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Radioactivities Produced in Foods by High-Energy Electrons

Period: 15 January 1958 - 14 January 1962

QUARTERMASTER FOOD AND CONTAINER INSTITUTE FOR THE ARMED FORCES
Research and Engineering Command
Quartermaster Corps, U.S. Army
Chicago, Illinois

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March 15, 1962

Final Report

Report No. 17

RADIOACTIVITIES PRODUCED IN FOODS BY HIGH-ENERGY ELECTRONS

Prepared for:

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Contract No. DA 19-129-QM-1100

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SRI Project No. PHU-2424

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I SUMMARY

An extensive current research effort is devoted to developing radiation processing of foods. With this type of food processing it might be possible to induce radioactivities in the foods which would impair their general wholesomeness. To determine whether or not such processing represents a potential health hazard, SRI has pursued a program for the past four years to establish the levels of induced radioactivities in foods. One part was concerned specifically with radioactivities induced by the (γ,γ') reaction in foods irradiated with gamma rays and x-rays of several energies. The other part of the program considered radioactivities induced by a variety of reactions during irradiation by high energy electrons. Phase I of this program measured the activity induced by 24-mev electrons. Phase II, reported here, extended the program to cover several foods and a wider range of energies (6 to 25 mev).

A large, uniform sample size (No. 10 can) was used throughout Phase II of the program. Because of the wide energy range considered, no attempt was made to secure uniform dose distributions in the samples, but the average sample dose was about 5 megarads in each case.

To allow efficient detection of the gamma activities in the samples, a special large-sample gamma counter was developed. The approximate lower limit of detectability of gamma activities for this detector is 0.001 micro-microcuries per gram food. Portions of the sample ash or chemical fractions were counted in existing small counters for any induced beta activity. The only induced radio isotopes observed were Na^{22}, Na^{24}, and, in one special sample, Rb^{84}. The induced sodium activity levels were observed to decrease sharply with decreasing electron energy.

A unique design for a large-sample beta counter was conceived and some effort was devoted to preliminary design considerations. Appropriate components were not available during the investigation to allow experimental verification of the preliminary considerations so

final design and construction of the counter was not attempted.

Some theoretical calculations of the induced levels of several isotopes at several electron energies were carried out. A comparison of the calculated and the experimentally measured Na^{22} values showed reasonable but not exact agreement. An attempt to calculate the radioactivities induced by x-rays of several energies was less successful.

As previously observed at 24 mev, Na^{22} continues to be the most important long-lived induced radioactivity at lower electron energies. The observed decrease in the measured levels with decreasing electron energy is consistent with the reported (γ,n) threshold of 12.1 mev. The variation in the measured Na^{22} levels under presumably similar conditions indicates a strong sensitivity of induced radioactivities on trace element variations. The occurrence of induced Rb^{84} in only one special beef sample further indicates a wide variation in trace elements which must be expected between samples of different foods and even between different samples of the same food.

The short-lived induced radioactivity of Na^{24} was observed generally in the foods irradiated at the higher electron energies and in one sample irradiated at as low an energy as 10 mev. This isotope was probably produced by both the (γ,p) and the (n,γ) reactions but the relative importance of the two reactions was not measured.

No beta-emitting radioactivities were observed during the investigation. However, an expression was developed to predict the levels of other activities produced by the (γ,p) or (n,γ) reactions relative to that measured for Na^{24}.

The only isotopes (besides Na^{24}) which have predicted activity levels sufficiently high to be measurable are P^{32}, P^{33}, K^{42}, and K^{43}. The last two (gamma-emitting) isotopes were not observed in the present investigation indicating that the predicted levels may be better interpreted as approximate upper limits for the expected activities.

Sufficient data were developed to allow a reasonably accurate evaluation of the potential health hazard presented by the induced radioactivities in radiation sterilized foods. The measured levels of induced radioactivities are in every case significantly less than the maximum permissible concentrations (MPC, in water from NBS Handbook 69. In foods irradiated with 14-mev electrons, the Na^{24} and Na^{22} levels are factors of 1,000 and 100,000, respectively, below the indicated MPC values.

