" button is pressed and another motor then operates the screws to move the source into the irradiation chamber. The timer on the control box may be set to stop the radiation automatically after a predetermined time.

RADIOSTERILIZATION OF BULK MEDICAL SUPPLIES

The greatest volume of medical supplies that are sterilized is found in cotton, gauze, adhesive bandages, surgical dressings, and other similar bulky materials. The sterilization measures and protection against subsequent contamination are costly operations performed by the manufacturer in the interests of public health and customer acceptance of his product. Current practice employs steam under pressure for sterilizing. Although procedures vary somewhat, in the method employed by one large manufacturer, a number of pallet loads of materials are placed in a chamber and subjected to pressurized steam for a period of time sufficient to kill any microorganisms present. The success of this method depends on the ability of the steam to penetrate the cartons and packages of materials to provide enough heat to destroy the organisms.

A new method of sterilization was proposed by Brownell and Bulmer[64] which consists of applying a sterilizing dose of gamma radiation to the materials after packaging and before ship-

ment to consumer markets. This new method has several distinct advantages and eliminates many of the disadvantages of the present practice of steam sterilization.

It is quite natural to consider gamma rays as a sterilizing agent for bulk medical supplies such as surgical dressings when examining the extensive work that is now being conducted on the pasteurization and sterilization of food by the use of gamma rays. The advantages offered by gamma-ray sterilization can effect economies of operation while still producing a product free of microorganisms. Some of the advantages of radiation sterilization are:

1. Gamma radiation is extremely penetrating and can pass through several pallet loads of materials, killing microorganisms in its path.
2. Complete sterilization could be assured in even the most tightly packed containers and thus could produce savings in packaging materials.
3. The restriction to certain types of packaging materials would be eliminated.
4. Manufacturers could exercise a wider choice of packaging-material colors that would have a greater sales appeal.
5. Economies that could be effected by eliminating the irreparable damage done to heat-sensitive materials by the steam. (Gamma irradiation produces no appreciable temperature rise in the materials.)
6. Continuous processing lends itself to move efficient and economical materials-handling methods compared to the present system of batch operations.

The question arises as to what effects, other than killing microorganisms, gamma radiation will have on the materials so processed. In experiments on the radiation dose required to sterilize food wrappers using two bacteria, *E. coli* and *B. stearothermophilus*, it was found that a dose of 2 Mrep was sufficient to effect complete sterilization. No visible damage, such as color change, embrittlement, and clouding, was observed in any of the samples of plastic, cloth, or paper irradiated at this dosage.[65]

Design for a Radiosterilizer for Bulk Medical Supplies

Figure 10.26 shows the plan and elevation views for the radiation chamber using reactor fuel elements as a source of radiation proposed by Brownell and Bulmer.[64] As shown, the materials to be sterilized will be conveyed by racks holding three pallet loads on a monorail conveyor system. This particular design arose out of considerations for maximum utilization of the radiation field that emanates in all directions from the radiation source.

With reference to Fig. 10.26 (lower), material handlers will deliver to area A pallet loads of materials from production areas. Here the loaded pallets will be raised by fork-lift trucks onto the racks ready for irradiation. The racks will travel on the monorail system through the labyrinthine passage B and into the chamber past barrier wall C. The materials then make two passes, D and E, on one side of the radiation source F and two passes, G and H, on the other side. This arrangement will provide a more uniform radiation dose for the materials by irradiating both sides of the pallet loads. After a sufficient dose of radiation is obtained in the chamber, the materials pass around concrete barrier I and out the labyrinthine passageway J to the unloading and removal area K. From area K the sterilized materials could be loaded into railroad cars or sent to accumulation areas to await shipment.

Provided in the chamber is a well L, which would be filled with water and would be used to store the radiation source when not in use. It will be necessary for the operator to enter the chamber for such things as routine maintenance, changing of radiation source, or recovery of spilled materials from the pallets. The well will permit this access simply by lowering the source beneath the protective shield of water. Since there is no residual activity in the chamber after the source if lowered into the water, the operator could perform all necessary duties without danger.

Sterilization of bulk medical supplies by gamma irradiation appears promising, considering both the limited experimental work and the feasibility studies. The preliminary design for the radiation sterilization of bulk medical supplies (in pallets, 4 by 4 by 4 ft using reactor fuel elements as a source of radiation) has an estimated capacity of 120 pallets per 8-hr day. This capacity is sufficient to handle the production of a large manufacturing plant, and the estimated cost of operation was stated to compare favorably with known costs for steam sterilization.[64]

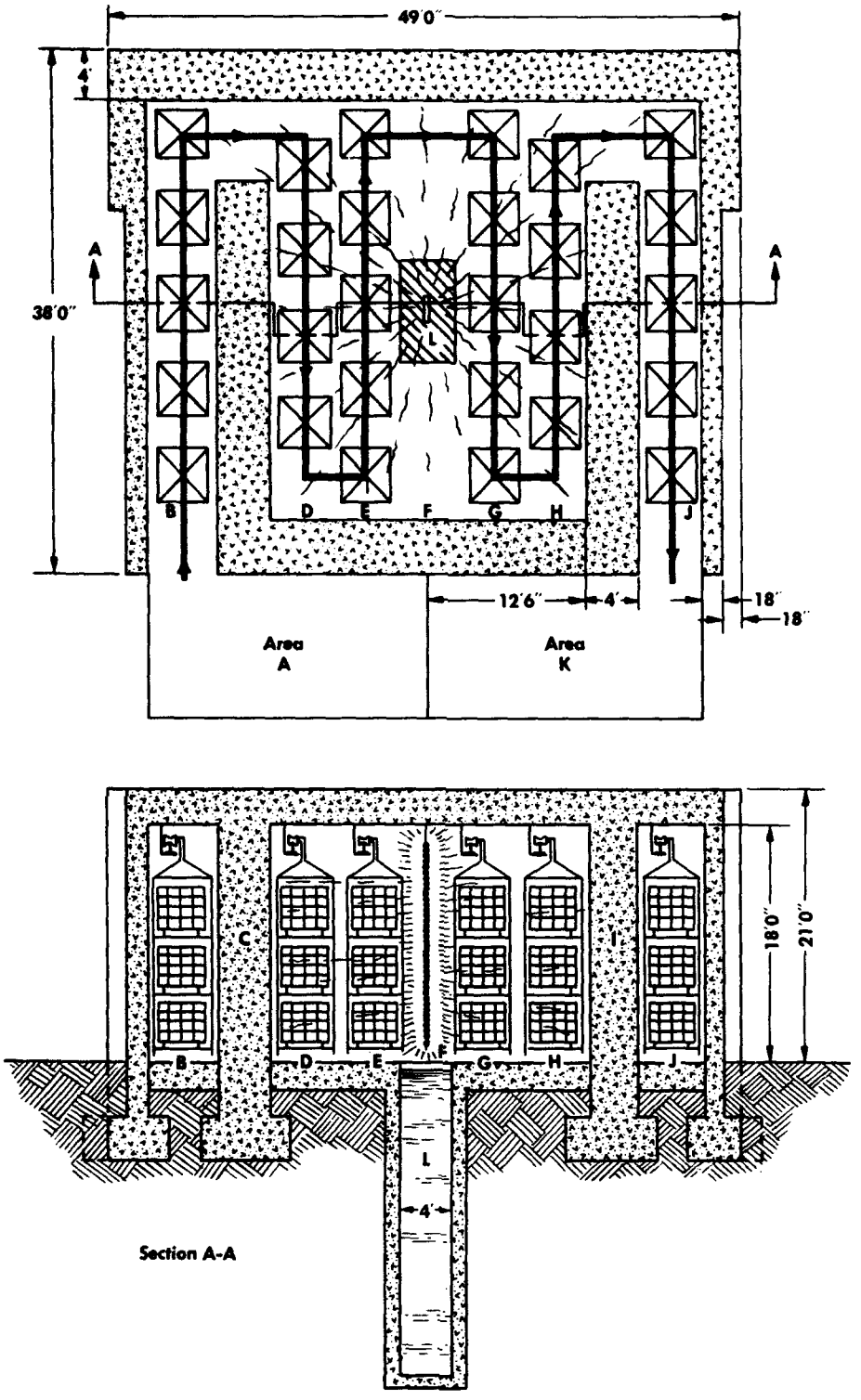


Fig. 10.26—Facility proposed for irradiation of bulk medical supplies. Upper: Plan view. Lower: Elevation view.[64]

RADIOSTERILIZATION OF HOSPITAL MATTRESSES AND SURGICAL ITEMS

The increase of staphylococcus infections currently plaguing hospitals has created a need for a new sterilizing process. "Staph," as it is commonly known, has developed a resistance to antibiotics and many patients contract staph infections after arriving at the hospital. To combat this microorganism it is desirable to sterilize all objects which might contain the germs. Hospital mattresses, pillows, and bed clothes are notorious carriers of many microorganisms, but because cotton and foam rubber fillings are damaged by heat and dampness, steam sterilization cannot be used.

Also awaiting more effective sterilization methods are many devices which are inserted and left in patients after operations. These include plastic organ parts, plastic arteries, and the like. A discussion with a staff neurosurgeon reavealed that certain siliconized polystyrene valves used to carry blood directly from the heart to the brain during operations develop post-operational infections in about one out of four cases. These valves are destroyed by steam sterilization and are currently "wet" sterilized at the factory and packed in an antibiotic fluid in plastic envelopes which are opened on the operating table. This seems to indicate that the infection-producing organisms are resistant to the antibiotic.

Many surgical instruments with hinged joints or close contact rubbing surfaces must be hand cleaned after each steam sterilization to insure cleanliness in jointed areas and to dry the areas between tight fitting surfaces which tend to collect water and corrode. These examples show clearly that additional sterilization methods are needed which are not destructive to the objects being sterilized, and which are dry and penetrating processes.

The gamma radiation dosage required varies with the item and the microorganisms in question. For the sterilization of bones, for example, a dose of 4.5 Mrad is used. To sterilize staphylococcus bacteria, however, only about 200,000 rads are needed. There are, of course, many organisms whose sterilization doses lie between these limits. The total acquired dose has been shown to be relatively independent of the rate at which the dosage is applied. However, for a hospital of any normal capacity, a method of fairly rapid sterilization is desirable.

Design for a Hospital Mattress Irradiator

The design of a hospital sterilizer is somewhat complicated because it must be versatile. The versatility could be defined in two ways: (1) ability to provide both high and low dosages in reasonable periods of time, and (2) ability to accommodate both large and small objects for sterilization.

In one large hospital having a 1200-bed capacity, 3215 contagious cases are admitted per year. The number of contaminated matresses averages 9 per day. If only about nine mattresses need to be sterilized per day a continuous irradiator such as those described for use in industrial irradiation processes with a conveyor system would be superfluous. Such a device would further confound the method for irradiating the small items with their necessary dosage. A batch system appears to be a more satisfactory solution. Latham and Orvis[66] proposed a design for a batch system having a capacity of twelve to sixteen mattresses per day, depending upon the size and material of the mattresses and the number of cycles employed. Figures 10.27 to 10.30 show the sketch of core and elevation, plan and detail drawings, respectively, for the irradiator.[66]

The irradiator consists of 3 horizontal drawers for containment of the material to be irradiated. The middle drawer also contains 4 cylindrical ports to receive 4 Co^{60} cylindrical sources.

The source is 16,000 curies of Co^{60} and is designed with sufficient activity and geometry to provide four cylindrical cores (4000 curies each) which will deliver 4.5 Mrads in 14 hr. There are three gaps between the cylinders which will deliver 120 krads in the same period, and two mattress-sized drawers which receive 200 krads (necessary for control of staph) in 12 hr.

The track-mounted drawer is made of open mesh stainless-steel and features four 36 in. long semicylindrical baskets rigidly attached to the drawer front, and three flat areas (between

baskets) of about 20 in. by 36 in. (see Fig. 10.27). This construction allows the source cylinders to slide over the core baskets.[66]

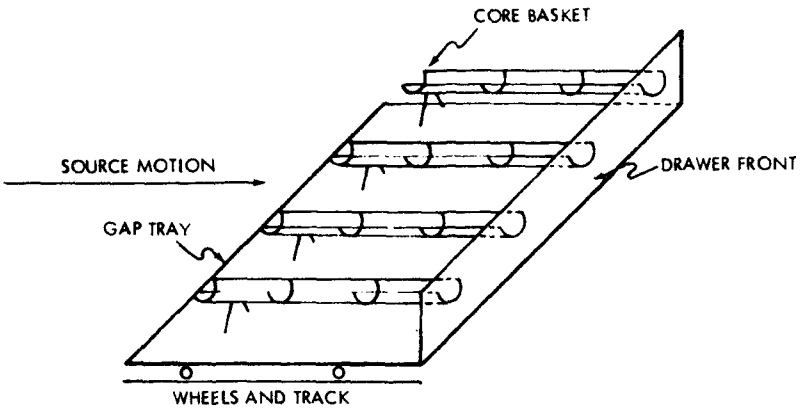


Fig. 10.27—Sketch of horizontal drawer for source core of hospital-mattress irradiator.[66]

The surgical instruments, bones, or other homograph material would be encapsulated in aluminum containers of about 3 in. outside diameter and of convenient length and placed in the semicylindrical core baskets. Larger items would be placed on the flat areas of the drawer. The drawer would then be pushed into the irradiating area[66] to await the arrival of the source (see Fig. 10.30).

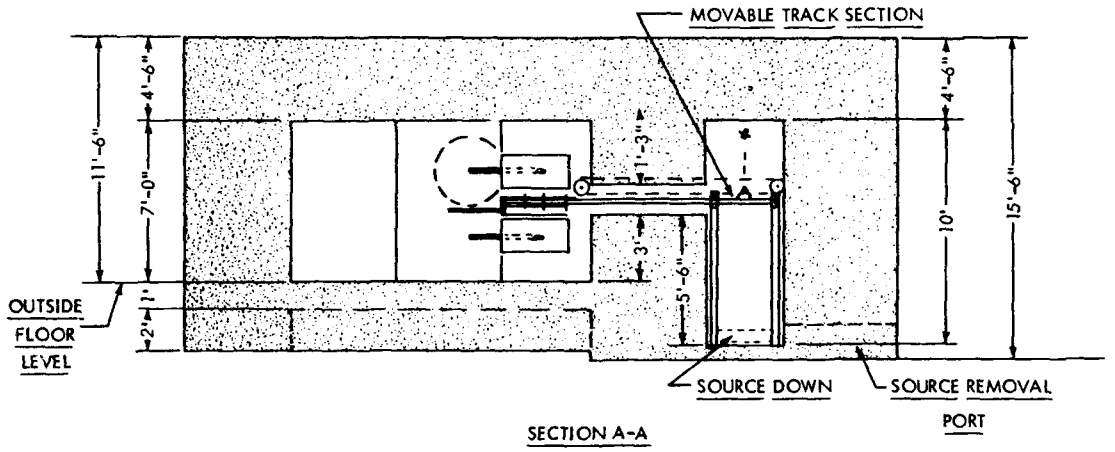


Fig. 10.28—Elevation sectional view of hospital-mattress irradiator.[66]

The matress baskets, which can also be used for other bulky items, would be utilized as follows: The baskets are constructed of thin aluminum sheet, or open mesh stainless-steel. The front and one-half of the top are hinged for ease in loading. The baskets are suspended on drawer rails like the core tray, and can be rotated about a pivot on the drawer glide when the basket is pulled out to the end of the track. The pivot can be locked to prevent the basket from rotating while it is in the irradiation chamber. Because the baskets are as close to the source as possible, only one basket can be rotated at a time, but this is all that is necessary.

In operation, matresses or other articles would be placed in paper wrappers (to prevent recontamination after leaving the sterilizer) and placed in the drawers. The drawers accommodate three cotton mattresses or four foam rubber mattresses per load. After the source is brought into position the contents of the drawers are irradiated until the innermost mattress has received-one-half the required dose. At this time the source is removed, the drawers are rotated, the source is returned, and the half cycle is repeated.

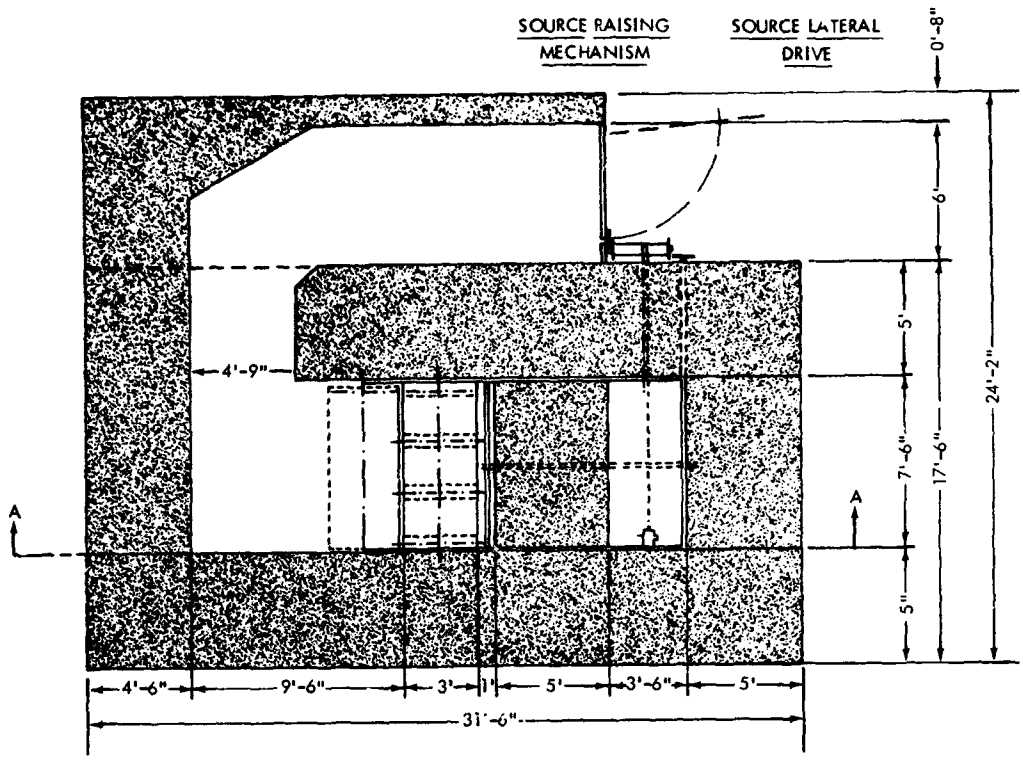


Fig. 10.29—Plan sectional view of hospital-mattress irradiator.[66]

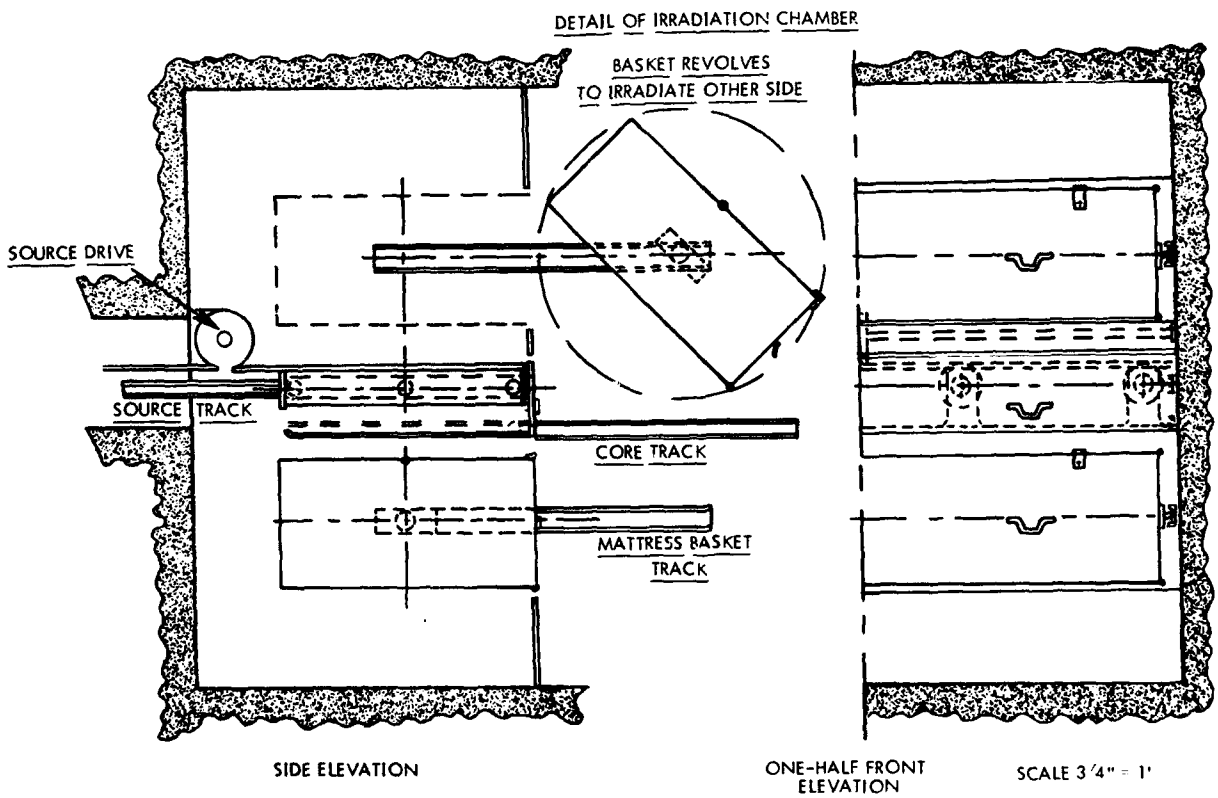


Fig. 10.30—Details of operation of hospital-mattress irradiator.[66]

In present practice packets which are to be steam sterilized are sealed with a special tape that reveals dark brown stripes after being steamed. A similar tape such as blue cellophane could be used that would indicate the radiation dose received.

Cost Estimate for a Hospital Mattress Irradiator

Latham and Orvis[66] estimated the cost of the hospital mattress irradiator to total $58,545 for capital outlay and $123 per day for operation (see Table 10.12).

At a daily operational cost of $123.00, use of the irradiating facility for the sole purpose of matress sterilization would cost $7.50 or $10.00 per mattress, depending upon whether cot-

Table 10.12—COST ESTIMATE FOR HOSPITAL MATTRESS IRRADIATOR[66]

Construction Cost	
Concrete construction, including forms, steel reinforcement, labor	$16,000
Fabrication of rear wall plugs	400
Fabrication of drawers	500
Drawer tracks and extenders	300
Hoist and drive	500
Wooden door	80
Interlock	65
Monitoring equipment	2,000
Electrical installation, lighting, wiring	400
Labor, other than concrete construction	1,200
Subtotal	$21,345
Source Cost	
Fabrication of frame	$ 750
Shipping cask	750
Co^{60} at $2.15 per curie encapsulated	34,400
Shipping and handling charges	800
Installation and testing	500
Subtotal	$37,200
Grand Total	$58,545
Operating Cost–Yearly	
Labor, operation	$22,400
Source replenishment at rate of 2,000 curies per year—includes transportation and reloading charges	6,000
6% interest on initial outlay	3,300
Amortization of initial outlay over 10 year period	5,850
Sterilizing supplies, tape, containers, wrappers	900
Repairs and maintenance	300
	$38,750
Daily operational cost (6 days per week) = $123.00	

ton or foam rubber were irradiated. However, with the use of the source cores and gaps between the cores for bone and other tissue, instrument, bandage, mask, gown, and plastics sterilization, the over-all cost per item would be considerably reduced. The controlling criterion in a sterilization method is not the immediate cost. The significant question is: "Does it do the job within reasonable financial limits?" Other financial considerations not as readily evident are in the cases of postoperational infections due to infectious plastic valves used in surgery as described previously, or an unsuccessful homograph procedure. These cases might be fatal

or might require repeated surgery involving considerable expense. Gamma radiation sterilization also could increase the useful lives of many electrically wired instruments and keen-edged surgical blades. In addition, if staph infection incidents are reduced to any degree, over-all sterilization expenses would be reduced by a decrease in volume, and shorter hospitalization periods could be expected for patients who are now potential staph cases.

With such varied materials to be irradiated, efficiency is extremely difficult to determine. The most that can be said is that the facility is designed for round the clock operation with all drawers filled at all times, making optimum use of the radiation available.

Taking all of the above mentioned benefits into consideration, a daily cost of $123.00 does not seem prohibitive.[66]

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Chapter 11

ELECTRONIC DETECTION AND MEASUREMENT OF RADIATION

Soon after the discovery of radioactivity, attempts were made to use the ionization produced by radiation as a method of measuring the intensity of the radiation. Calorimetric methods were used as early as 1897 and simple ionization chambers were used by Madame Curie in 1898 and by Rutherford in 1900. After the beginning of this century, detection methods based on chemical reactions induced by radiation were tried. The fluorescence and scintillation of certain materials when irradiated were also investigated.

Photographic emulsions, used so extensively today for film badge dosimeters, were developed as a quantitative method of detection[1] as early as 1905. From then until World War I, the principal development in radiation detection occurred in instruments based on the ionization of gases. During this period the effect of radiation on electrical resistance and a number of biological effects were explored as possible methods of radiation dosimetry. The development of radio and electronic circuits and reliable mass-produced electronic parts encouraged the continuing development and use of electronic methods of measuring the ionization of gases.

This chapter discusses devices used for electronic detection and measurement of radiation, including ionization chambers, condenser dosimeters, proportional counters, Geiger-Müller counters, neutron counters and scintillation counters. These devices have various ranges of sensitivity. Those that measure radiation dose rate may vary in sensitivity from a few milliroentgens per hour to several hundred roentgens per hour, and "thimble" chamber ratemeters may be used for dose rates of several hundred thousand roentgens per hour. Those that measure radiation dosage may range from a fraction of a milliroentgen to a few thousand roentgens.

The above-mentioned devices, except for the thimble chamber, are not usually suitable for measuring dose rates in the range of 10^4 to 10^8 r/hr or for measuring dosages in the Mrad range. These are, however, the ranges of greatest interest in studying industrial uses of gamma radiation. For the highest ranges, chemical dosimeters (ferrous sulfate or ceric sulfate), special films (such as polyvinyl chloride and cellophane), and special glass dosimeters have been found useful.

IONIZATION OF GASES

High-speed electrons, alpha particles, or photons passing through a gas may, if they have sufficient energy, eject an electron from the electron cloud surrounding a gaseous atom or molecule (see Absorption of Gamma Radiation, Chap. 2). This interaction produces a pair of charged particles known as an ion pair. (This is not to be confused with pair production resulting in the formation of an electron and a positron by the annihilation of an energetic gamma photon described in Chap. 2.) The negatively charged particle of the pair of ions is an electron, and the atom or molecule of gas losing the electron becomes a positive ion.

The "ionizing energy," sometimes referred to as the ionizing potential, is the energy required to remove the most loosely bound electron in the gas molecule. Measurements have shown that the average energy absorbed in air per ion pair produced is considerably greater than the average ionizing energy of the gas. Table 11.1 gives a comparison of the ionizing energy of some gases with the average energy loss per ion pair produced.

Table 11.1—IONIZING ENERGY AND ENERGY LOSS PER ION PAIR FOR VARIOUS GASES[2]

Gas	Ionizing energy, ev	Energy loss per ion pair, ev
Hydrogen	13.9	32
Helium	24.5	28
Nitrogen	15.7	36
Oxygen	12.7	33
Argon	15.7	25

Some collisions of high-speed charged particles with gas molecules produce excited (more energetic) atoms or molecules without producing ionization. This in part explains the fact that the energy loss per ion pair produced is appreciably greater than the ionizing energy. In the case of the heavier gases, some of the electrons ejected may come from an inner shell where the binding energy is appreciably greater than that of an electron ejected from an outer shell. The average energy necessary to produce an ion pair is about 32.5 ev. Thus a gamma photon with an energy of 1.0 Mev will produce in air about 1.0×10^6 ev/32.5 ev per particles, or 3.08×10^4 ion pairs.

High-speed charged particles produce ionization along their paths by interaction between the electric field associated with the charged particle and the electric field of the atomic electrons. Force between two charged particles varies inversely as the square of the distance between them. A high-speed charged particle having sufficient kinetic energy can eject an orbital electron by means of a transfer of energy between the electric fields of the moving particle and of the electron. This transfer of energy will slow the original charged particle and may deflect it from its path. Many ionizations may be produced by a single high-speed electron, and its path will be tortuous as it ionizes one atom after another. Because most positively charged particles are much heavier than electrons, there is little deflection of these particles during the production of ion pairs.

Gamma rays and X rays first interact by one of the three mechanisms described for the absorption of gamma radiation (photoelectric effect, Compton scatter, and pair production). The high-speed electrons resulting from these processes have considerable kinetic energy and can themselves produce ionization.

PRINCIPLE OF OPERATION OF IONIZATION CHAMBER

Ionization chambers are among the most useful devices for the measurement of radiation dosages. A gaseous ionization chamber consists of a set of electrodes in an enclosed volume of gas. A potential difference applied between electrodes produces an electric field in the gas. When ionizing radiation penetrates into the contained gas, electrons and positive ions are formed by the interaction of the radiation with the molecules of the gas. Under the influence of the electrical field these charged particles migrate through the gas to the electrodes.

The design and construction of ionization chambers depends a great deal on the type of ionizing radiation to be measured. If alpha radiation is to be measured, very thin windows must be used to admit the particles into the gas-detecting volume so that the particles will not be absorbed in the chamber walls. Thin mica, or nylon stretched to 0.1 mil in thickness, has been found satisfactory for this purpose. The window need not be so thin if beta particles are to be measured. However, as high-speed electrons may have a relatively long range in the

gas, they may pass completely through the ionization chamber without expending all their energy in ionizing the gas. If a heavy gas such as argon is used to fill the chamber, the range will be decreased and more energy will be expended in the gas.

The probability of absorbing penetrating gamma radiation in an ion chamber may be quite low. The radiation enters the chamber readily, but it and the high-speed secondaries resulting from interaction are not readily absorbed by the gas in a chamber of limited size.

A variety of gases is used in ionization chambers, but air is used most frequently. Chambers designed to operate with detection equipment that measures the time rate of accumulation of charge usually are filled with a gas of low molecular weight. Oxygen, water, carbon monoxide, and the halogens have been used. The probability of interaction in the chamber volume can be increased by pressurizing to increase the gas density and by using the dense gases argon or krypton.

EFFECT OF VARYING THE ELECTRIC FIELD

Ideally the charge collected at the chamber electrodes should exactly equal the total charge formed by the interaction of radiation with the gas molecules in the sensitive volume of the ionization chamber. At least two effects tend to prevent this. The first is the normal thermal diffusion of ions, which will tend to cause some of the ions to diffuse into the chamber end walls away from the collecting electrodes. The second effect is recombination of an ion and electron to form a normal molecule, which may take place before the ions are collected by the electrodes. Each of these effects can be inhibited by making the collecting electric field high and thereby collecting the ions more rapidly. However, the voltage should not be so high that additional ions are formed by the fast migration of the original ions to the electrodes; otherwise, the linear relation between primary ionization and the charge collected at the electrodes may be lost. Figure 11.1 shows the relation between the applied voltage and the charge collected for a constant radiation intensity at the chamber.

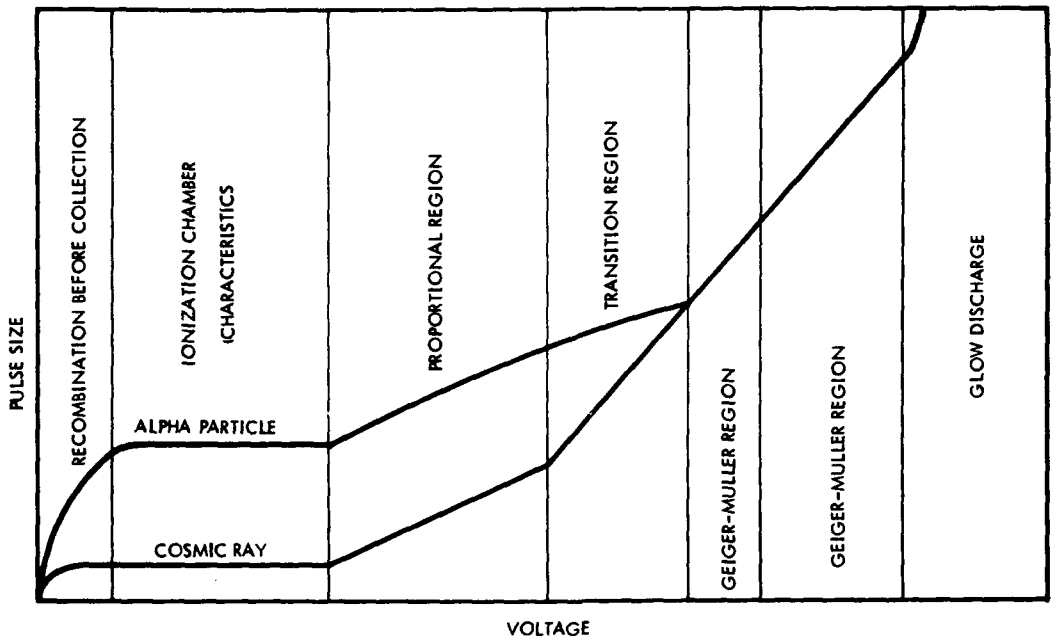


Fig. 11.1— Effect of voltage on charge collected at electrode of ionization chamber.[3]

After reaching a certain minimum voltage the charge collected is constant for a considerable range of higher voltages. This makes the equipment insensitive to slight voltage changes over a reasonable voltage range. Operation in the region where the charge collected is equal to the primary ionization produced by the radiation distinguishes ionization chambers from other types of counters.

At still higher voltages the electrons produced by the primary ionization acquire sufficient energy between collisions in the stronger electric field to produce additional ions which results in the "proportional" region. The number of additional ions produced per original primary ion depends on the voltage and increases with increasing voltage, reaching a maximum of about 10^4.

As the voltage is increased beyond the proportional region, the multiplication factor becomes so great that a single primary ionization produced by radiation causes a cascade of additional ionization, resulting in multiplication factors as high as 10^{10}. Because of these multiplications, the total ionization is no longer proportional to the original primary ionization, but depends only on the operating voltage. This region of operation is called the "Geiger" region. This large multiplication makes the Geiger tube very sensitive to low levels of ionizing radiation but useless for measuring high levels of radiation. With high levels of radiation, primary ionizing events occur so frequently that there is insufficient time between primary ionizations for the tube to recover and function as a counter of the individual events. Thus, ionization chambers are more satisfactory in measuring radiation fields of high flux than are detectors which depend on gas multiplication.

The range of operating voltage for an ionization chamber can be determined by gradually increasing the voltage. The measured response will increase from zero to a fixed value. The voltage may then be increased over an appreciable range without further increase in response until multiplication occurs, indicating the beginning of the proportional region.

ELECTROSCOPES AND CONDENSER TYPE DOSIMETERS

The first radiation measuring device to make use of ionization was the gold-leaf electroscope. This was the first instrument that could be used to estimate radiation dose quantitatively. The early form of the instrument consisted of two gold leaves suspended in a container by an electrode which was electrically insulated from the walls of the container.

When a charge was applied to the electrode, the gold leaves separated and the degree of separation was measured with a scale. With even the best quality electrical insulation the leaves would not remain spread but would slowly return to their original position, indicating a loss of the applied charge. This occurred because of cosmic radiation and background radioactivity which caused ionization of the air in the electroscope. These ions migrate either to the gold leaf or to the container wall, depending on their charge, and slowly neutralize the original charge. Although the gold-leaf electroscope, with the leaves serving as an electrostatic voltmeter, is a complete instrument, it is not very accurate.

If the voltage is determined by an external electrostatic voltmeter, greater accuracy is possible. The gold leaves may then be replaced with a larger capacitance. An instrument of this type is simply a condenser with a gas dielectric. By charging such instruments, and then accurately measuring the charge remaining after an exposure to ionizing radiation, a reliable indication of the dosage of ionizing radiation is obtained. The loss in charge is dependent on the number of ion pairs produced, which is a measure of the dosage of ionizing radiation. Such instruments can be made in small sizes. They are useful as pocket or ring dosimeters to measure the accumulated dosage of personnel working in a radiation field. Figure 11.2 shows a sectional view of such a condenser type pocket ion chamber.

Referring to Fig. 11.3, the chamber C is first inserted in charging unit A and charged to potential E by depressing button B. The chamber C is now removed and exposed to the radiation dose that is to be measured. The chamber is next reinserted in the charging unit and the change in potential is measured by the electrometer S. In this case a string type electrometer is indicated; however, other types can be used. In the string electrometer the deflection of the string is observed with a microscope on a scale calibrated in roentgens. The scale may be on the ocular, or the image of the string may be projected onto a calibrated scale. A cap is usually placed on the end of the chamber unit when it is not in use to eliminate the collection of moisture and dust.

Figure 11.4 shows a finger ring type chamber (which is particularly useful when the hands must be placed in a radiation field but the body is shielded), and the reading of a pocket chamber.

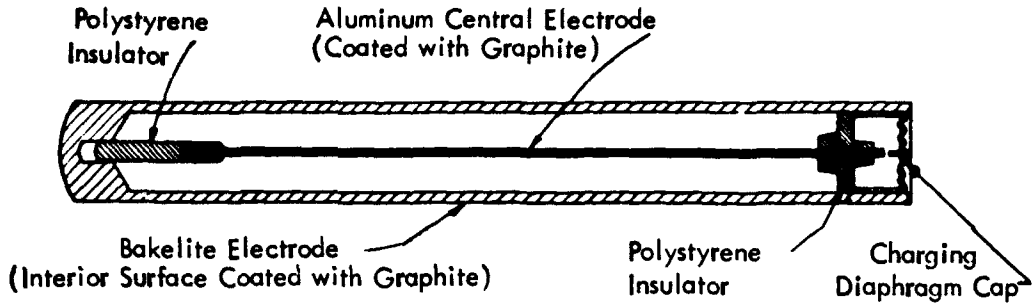


Fig. 11.2—Sectional view of condenser-type pocket ion chamber.[4]

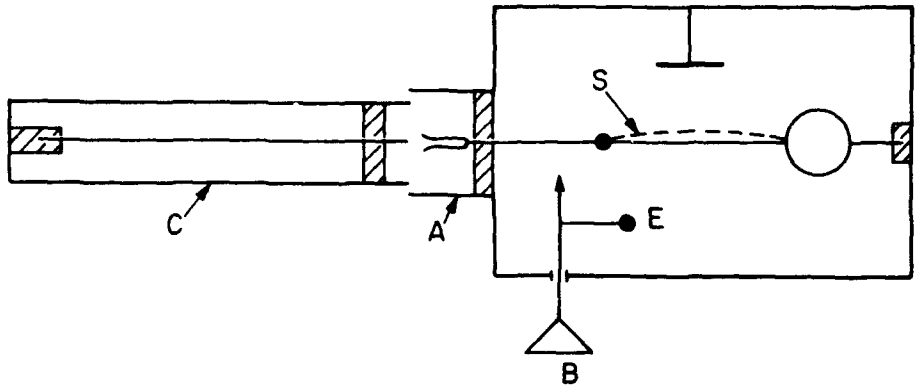


Fig. 11.3—Schematic construction of a capacitor type "r" meter.[5]



Fig. 11.4—Reading condenser type pocket ionization chamber.

Another electroscope that represents an improvement in the old gold-leaf instrument was developed by the Lauritsens in 1937, and is known as the Lauritsen electroscope. This instrument uses a fine quartz fiber about 6 mm long that is metal-coated and mounted in contact with a rigid horizontal wire. This combination of fiber and wire replaces the leaves of the gold-leaf electroscope. As in the earlier instrument, the electrode which holds the fiber and wire is insulated from the container walls. The electroscope is usually charged by connecting it to a d-c voltage scurce of 100 to 200 volts. The induced charge produces a repulsion between the wire and the fiber, causing a deflection of the fiber. If the instrument is exposed to ionizing radiation, the charge will be slowly dissipated by the collected ions and the fiber will return to its original position. The displacement of the fiber is approximately proportional to the remaining charge, and the amount of charge lost is proportional to the dosage of ionizing radiation. Thus, the instrument may be used as a direct-reading dosimeter by means of a magnifying lens and a scale calibrated to read the deflection of the fiber in units of radiation dosage. Direct-reading pocket dosimeters operate on this principle and are very useful in keeping a continuous check on accumulated dosage. Figure 11.5 is a sectional view of a self-reading quartz-fiber pocket dosimeter that operates on the principle of the Lauritsen electroscope.

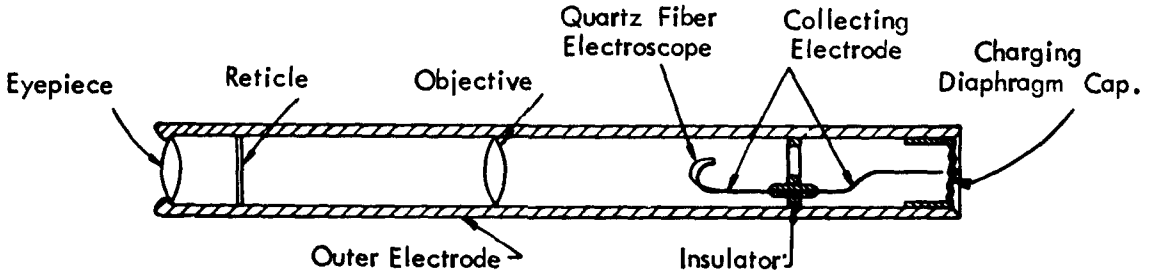


Fig. 11.5—Self-reading quartz-fiber pocket dosimeter.[4]

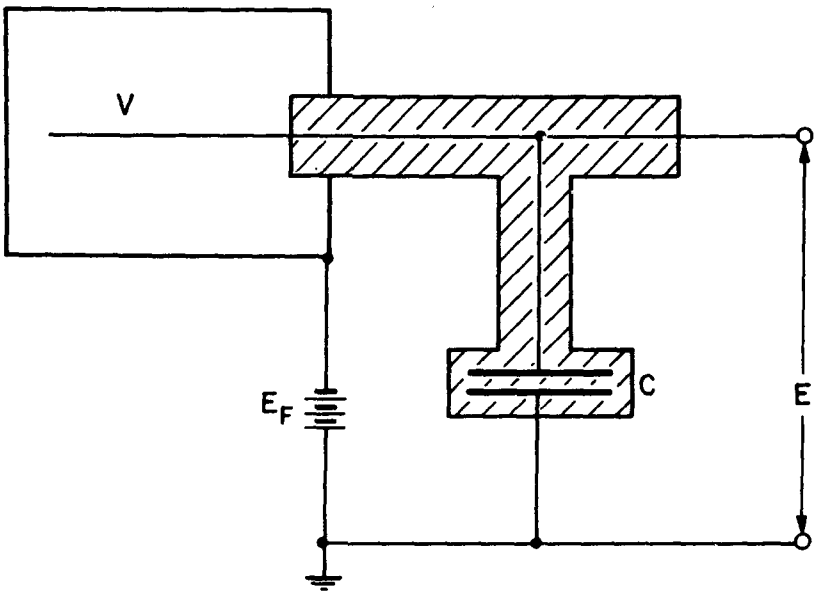


Fig. 11.6—Basic circuit used with practical ionization chambers.[5]

CONDENSER TYPE METERS

The basic circuit on which ionization chamber instrumentation is based is shown in Fig. 11.6. Here C is the capacitance of the system, V is the gas volume, and E is the potential change in volts occurring across the capacitor owing to radiation.

Connecting the chamber to an electrometer or amplifier completes the instrument. The ions in the gas which are produced as a result of interaction with radiation migrate under the influence of the electric field to the chamber wall and center electrode where they are collected to produce an ionization current. This current is a measure of the ions produced and thus of the original radiation. This causes a change in the voltage across the capacitance. This change in voltage E is directly proportional to the charge Q and inversely proportional to the capacitance of the condenser C as given by

$$E = \frac{Q}{C} \tag{11.1}$$

where E is in volts; Q, in coulombs; and C, in farads.

The charge is also directly proportional to the radiation dose r and inversely proportional to the volume of air v, and depends on pressure and temperature as indicated by

$$Q = \frac{r}{v}\,\frac{p}{760}\,\frac{273}{273 + t} \tag{11.2}$$

where Q is in electrostatic units, r is in roentgens, v is in cubic centimeters, t is in degrees centigrade, and p is in millimeters of mercury.

Equation 11.2 is based on the original statement of the roentgen unit by Villard in 1908 as the "unit of quantity of X radiation that by ionization liberates one electrostatic unit of electricity per cubic centimeter of air under normal conditions of temperature and pressure." At standard conditions, p is equal to 760 mm Hg pressure, and t is equal to 0°C. Therefore, as there are 3×10^9 esu per coulomb, the relation between dosage and voltage drop at standard conditions of pressure and temperature becomes

$$r = \frac{vQ}{3 \times 10^9} = \frac{vEC}{3 \times 10^9} \tag{11.3}$$

An unusual feature of ionization-chamber circuits is the spurious ionization current picked up by the highly insulated circuit. Since the circuit cannot usually be shielded from penetrating radiation, it must be made ionization-proof by enclosing with high-quality insulation as indicated by the shaded area in Fig. 11.7. Ordinary varnishes and waxes are unsatisfactory. Amber and pure polystyrene are preferred as insulators. Glass treated to prevent surface leakage and silicone preparations are also used.

Ionization chambers are sensitive to all types of ionizing nuclear radiations and X rays, and their use has been sufficiently successful to make this a primary standard of measuring radiation dosage. Dry air of uniform composition and at atmospheric conditions is a convenient reference. Standard instruments have been developed to measure the dosage from a collimated X-ray beam. The most common of these instruments is the parallel-plate or "free-air" chamber described by Victoreen.[5] A schematic diagram of such a chamber is shown in Fig. 11.7.

In Fig. 11.7 the volume of a standard air chamber is defined electrostatically to eliminate the effects of the wall. The volume is equal to $\pi r^2 H$ in which H is the length of the collector plate and r is the radius of the aperture D_2. Apertures D_1 and D_3 are used to collimate the beam from a source T.

A potential of about 1500 volts is maintained between plates G-H and E_2. The guard plates G serve to eliminate fringing of the electric field between plates E_2 and H. For this purpose guard plates G must be appreciably longer than the collector plates H, otherwise, the electric field will be curved as indicated at B. The guard wires W aid in preventing distortion of the field. At spaced intervals the guard wires are connected to the potential divider R, which maintains a uniform potential gradient. Although Fig. 11.7 shows guard wires only at the inlet end of the chamber, the same construction is used at both ends of the chambers of standard instruments to align the electric field perpendicular to the plate H. The field will also be distorted if the potential between G and E_2 differs from that between H and E_2. To prevent

this, a "null" type of circuit is used which maintains E_0 at zero. The potential E_0 is indicated by a string electrometer with a zero at center. As electrons are collected at capacitor C, a compensating potential E_C is added to maintain $E_0 = 0$. At the end of the exposure, E_C is equal to E of Eq. 11.3 and gives the radiation dosage.

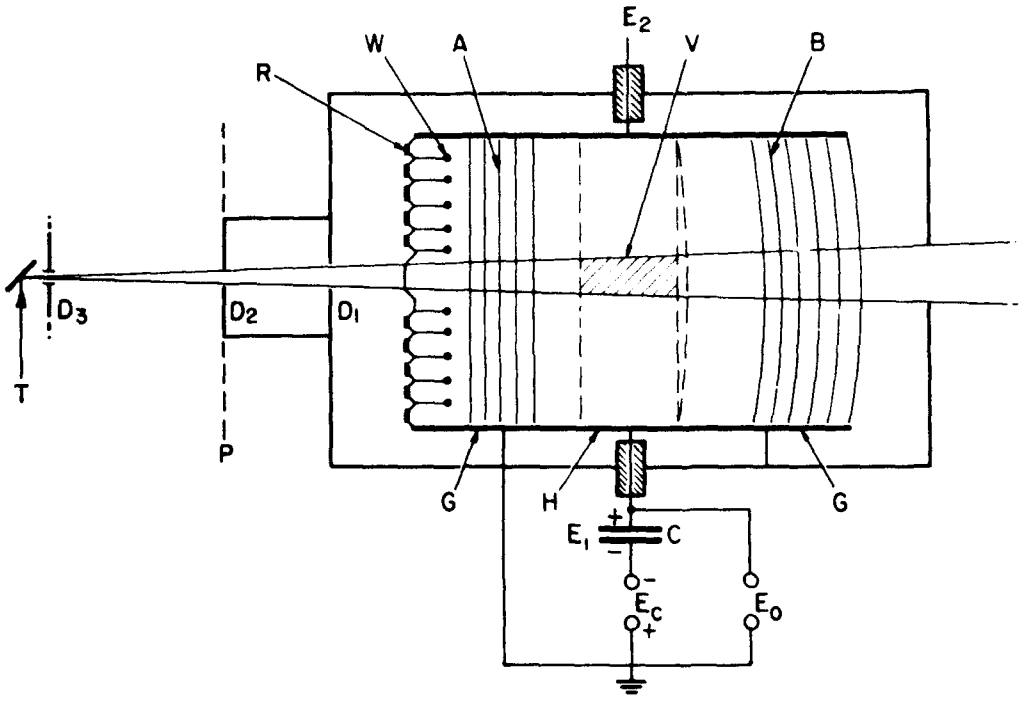


Fig. 11.7—Schematic construction of a primary standard air chamber as used for determining the International Roentgen.[5]

The plates E_2 and G-H are spaced a sufficient distance apart to avoid collecting scattered electrons before they interact with the air in the chamber. The spacing increases as the wavelength of the measured radiation decreases and becomes excessively great with ionization radiation of very short wavelength. This limits the use and practicability of standard air chambers. However, such standard chambers are useful for accurate calibration of other instruments. The first international definition of the roentgen in 1928 was based on the use of a standard air chamber: "The roentgen shall be that quantity of X radiation, which, when the secondary electrons are fully utilized, and the wall effect avoided, produces in one cubic centimeter of atmospheric air, at 0°C and 76 cm mercury pressure, such a degree of conductivity that one e.s.u. of charge is measured under saturation conditions."

Equation 11.3 is rigorous if air is the sole source of ionization. Thus, a chamber with a large volume of air and a minimum of wall effects obeys the relation expressed by Eq. 11.3 more closely than a smaller chamber with greater wall effects. However, large ion chambers are unwieldy and are not satisfactory for use in portable meters. The relations expressed in Eq. 11.3 may be approximately obeyed by small chambers fabricated of material having the same radiation "stopping power" per unit mass as air. This requirement can be met if the wall material of the chamber has an atomic number similar to that of the gas used in the chamber. The stopping power of an absorber for gamma radiation depends primarily on the atomic number of the absorber.

Absorption in air may be simulated by making the walls of the chamber from a material having an atomic number close to that of air but having a much greater density. Carbon has an atomic number of 6, which is comparatively close to that of oxygen, 8, and nitrogen, 7. Therefore, the wall of the chamber may be made of a plastic such as bakelite, which contains a high percentage of carbon. A thin wall of bakelite may be considered to be approximately equivalent to a much greater thickness of air because of the closer packing of the molecules, and a very small bakelite chamber may be approximately equivalent to a large standard air chamber.

ION-CHAMBER DOSE-RATE METERS

The collection of charge on the electrodes of an ionization chamber results in the flow of a small amount of current. If a battery or other source of voltage is kept connected to the ionization chamber during exposure to radiation, the potential can be maintained constant instead of being changed as in the case of the electroscope. The small current resulting from the collection of ions can be measured by a sensitive electrometer or a microammeter. The primary current strength of such an ionization chamber is directly proportional to the rate at which ionizing radiation enters the ionization chamber. A portable ionization-chamber rate meter with a pistol grip was dubbed "cutie-pie" during the operation of the Manhattan project and this name has continued to be used. Figure 11.8 is a photograph showing a cutie-pie type meter being used to survey the radiation field at a waste disposal area at the Hanford Atomic Products Operation.



Fig. 11.8—"Cutie-pie" type portable radiation survey meter used to monitor radiation field at a waste disposal area of the Hanford Atomic Products Operation. (Courtesy of General Electric Co.)

This type of meter is used for measuring beta and gamma radiation in "hot" laboratories. The scale reading can usually be multiplied by factors of 10 or 100 by a scale button on the top or face of the instrument. Typical full-scale readings are 15, 150, and 1500 mr/hr. The power supply for such an instrument is usually a number of dry cells. A cross-section pistol type portable dose-rate meter of the design described by Taylor and Sharpe[7] is shown in Fig. 11.9.

The meter circuit shown in Fig. 11.10 may use a total of 11 small batteries consisting of 6 of $1\frac{1}{3}$ volts, 2 of 15 volts, and 3 of 30 volts. The approximate life of the group of batteries is

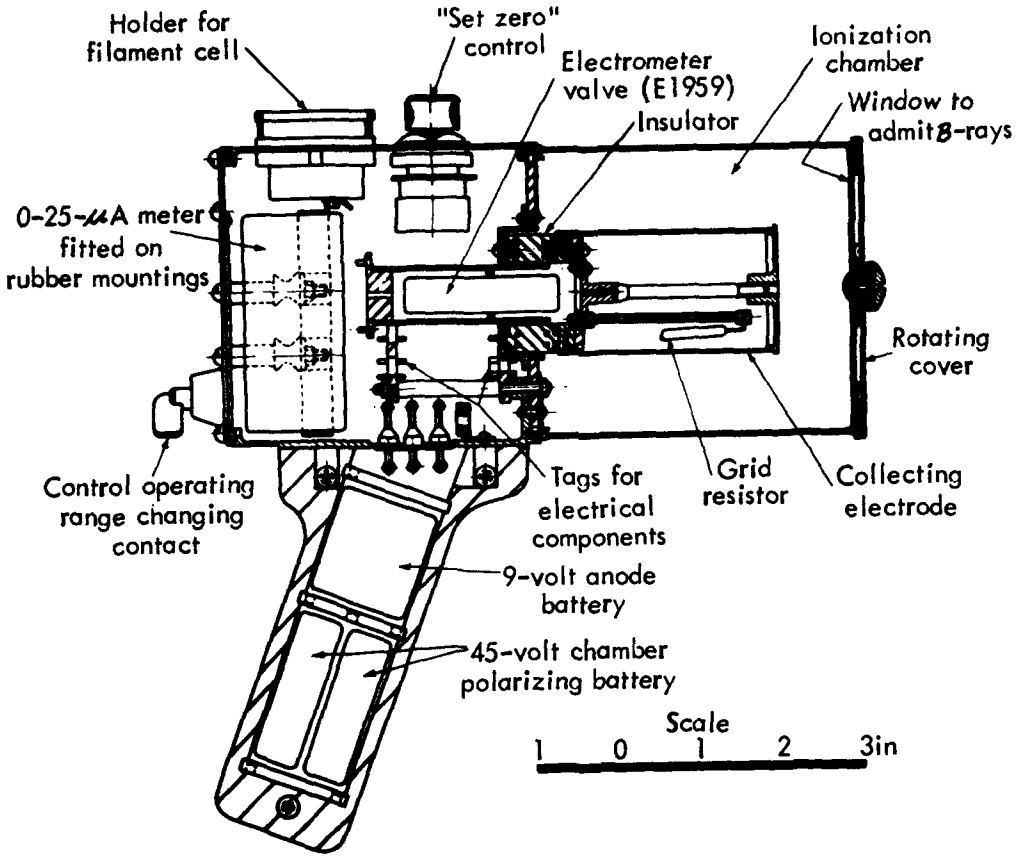


Fig. 11.9—Cross-section of pistol type portable dose-rate meter. (Courtesy of *Atomic Energy Research Establishment.*)[7]

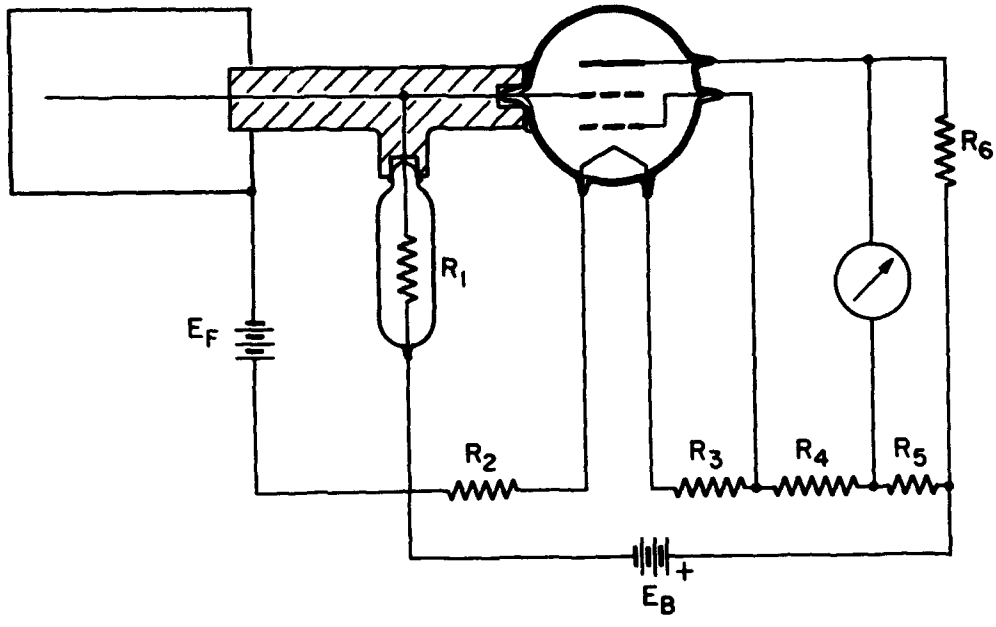


Fig. 11.10—Conventional tetrode electron-tube circuit as used with an ionization chamber as a rate meter.[5]

800 hr of meter use or one year of shelf life.[6] Two vacuum tubes (one tetrode and one pentode) are used in the circuit to multiply the current from the ionization chamber associated with the instrument. The ionization chamber of the meter shown has bakelite walls $\frac{1}{8}$ in. thick, coated with "aquadag" (graphite). The chamber volume is 400 cm^3 and a pliofilm "window" 0.00025 in. thick permits entry of low-energy beta-particle radiation into the chamber. A rotary shield can be used to close the "window" to permit measurement of the gamma radiation component in the presence of a beta-ray background.

MEASUREMENTS WITH DOSE-RATE METERS

Two methods are used to measure the ionization produced by radiation. Either the rate of change of voltage as the charge is collected, or the flow of collected charge through a resistance, may be measured. If a high resistance is placed between the electrodes, the relation for a constant rate of ionization is

$$E = neR(1 - e^{-t/RC}) \tag{11.4}$$

where E = change in potential due to ionization
n = number of ions formed per second
e = electronic charge
R = effective circuit resistance
t = time of exposure to radiation
C = capacitance of electrode system

If t is very large with respect to the quantity RC the relation reduces to

$$E = neR \tag{11.5}$$

The movement of the ions to the electrodes of an ionization chamber produces a very small electrical current. If the chamber is used to measure a radiation field of 1 r/hr, the primary current predicted by Eq. 11.3 is

$$I = \frac{400\ cm^3}{3 \times 10^9\ (3600)} = 3.7 \times 10^{-11}\ amp$$

The magnitude of the current may be amplified by the use of vacuum tubes to the point where a meter will give a visual indication of the amount of radiation absorbed in the chamber.

A simple triode vacuum-tube circuit may be used for such amplification. The triode contains a filament, a grid, and a plate. The hot filament emits electrons which migrate to the plate as a result of a potential between the filament and plate. The grid is a wire-mesh element placed between the filament and plate so that a very slight change in input to the grid will produce a much greater change in the flow of electrons from the filament to the plate. Typically a change of one volt on the grid may produce a change in the plate current equal to that produced by a hundredfold change in plate voltage. In this case, the tube is said to have an amplification factor of 100. However, ordinary triode tubes are not very satisfactory for use in ionization chambers because they have a grid current of about 10^{-10} amp, which may completely mask the small current of an ion chamber. Special vacuum tubes called electrometer tubes, which have grid currents of only 10^{-15} amp, give more satisfactory operation with ionization chambers.

The basic circuit can be improved considerably by the use of inverse feedback to permit extremely stable operation independent of variations in tube characteristics or battery voltage.[6] A large reduction in the input time constant may also be obtained, resulting in faster meter response.

The current from the ionization chamber passes through the input high-megohm grid resistor and develops a signal voltage across the resistor. This signal is amplified in the tetrode tube, the plate of which is directly coupled to the grid of the cathode-follower tube which provides the output for the 100-μamp meter and the feedback voltage.

Since 1958 the size and weight of many electronic instruments have been decreased by replacing electron tubes with transistors. However, the basic principles of the circuits used are the same in most cases.

THIMBLE CHAMBERS

The accurate measurement of the ionization produced in an ion chamber by gamma radiation involves the requirement that all the electrons capable of ionization be absorbed either in the gas or in an air equivalent wall. Absorption entirely in the gas is difficult to accomplish in a chamber of limited size because of the penetrating nature of gamma radiation and the high speed of the electrons from secondary radiation.

The Bragg-Gray principle reported by Bragg[8] in 1912 and developed in the 1930's by Gray[9] and others brought a new approach to the problem of dosimetry. This principle involves the determination of absorption of the energy of radiation in a medium (usually solid) in terms of the physical measurement of ionization in a very small volume of gas held in a cavity within the solid. The relation describing this principle is stated mathematically by Eq. 11.6.

$$E = SJW \tag{11.6}$$

where E = energy absorbed per unit volume per second in wall material (ergs or mev/cm^3-sec)
S = ratio of stopping power for secondary particles in wall material to stopping power in gas
W = average energy absorbed from secondary to form one ion pair in the gas
J = number of ion pairs formed in the cavity per unit volume per second

This simple relation given in Eq. 11.6 is valid only under the following conditions:

1. The thickness of the chamber wall must be equal to or greater than the maximum range of the secondary particles in the wall material.
2. The chamber must be sufficiently small so that the dimensions are less than the average range of the secondary electrons in the gas.
3. The primary radiation should not be reduced appreciably in passing through the radiation chamber.
4. The relative stopping power should be independent of the velocity of the secondary particles.

These limitations show that for Eq. 11.6 to be valid the intensity of secondary electron radiation in the chamber must exactly equal its intensity within the wall material. In chambers of small size the ionization per unit volume is independent of the dimensions, so small chambers are desired.

The presence of the wall material alters to some extent the direction and distribution of the secondary electron radiation. It is also generally true that the ionization produced in the gas is not exactly equal to the ionization produced in the approximate equivalent volume of wall material. However, these factors are not significant if the wall material has the same atomic number as the gas.

Although the current produced by the ionization of gases in the thimble chamber is the means of measuring the radiation dose rate, this instrument is not simply a version of the ionization chamber. The thimble chamber is a different kind of instrument about which Parker[1] states, "the wall material is the important medium and the nature of the ionized gas is subordinate." In fact, Gray[9] proposed that a new dosimetry unit be used with a pure substance such as carbon as the chamber walls. Although this proposal was not adopted, the definition of the roentgen was revised in 1937 to include the use of an air-wall chamber with an air-filled cavity: "The roentgen shall be that quantity of X- or gamma-ray radiation such that the associated corpuscular emission per 0.001293 gm of air produces, in air, ions carrying 1 e.s.u. of quantity of electricity of either sign."

The bakelite thimble is usually lined with a thin graphite coating to make it a conductor. The collecting electrode in the center is well insulated, is connected directly to the detecting equipment, and is charged to the desired operating potential. The guard ring is charged to the same potential and prevents leakage across the insulator surrounding the electrode.

Thimble chambers have been used as radiation-monitoring equipment for many years. The "roentgen ratemeter" is the name given by one manufacturer to such a meter, which was originally developed for continuous monitoring of the intensity of X-ray beams, but, when suitably calibrated, is very useful for measuring gamma radiation. This instrument is reported to be independent of the energy of the radiation within ±1 per cent over the range of 30 kev to 1.0 Mev. Figure 11.11 is a photograph of the instrument and probe. The ionization-chamber preampli-



Fig. 11.11—Photograph of Victoreen "Roentgen Ratemeter" and probe. (Courtesy of Victoreen Co.)

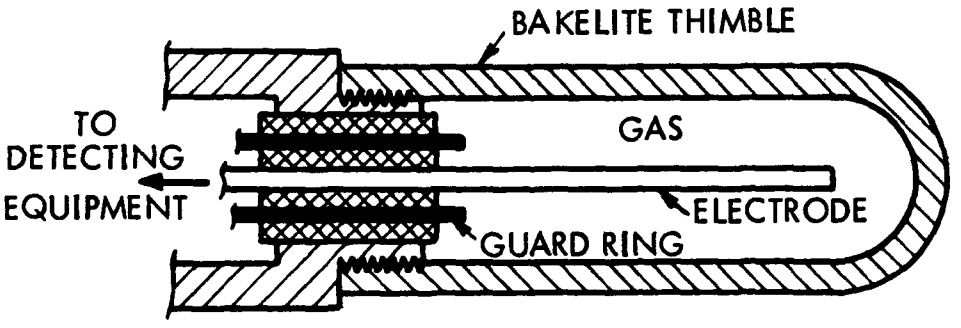


Fig. 11.12—Diagram of thimble ionization chamber.

fier probe is connected to a control unit by means of 35 ft of shielded cable. The probe is $\frac{1}{2}$ in. in diameter at the thimble end, 1 in. in diameter at the body end, and $8\frac{3}{4}$ in. long. The small size of the probe assembly and the extension cable makes it very easy to mount the probe

assembly in a variety of positions by means of clamps, ring stands, etc., for the calibration of a radiation field.

Since the solid wall of the thimble chamber absorbs radiation much more effectively than the same volume of air alone, the thimble chamber is useful in solving the problem of measuring ionization from fields with high dose rates of gamma radiation. Figure 11.12 is a sketch of a thimble ionization chamber.

A thimble type ionization chamber $\frac{3}{4}$ in. long, $\frac{1}{2}$ in. in diameter, with a nominal volume of 0.2 cm^3 is mounted on the end of the probe assembly. This chamber is of the air-equivalent type described previously, with a bakelite wall. The bakelite and the dimensions used for the chamber are selected to provide a special response equivalent to that of a standard air chamber over a range of 30 kev to 1 Mev. The walls of the thimble are positive and the central electrode is negative with a potential difference between them of 300 volts. The chamber in the thimble is filled with air which, when ionized by radiation, produces a very weak current through the ionization chamber. This minute current flows through a large resistance to produce a voltage drop which constitutes the signal to be amplified by electrometer type vacuum tubes.

The current through the ionization chamber is directly proportional to the ionization of the gas, which is in turn, directly proportional to the absorption of the gamma radiation. Therefore, the potential on the grid of the amplifier tube is directly proportional to the absorption of the radiation field.

The amplification system, controls, fuses, etc., are housed in a control unit about 8 in. square and 10 in. high. A two-stage amplifier of the balanced direct-coupled type is used with all operating voltages regulated.

Additional information on condenser type dosimeters and thimble chambers is given in Refs. 10 to 49.

PROPORTIONAL COUNTERS

The ionization chamber provides a convenient method of measuring both radiation dosage and dose rates. However, ionization chambers require the use of very sensitive electrometers or the use of high-amplification vacuum-tube equipment. By increasing the voltage across the electrodes of an ionization chamber it is possible to produce the internal multiplication as indicated by the "proportional" region in Fig. 11.1. In the proportional region the pulse is not independent of the voltage, as in the lower voltage plateau region used with ionization chambers, but increases in proportion to the voltage. In this region of operation, the electrons produced as a result of primary ionization of the gas atoms by gamma photons, alpha, or beta particles are accelerated by the high potential gradient near the positive electrode, and gain sufficient energy as a result of this acceleration to eject additional electrons from gas molecules in their path. The avalanche of secondaries gives a multiplication of ionization current by a factor between a thousand and a million. As the electrode voltage is increased, the energy of these electrons is increased so the pulse size increases with voltage.

The phenomenon may be understood more easily by considering the variation of the electric-field intensity with radius for a cylindrical ion chamber

$$E_{(r)} = \frac{V}{r \ln (r_2/r_1)} \tag{11.7}$$

where $E_{(r)}$ = electrical field intensity at any point r
V = voltage between inner and outer electrodes
r = radical distance from center
r_1 = radius of inner electrode
r_2 = radius of outer electrode

Equation 11.7 indicates that for electrodes of given radii and for a given voltage, the field strength varies inversely with the radius and, therefore, is much greater near the inner electrode, which is positive. The total multiplication of about 10^3 to 10^6 depends on the voltage and

gas pressure used. The zone of multiplication is very small, being usually a few millimeters around the anode. The volume of the multiplication zone may be increased or decreased by increasing or decreasing the voltage. If the zone of multiplication is kept small, essentially all the primary ionizations produced in the chamber by radiation will occur outside the zone and the electrons produced will give the same multiplication factor per electron regardless of the location of primary ionization. For a given voltage, gas, and pressure the electron yield at the anode then becomes proportional to the absorption of the radiation for a given radiation such as beta particles. Most proportional counters operate at less than atmospheric pressure and use a gas other than air for optimum performance. Methane with 10 to 25 per cent argon is a gas mixture commonly used. The methane renders the instrument more stable in operation and the argon increases the multiplication factor. The usual operating voltages range from about 500 to 800.

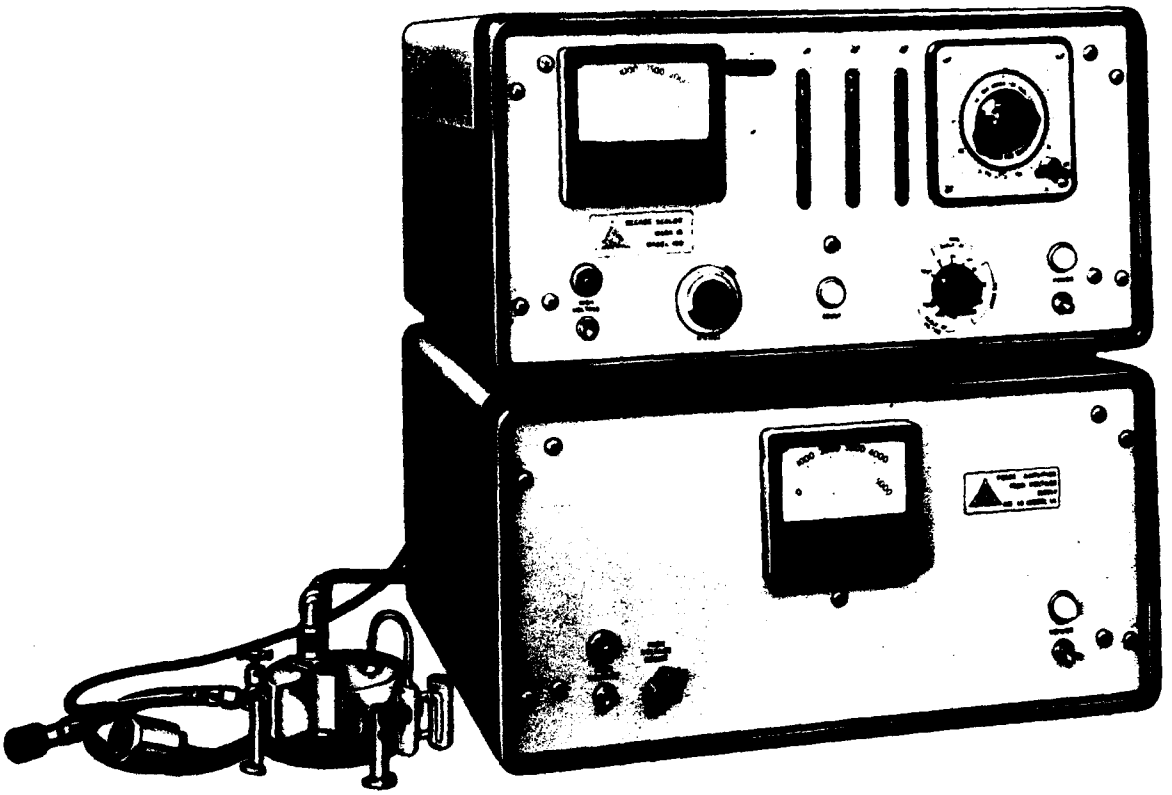


Fig. 11.13—Windowless flow counter connected to a scaler. (Courtesy of Radiation Laboratories, Inc.)

A unique characteristic of the proportional counter is the variation in the pulse height with various types of ionizing radiation as a result of the proportional amplification of the counter tube. For example, an alpha particle will give a larger pulse than a gamma photon or a beta particle. For this reason proportional counters are particularly useful in counting alpha particles in the presence of beta particles and/or gamma photons. The instrument can be adjusted so that only the larger pulses caused by alpha particle ionization are counted and beta particles and gamma photons are ignored.

Because of the large multiplication, that is, each primary ionization is followed by a cascade of ionizations near the anode giving a measurable current, it is possible to count individual ionizations. The output is sufficient to operate an electrometer, or the chamber may be connected to a vacuum-tube amplifier. The condenser type ionization chamber measures accumulated dosage and the ionization-chamber ratemeter measures the current resulting from ionization after amplification. The proportional counter produces a pulse as a result of each primary

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ionization and, therefore, measures individual ionizing events. The frequency of the pulses or the total number of pulses in a given time interval can be counted. The count rate is a measure of the radioactivity of a sample and is an essential determination in analytical techniques involving radioactive tracers. The counting may be performed by means of a timer and a flashing light, audible sound, or pulse of the indicating needle of a meter since any of these may be operated from the output of the amplification circuit. However, for convenience the counting is usually performed automatically by an instrument known as a scaler. Automatic counting is necessary when the count rate is very high and can be performed in the absence of an operator when the counting time is long. A typical scaler which might be used with a proportional counter tube was described in Chap. 6.

The "flow counter" is an instrument that can be used either in the Geiger-Müller region or in the proportional region. Only its use as a proportional counter will be discussed here. Essentially the flow counter is a shielded counter tube through which a constant flow of gas is maintained to prevent infiltration of the chamber with air. The greatest advantage of the flow counter is that the sample to be counted may be placed directly in the chamber, completely eliminating absorption in the windows required on closed tubes. For this reason these instruments are often called "windowless" flow counters to distinguish them from other types of counters. The absence of a window greatly increases the sensitivity of the instrument and makes it useful in counting alphas and weak betas such as those emitted by C^{14} and S^{35}. In the case of C^{14} the flow counter is more than twice as sensitive as a Geiger tube with the thinnest practical mica window. Figure 11.13 shows a windowless flow counter connected to a combination of an amplifier and scaler.

Additional information on proportional counters is given in Refs. 50 to 58.

GEIGER-MÜLLER COUNTERS

The forerunner of the Geiger-Müller tube was first used by Rutherford and Geiger in 1908 to count alpha particles. This first Geiger tube consisted of a brass tube with a wire along the axis of the cylinder. The cylinder was filled with a gas at reduced pressure and a potential slightly less than the sparking potential was applied between the wire and cylinder. In reporting their studies, these authors state that "...the small ionization produced by one alpha particle in passing along the gas could be magnified several thousand times. The sudden current through the gas due to the entrance of an alpha particle in the testing vessel was thus increased sufficiently to give an easily measurable movement of the needle of an ordinary electrometer."[2]

The G-M tube operates at a higher voltage than the proportional counter as indicated in Fig. 11.1. The voltage region between the "proportional" zone and the G-M zone, known as the "transition" region, is not generally used for instruments designed to measure radiation.

The G-M counter is similar to the proportional counter in many respects and a pulse is produced for each ionization event. As in the case of the proportional counter, the high voltage used with a Geiger tube results in a large multiplication of an original ion pair produced by the interaction of radiation and a gas atom or molecule. Unlike the proportional counter, the G-M counter produces a pulse the magnitude of which is the same for all the basic types of ionizing radiations.

Williams[119] in 1958 commented that, because of the low initial cost and other advantages, the G-M counter is at present the most widely used device for detection of radiation. The fact that the response of the G-M tube is nondiscriminating, being independent of the type and energy of the radiation, is often an advantage. In the majority of applications, where only one radioisotope is used, a discriminating device is not needed.

For the measurement of beta-particle radiation, the efficiency of counting is comparable with gas-flow proportional counters without the complications of the gas-flow apparatus. As compared to scintillation counters, the dimensions are smaller and the weight of lead shielding is less. Because of low gamma sensitivity the background counting rate is low.

For use in low-level detection the small size and low gamma sensitivity of G-M tubes are definite advantages. For use in portable dose-rate meters, the small size, ruggedness, and simplicity of G-M instruments are helpful. The advantages are increased with the use of halogen-quenched G-M tubes used in combination with transistor circuits.

Quenching G-M Tubes

One of the difficulties encountered in the operation of G-M tubes is the tendency of the tube to continue the discharge after the original avalanche has ceased. This results in low maximum counting rates and poor resolution of the counts. The suppression of the discharge is known as "quenching" and is necessary for satisfactory operation of the G-M tube. Quenching prevents the process whereby a positive ion becomes an excited atom at the cathode and de-excites by emitting a quantum of ultraviolet light which causes the further emission of electrons from other gas atoms. Quenching may be by a vacuum-tube circuit or an external resistance that lowers the voltage below the starting potential after each avalanche. This method of quenching requires a more complex circuit but has the advantage that the tube has a very long life. Quenching may also be accomplished by the use of organic gases in the tube. When organic "self-quenching" gases are used, decomposition products are deposited on the electrodes and limit the useful life of the tube. Essentially pure argon (98 per cent) is used as a non-self-quenching G-M tube gas.



Fig. 11.14—Portable G-M tube survey meter used to check for radioactivity.

It should be mentioned that even when a self-quenching G-M tube is used, an external quenching circuit is also sometimes employed. Such a circuit may serve as a preamplifier and also provide a low-impedance output, thus permitting the use of longer cables to the indicating or recording circuit.

The G-M tube may be made self-quenching by adding a small quantity of an organic gas such as methane or ethane to argon or isobutane to helium. The inert gas provides a low starting potential and a high specific ionization and the organic gas quenches the discharge. To explain how such organic gases quench the discharge, it is necessary to discuss the mechanism of the discharge.

As mentioned previously, the difference in mass between the electrons traveling toward the anode and positively charged gas ions traveling toward the cathode leads to more rapid collection of the electrons than the positive ions. Thus, a positive space charge accumulates tending to reduce the potential gradient and halt the discharge. When the positive ions produced by an avalanche reach the cathode, they are neutralized. The neutralized gas atoms have an excess of energy equal to the work function of the cathode surface minus the ionizing energy of the atom. This excess of energy may be released as an energetic photon with sufficient energy to eject an electron from the cathode. The net result is the release of excess electrons at the cathode which move rapidly to the anode and renew the discharge, creating a "spurious" pulse. In self-quenching G-M tubes, the polyatomic organic gas ionizes more readily than the noble gas (argon or helium). This results in a transfer of charges from noble-gas ions to organic gas molecules. Essentially all ions reaching the cathode will, therefore, be organic. The organic molecules can be decomposed, whereas the noble gases are stable. The energy exchange at the cathode is sufficient to decompose the organic-gas ions. This decomposition consumes energy that otherwise would release electrons and renew the discharge. However, this process gradually consumes the organic gas and results in the accumulation of decomposition products at the cathode. A high-quality self-quenching G-M tube may have a life of 10^8 to 10^9 pulses.

Another method of self-quenching uses a halogen, such as gaseous chlorine or bromine, in place of the organic gas. The diatomic halogen gas molecules dissociate at the cathode, thereby consuming energy and reducing the release of electrons. The halogen atoms recombine so as to renew the supply of diatomic halogen molecules. Such tubes have very long lives.

G-M Meters

One important application of the G-M tube is as a detection element in portable survey meters. Figure 11.14 shows a G-M tube portable survey meter being used to check the radioactivity of a potentially contaminated beaker.

In addition to checking for contamination, such portable survey meters are widely used in uranium prospecting. For both of these applications, the detachable G-M tube probe is extremely useful in exploring inside vessels, crevices, etc., into which a cutie-pie type meter could not be inserted. When low activities are to be detected, the G-M tube meter is much more sensitive than ion-chamber ratemeters because of the great internal amplification factor of the G-M tube. The instrument may also be used as a monitor to indicate radiation levels near areas of high radiation intensity.

The portable survey meter shown in Fig. 11.14 has three ranges: 0 to 0.2, 0 to 2, and 0 to 20 mr/hr. The threshold voltage for this G-M tube is 800 to 850 volts and the operating voltage is 900 volts. The G-M tube of this instrument is a glass tube of uniform thickness (no window), sufficiently thin to permit betas as well as gammas to enter but too thick for alphas. If the instrument is to be used to indicate gammas alone, the tube housing may be rotated 180° so as to shield out beta particles.

Earphones may be used with the portable survey meter. A change in the frequency of the Geiger "clicking" is usually a more sensitive indication of a change in radiation intensity than the meter needle, which usually is slightly "damped." In prospecting or in surveying for contaminated areas, earphones are often used. The probe is manipulated with one hand and the meter held in the other. When used as a portable monitor for gamma radiation, the earphones are not always used and the probe may be left in the holder if the meter is sufficiently sensitive to indicate the presence and level of gamma radiation.

The laboratory monitor uses a thin-window type of G-M tube and is designed primarily as a stationary meter rather than a portable meter so it can be powered by 60-cycle alternating current. This eliminates the cost and inconvenience of battery replacement.

One design of laboratory monitor has a thin mica window (2 to 4 mg/cm^2) at the end of the tube. This window is sufficiently thin to permit the use of this instrument for detection of low energy emitters such as C^{14} and S^{35}. The tube has a starting potential of 1100 to 1250 volts, a plateau of 300 volts, and a maximum plateau slope of 5 per cent per 100 volts. Although the G-M tube is self-quenching, the circuit contains an external quenching (multivibrator) circuit to improve performance. The instrument also contains a loudspeaker to produce an audible Geiger tick without the use of earphones. The instrument is useful in monitoring hands, clothing, bench tops, and miscellaneous equipment for contamination. It may also be used as a monitor to indicate the direction of remote radiation sources and to detect fluctuations in the "background" counting rate that might interfere with precise radioassays.

Geiger-tube Designs

The geometric form of the conventional G-M counter tube has remained substantially the same since its invention in 1908. The anode is generally a wire of 0.1 to 1 mm diameter, coaxially fitted in the hollow cathode. With the conventional types of gas it is necessary to keep the anode diameter small to achieve adequate quenching of the discharge. Increasing the anode diameter inevitably results in higher working voltage, greater slope of the plateau, and shorter plateau length.

Experiments by Hermsen et al.[59] have shown that the use of the halogen gas mixtures makes it possible to construct counters with thick anodes which have very good counting characteristics. This alteration has the advantage that the geometry of the design can be much better adapted to the pertinent application of the G-M tube. An additional advantage is that the dead time can be made smaller. Various experimental counter tubes were made with large anode diameter.

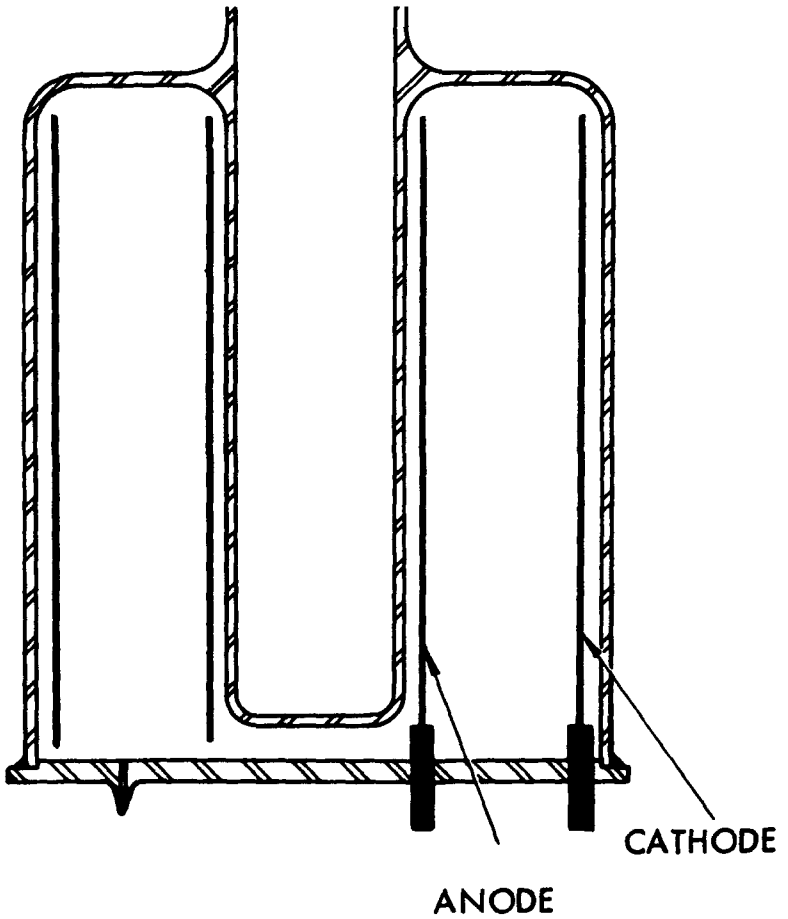


Fig. 11.15—Well type of 4π (G-M tube for gamma counting.)[118]

Increasing the anode diameter allows the possibility of making the anode hollow. The radioactive source may then be placed within the counter and by this procedure the solid angle increases considerably. The "well type" or "4π" gamma counter constructed according to this principle, as shown in Fig. 11.15, is used for counting the radioactivity of solutions. In the hollow anode a glass tube is inserted into which liquids can be poured.

The thin-wall, "pipe type," hollow-anode beta-particle counter (shown in Fig. 11.16) is suitable for measuring the activity of a flowing liquid, for example, with an ion-exchange column. In this design the solution flows through a thin glass tube that has a wall thickness of 30 mg/cm^2.

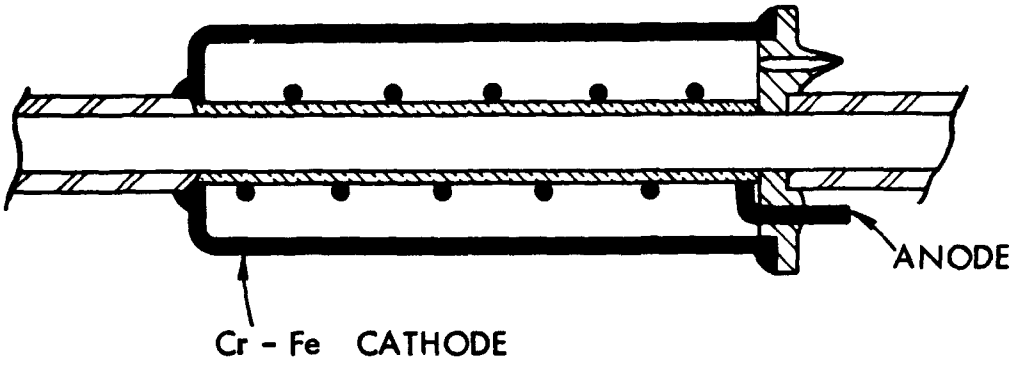


Fig. 11.16 — Pipe type hollow anode G-M tube for beta-particle counting.[118]

It is also possible to discard the cylindrical geometry of G-M tubes. Flat parallel plate type counters with a mica window have a small dead time. The window can be internally covered with a thin conducting layer or supported by a metal grid. These and other special designs of G-M counters have recently been reviewed and described by Van Duuren et al.[118]

The plateau and the dead time of irregular-shaped counters are strongly dependent on the value of the series resistance R. High values of R, 10 megohms for instance, give excellent plateaus. The dead time, however, is optimum with smaller resistance values. Therefore, the value of this resistance can be determined in accordance with the requirements of the experiment.

Additional information on G-M tubes and counters is given in Refs. 60 to 89 and 118.

NEUTRON COUNTERS

Neutron counters usually operate in the proportional region rather than the G-M region so that beta and gamma backgrounds may be screened out. Neutrons produce no direct ionization in gases because they have no charge and, therefore, no associated electric field. Ionization from secondary mechanisms must be employed to increase the ion pair yield in a neutron field if the neutron counter is to be sensitive. In the case of slow neutrons, the emission of alpha particles as a result of neutron capture is a typical mechanism. In the case of fast neutrons, the recoil energy produced by elastic collisions with light nuclei produce ionization. These processes will be described in greater detail.

Counter for Fast Neutrons

Fast neutrons are, in general, not as readily captured as slow neutrons. Therefore, transfer of kinetic energy by elastic collision is the most effective method of detecting neutrons with high energy. The average kinetic energy transferred in such interactions is given by Eq. 11.8.

$$E_{avg} = \bar{E}_n \frac{2M}{(M+1)^2} \tag{11.8}$$

where E_{avg} = average recoil kinetic energy of interacting nucleus
E_n = kinetic energy of interacting neutron
M = mass of interacting nucleus (per unit neutron mass)

The maximum kinetic energy that may be imparted to an interacting nucleus will be twice the average value. Thus, if a hydrogen atom is the interacting nucleus and the mass of hydrogen nucleus per unit neutron mass is taken as unity, the average recoil energy will be equal to $\frac{1}{2}E_n$. The maximum recoil energy will result from a head-on collision and will be equal to E_n.

A threshold exists for the recoil energy of nuclei of a gas below which a pulse will not be produced in a counting tube. This threshold B_0, is termed the bias energy. The lowest neutron energy, B, that can cause a pulse is equal to $2B_0(E_{avg}/E_n)$ or

$$B = 2B_0 \frac{E_{avg}}{E_n} = B_0 \frac{E_{max}}{E_n} \tag{11.9}$$

As E_{max}, the maximum recoil energy, is equal to E_n when hydrogen is used as the gas, $B = B_0$ for hydrogen. The sensitivity of the hydrogen-filled detection tube rises rapidly from zero for energies below B_0 and increases approximately with the inverse square root of the neutron energy for energies above 0.050 Mev. The sensitivity reaches a plateau between the limits of

$$\text{Plateau range} = 1.57B < E_n < 9.6B \tag{11.10}$$

In addition to measuring the neutron flux, the hydrogen-filled proportional counting tube may provide a rough method of measuring neutron velocities. This is accomplished by utilizing the existence of a detection threshold. As this threshold is dependent on both the neutron energy, E_n, and the bias energy, B_0, a change in the bias will change the lower energy limit for detectable neutrons.

Another type of proton-recoil detection tube uses films of a hydrocarbon or carbohydrate solid within the tube as the proton source. The interaction of neutrons with hydrogen atoms in such solids produces recoil protons which ionize the gas in the tube. As hydrogen is no longer needed as the proton source, heavy noble gases, such as argon, krypton, or xenon, which give better tube performance, are used for the fill gas.

Counters for Slow Neutrons

Slow neutrons are readily captured to produce n,α reactions. The resulting alpha particle may be used to ionize the fill gas and produce pulses. In such an instrument, nuclei with high cross-sections for the n,α reaction are required, such as B^{10}, which, by neutron capture, emits an alpha particle to produce Li^7. The boron may exist in the fill gas as boron trifluoride or as a solid metallic film on the inner surface of the cathode wall.[89]

The efficiency of boron trifluoride gas-filled counters can be increased by increasing the gas pressure and/or the B^{10} concentration over the 18 per cent present in natural boron. The increase in pressure is limited by the accompanying increase in operating voltage and also by the increase in size of the electron pulse. The dependence of counter efficiencies, defined as the probability of neutron capture in traveling through the counter an average distance X is given by Eq. 11.11.

$$\text{eff} = X \sigma pcL \tag{11.11}$$

where X = average length of neutron path in counter
σ = B^{10} cross section
p = pressure of BF_3 gas
c = B^{10} concentration
$= B^{10}/(B^{10} + B^{11})$
L = Loschmidt's number

Figure 11.17 shows a sectional drawing of a boron trifluoride neutron counter tube available at pressures up to 120 psi and containing 96 per cent B^{10} enriched BF_3 gas.

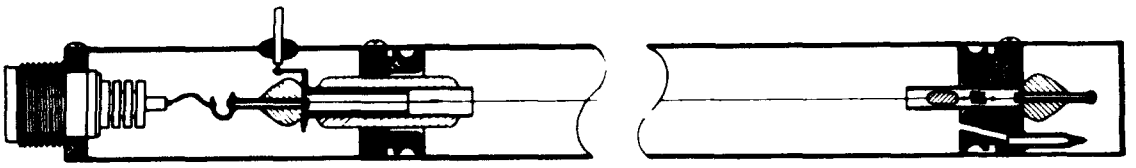


Fig. 11.17—Diagram of boron trifluoride neutron counter tube. (Courtesy of Radiation Counter Laboratories, Inc.)

The counter tube shown in Fig. 11.17 is of aluminum construction, except for the anode, Kovar seal, and connector. A ring guard is used to prevent spurious counts.

If the counter utilizes a film of boron on the cathode rather than boron trifluoride, BF_3 gas, a gas mixture of argon and alcohol may be used. For maximum efficiency of such counters the boron film thickness should be restricted to about 0.1 mm. This will permit the escape of an alpha particle ejected as a result of capture of a slow neutron by a B^{10} nucleus. A greater thickness results in the stopping of ejected alpha particles before they escape to the gas in the tube, thereby decreasing the efficiency. The relation between the maximum efficiency of such counters and other properties of the system is given by Eq. 11.12

$$\text{eff} = \frac{\sigma p R N}{A} \tag{11.12}$$

where σ = boron cross section for neutron capture
p = density of boron
R = range of alpha particle in boron
N = Avogadro's number
A = atomic weight of boron

Another type of neutron counter may be used in which the boron film is replaced with a film of uranium. In this case neutron capture in the film leads to fission of U^{235}. The fission fragments produce intense ionization in the gas. As in the case of boron counters, enrichment of the fertile material, in this case U^{235}, increases the efficiency of the counter.

Neutron Detectors for Various Flux Ranges

The BF_3 proportional counter is used to cover a neutron thermal flux range of from about 10^5 down to 1 neutron/cm^2-sec. Typical chambers are about 1 ft long, 1 in. in diameter and may be filled with 95 per cent B^{10} enriched BF_3 gas at about 2 atm of pressure. Such a chamber[119] is usually operated at about 2000 volts direct current, produces a gas amplification factor of about 500, and has a sensitivity of about 4 counts/neutron-cm^2-sec. If large amounts of gamma radiation are present, ion pairs will be produced by the Compton effect. However, the pulses from such reactions are less energetic than those produced by the alpha particles given off by the boron and can be biased out by suitable discriminator circuits in the linear amplifier. The design of a rugged BF_3 proportional counter was described recently by Swift and Bayard.[120]

The fission chamber is used for slightly higher ranges of neutron flux of about 100 to 10^6 neutrons/cm^2-sec. It is not as sensitive as the BF_3 proportional counter and gives only about 1 count/neutron-cm^2-sec. However, it has the advantage of being capable of operation in gamma fluxes as high as 10^{10} photons/cm^2-sec without loss in sensitivity. The typical fission chamber is about 1 ft long, 2 in. in diameter and contains an electrode coated with enriched U^{235} oxide. The fill gas may be a mixture of nitrogen and argon.

For higher ranges of neutron flux of about 10^4 to 10^{10} neutrons/cm^2-sec the compensated ionization chamber is useful. These chambers are larger, about 2 ft long and 3 in. in diameter and they have the advantage of compensating for gamma radiation. They consist essentially of two ionization chambers in combination. One chamber is coated with B^{10} enriched boron and is sensitive to both neutron and gamma radiation. The other chamber is uncoated and, therefore, sensitive to gamma radiation only. If the signals are combined, the current due to the neutron flux alone can be measured. These chambers have a sensitivity that will produce a current of about 2×10^{-14} amp/neutron-cm^2-sec.

For very high neutron fluxes of 10^5 to 2×10^{10} neutrons/cm^2-sec, uncompensated ionization chambers may be used. With such high power levels, compensation for gamma radiation is not considered essential because the relative contribution of the gamma radiation is less. The chambers are lined with B^{10} enriched boron and are similar to compensated chambers except that only one chamber is required.[119]

SCINTILLATION COUNTERS

A block diagram of a typical scintillation counter system is shown in Fig. 11.18. The scintillation counter consists basically of two parts, a phosphor and a photomultiplier circuit.

PHOSPHORS

The phosphor used in a particular application depends on the particle being detected and the information desired. The mechanism by which the energy-conversion process takes place

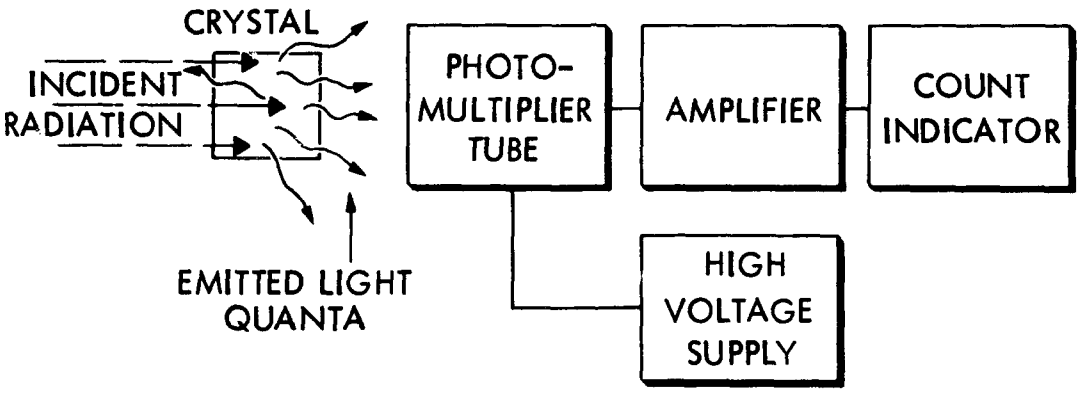


Fig. 11.18—Block diagram of scintillation counter system.

depends on the molecular structure of the phosphor and is somewhat different, depending on whether the phosphor is an organic or inorganic solid, a suitable material in an organic solution, or in some cases a gas. The process involves the transfer of energy from the ionizing radiation to the phosphor, usually as a dislocation of the normal arrangement of some of the electrons making up the material. This dislocation leaves the structure with an excess of energy. To reach equilibrium, the electrons undergo transitions to lower energy levels, giving up energy. Part of this energy, not more than 10 to 20 per cent, is emitted as visible radiation. The rest is usually expended as heat.

Scintillators are described as fast or slow on the basis of the decay time of the phosphorescence. A simplified model of the decay process may be described to illustrate. Since the scintillation is produced by secondaries trapped in the scintillator, one may say that for a total of n trapped electrons existing at any time, the number falling back to ground or normal states per second is proportional to n, and thus

$$dn = -\lambda n(dt) \qquad (11.13)$$

Hence, the number

$$n = n_0 E^{-\lambda t} \tag{11.14}$$

Here n_0 is the number present at zero time and $1/\lambda$ is a constant, characteristic of the material. It may vary for typical materials from about 10^{-9} sec to as much as 10^{-2} sec. Phosphors with small $1/\lambda$ are said to be fast and those with large $1/\lambda$, slow.

A number of phosphors have been used and some of the more common ones are listed in Table 11.2. The most satisfactory inorganic phosphors are zinc sulfide (silver-activated) and sodium iodide (thallium-activated). The silver-activated zinc sulfide is very efficient in the

Table 11.2—REPRESENTATIVE SCINTILLATORS[116]

Scintillator	Used to detect	Conversion efficiency	Decay time sec	Wavelength of scintillations at maximum response, A
ZnS(Ag)	Alpha	0.28	1×10^{-5}	4500
Anthracene	Beta	0.10	3×10^{-8}	4400
Naphthalene	Gamma	0.05	7×10^{-8}	3450
Stilbene	Gamma	0.06	5×10^{-9}	4080
NaI(Tl)	Gamma	0.20	2.5×10^{-7}	4050
Typical plastic phosphor	Gamma	0.024	1×10^{-8}	4550
Terphenyl (3 g/liter) in phenylcyclohexane	Gamma	0.05	8×10^{-9}	~4000

detection of alpha particles and the thallium-activated sodium iodide has been found efficient for the detection of both beta particles and gamma photons and shows a nearly linear relation between light output and radiation energy. The most satisfactory organic phosphors are anthracene, stilbene, and naphthalene. Anthracene is transparent to the luminescence produced, which permits collection of much of the light generated internally. It is an efficient detector for both beta particles and gamma radiation. Stilbene is useful if a very short resolving time is desired. Organic solutions such as anthracene or terphenyl in benzene or xylene fluoresce under gamma radiation or alpha or beta particles. Limited success has been obtained with solid noncrystalline phosphors such as terphenyl in polystyrene.

No high-efficiency phosphor has yet been found for the detection of slow neutrons; however, the organics and sodium iodide (thallium-activated) may both be used for this purpose. In the case of fast neutrons, the organics are very efficient because of the production of recoil protons. Scintillators for neutron detection may be prepared by incorporating in the scintillator some material which produces a detectable nuclear reaction in a neutron flux. A fast-neutron detector may be prepared by incorporating zinc sulphide powder in a plastic. The n,p reaction of the fast neutrons produces protons which are detected by the zinc sulphide. Thermal-neutron scintillators have been prepared by placing a layer of a compound containing U^{235} against a zinc sulphide phosphor. In this case the fast fission fragments due to thermal fission of U^{235} are detected by the zinc sulphide.[94,115]

Gas scintillators have been used for the detection of heavy charged particles in the presence of gamma radiation. The charged particles produce excitation of the gas and the resultant radiation is an indication of the particle-gas interaction. Since the gamma interaction probability is very low, such detectors have a very low background. Noble gases such as argon and xenon have been used successfully.

Plastic phosphors have certain desirable characteristics which have made them popular to some applications. They are usually more economical to fabricate than the large crystal phosphors and have practically no size and shape limitation. They may be prepared by dis-

solving highly purified scintillators in vacuum-distilled vinyl toluene monomer and then polymerizing[117] under controlled temperature conditions. They exhibit very little absorption of the scintillations. Since they are nonbreakable and noncorrosive, no special precautions in handling are necessary. The conversion efficiency may be low, but this is somewhat compensated for by the advantages. Liquid scintillators share most of the advantages of the plastic phosphor, but require special containers.

In selecting a phosphor the following characteristics are important: efficiency (light output per unit of energy); emission spectrum; decay constant; and problems of fabrication. In general, a thin screen of silver-activated zinc sulfide is adequate for the detection of alpha particles from natural radioisotopes. However, gamma photons, X radiation, beta particles, mesons, and high-energy particles are not as efficiently detected by zinc sulfide as by the other phosphors mentioned. For such radiations the luminescent organics are preferred for high-speed counting. Thallium-activated sodium iodide is preferred for greater sensitivity because it has greater stopping power and produces more light than the organics.

PHOTOMULTIPLIER TUBES

Two common types of photomultiplier tubes are the electrostatically focused and the unfocused multipliers shown schematically in Fig. 11.19.

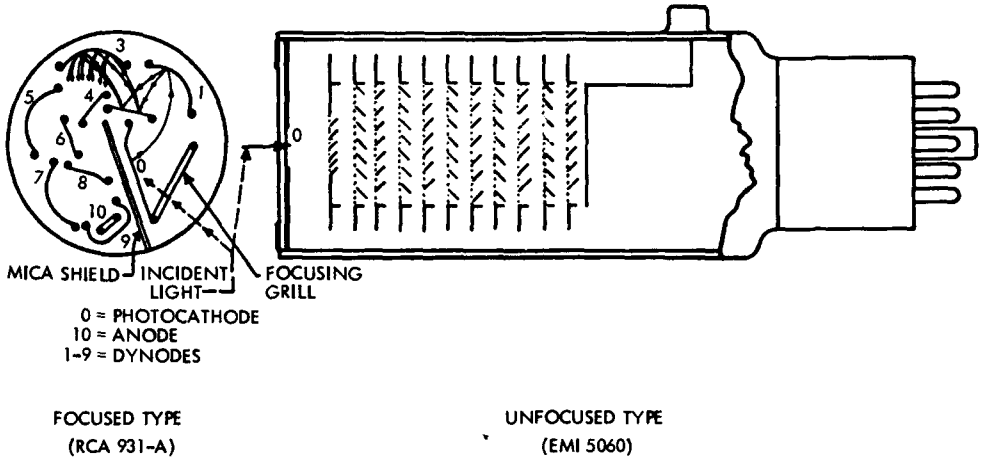


Fig. 11.19—Schematic diagrams of two types of photomultiplier tubes. (Courtesy of Tracerlab, Inc.)

The photomultiplier tubes shown schematically in Fig. 11.19 are basically vacuum phototubes with internal current amplifiers. When a light photon strikes the photocathode surface a photoelectron is ejected and accelerated by the applied field to the first dynode. The photoelectron is collected at the first dynode, causing the emission of three to five additional electrons which are accelerated to the second dynode followed by the emission of additional electrons and so on to produce an eventual total electron multiplication of 10^6 to 10^7. The multiplication depends on the number of stages, n, the secondary emission ratio, θ, (ratio of electrons emitted per incident electron), and a constant, K (usually ≈ 1), as shown by Eq. 11.15.

$$\text{Gain} = K\theta^n \tag{11.15}$$

For example, if K is taken as unity, θ as 4, and 10 stages are used, the gain is equal to 4^{10}, which is about 10^6.

Figure 11.20 shows a 14-stage head-on multiplier phototube that can deliver very high output pulse currents.

It may be used to multiply weak photoelectric currents by 10^7 to 10^8 when operated at 2300 volts and has short resolution time of the order of 1 to 2 millimicroseconds.

APPLICATIONS OF SCINTILLATION COUNTERS

Another important use of the scintillation counter is the field of gamma-ray spectroscopy for making accurate determinations of the energy of gamma photons. In such studies, and for research experiments requiring greater sensitivity than possible with ordinary G-M counters, the well type scintillation counter is used. A sectional diagram of such a counter was shown in Fig. 6.4 (see Chap. 6).

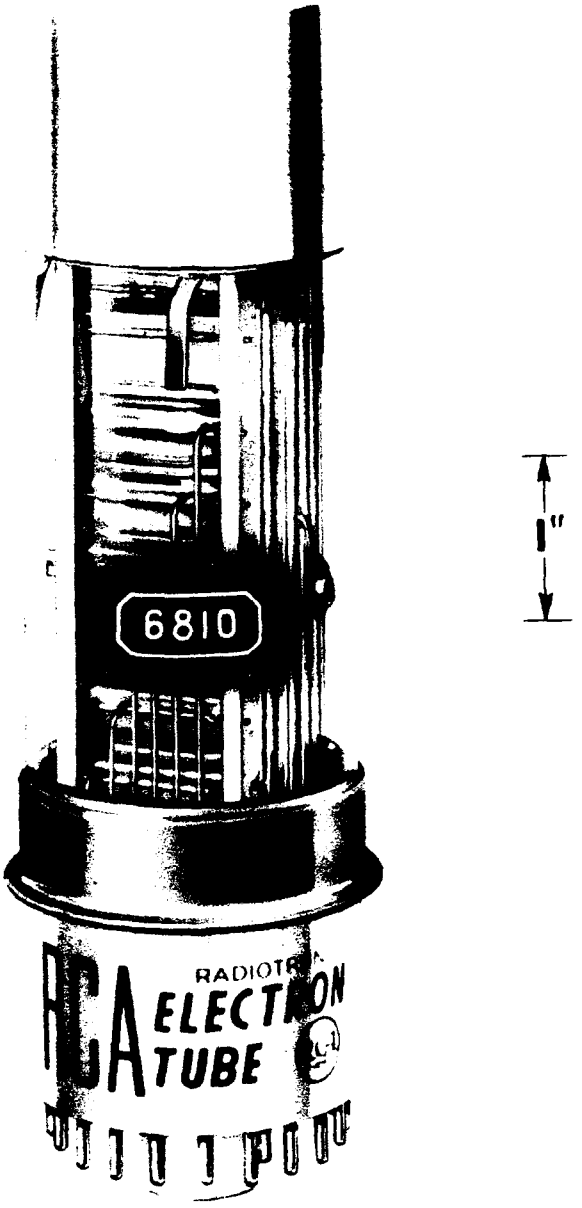


Fig. 11.20—Fourteen-stage multiplier phototube. (Courtesy of RCA.)

The scintillation counter is still undergoing rapid development and improvement.[91,92] One development is gaseous scintillation counting, which permits higher pulse rates and is almost completely insensitive to gamma radiation. The first studies with noble gas scintillation counters were made in 1953 by Muehlhause[93] who observed a barely detectable light output

from the alpha-particle excitement of argon or helium. Improvements since then have made gaseous scintillation counting an accepted procedure. Krypton, xenon, and argon-nitrogen mixture give improved performance.

Additional information on scintillation counters is provided in Refs. 94 to 116.

USE OF TRANSISTORS IN RADIATION DETECTORS

Goldsworthy[121] describes the design and use of a transistorized portable counting-rate meter. By means of interchangeable detector probes the instrument can be used for detecting alpha particles, beta particles, gamma radiation, or neutrons. An air-proportional probe, a scintillation probe and a G-M probe are used. Exclusive of the probes the instrument weighs less than 4 lb and measures about 4 by 4 by 9 in. Goldsworthy suggests that because of compactness, versatility, low battery drain, and ease of servicing, these instruments should make very useful multipurpose radiation detectors. Additional information on use of transistors in radiation detectors is given in Refs. 121 to 128.

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