Several areas are suggested for further investigation.

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II INTRODUCTION

The U. S. Army Quartermaster Corps is currently responsible for an extensive research effort devoted to obtaining basic information on radiation processing of foods. Many scientific and economic disciplines are necessarily being combined to help solve the complex problems associated with this new type of food processing. Since the ultimate aim of this research is to develop a product for human consumption, the general wholesomeness of the foods is of major importance and it is necessary to assure that the foods will not contain greater than permissible concentrations of radioactivity.

Radioactivity is a naturally occurring phenomenon in our environment and only recently has man altered the kinds and levels of radioactive isotopes. All foods, regardless of type or location of growth, contain measurable amounts of these natural radioactivities and presently many foods also contain any of several radioisotopes which are a result of atomic fallout. Still more radioactivity may be added with the preservatives and seasonings used in conventional food processing (potassium-40 is the principal contributor). In the sterilization of foods with ionizing radiations, an additional and unique potential source of radioactivity must be considered. It is possible that the radiations may interact with the stable nuclei of the food elements, causing them to become unstable or radioactive. It is important, therefore, as one of the initial steps in designing a radiation sterilization program, to establish the levels of any induced radioactivities, so that the potential health hazard can be evaluated.

Several investigations have been made to determine the level of induced radioactivities under a variety of irradiation conditions. Meinke[1]* reported on activation by cobalt-60 gamma rays and Kruger[2]

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* Superscript numerals refer to references collected at the end of this report.

measured neutron fluxes and induced radioactivities at several irradiation facilities. Five investigations have been concerned specifically with activation by high energy electrons. Two of these were primarily theoretical studies and no definite results were reported.[3,4] The third study made using 16-mev electrons found no measurable induced activities[5]. In a fourth study aqueous solutions of food elements, bacon, and cheese were irradiated with 25-mev electrons[6], and the levels of several induced radioactivities observed in this investigation were measured. In addition to these, an investigation currently being carried out by Vanderbilt University and Tracerlab, Inc. is designed to measure the radioactivities induced in foods by electrons of a variety of energies[7].

An extensive experimental program to determine the abundance of radionuclei in food has been conducted at Stanford Research Institute (SRI) over the past four years. This program consisted of two separate investigations, each sponsored by the Quartermaster Corps (QMC). One of these, Contract DA 19-129-QM-1511, measured the radioactivities induced in elements and foods by gamma rays and x-rays of several energies up to 24 mev[8]. Results of this project showed that the process of isomer activation could lead to induced radioactivity in selected food elements when they were exposed to radiations as low in energy as 1 mev. However, no radioactivities attributable to this process were observed in actual foods.

The other investigation, Contract DA 19-129-QM-1100, is the subject of the present report. This project was carried out in two distinct phases. The first phase was the measurement of radioactivities induced in foods by 24-mev electrons. To enhance the production and detectibility of some normally undetectable induced activities, small samples of aqueous solutions of food elements, or of foods which were artificially enriched with certain food elements, were irradiated, generally to doses in excess of 5 megarads. The results of these irradiations were then corrected to the elemental abundances and doses normally anticipated in food processing and were used to predict the

levels of other activities which might occur in the foods. Additional irradiations of untreated food were carried out to verify the predicted levels for observable radioactivities. The results of this phase of the investigation and predictions of the radioactivity levels which might be expected in foods processed with 24-mev electrons were given in Summary Technical Report (Report No. 10)[9] issued at the end of the first two years of work. Also included in this report was an extensive discussion of activation processes, a description of experimental procedures and apparatus, the results of a literature survey of elemental abundances in food, and a calculation of the radiation levels which might be realized in the vicinity of food samples irradiated with 24-mev electrons.

An extensive theoretical consideration of the processes leading to induced radioactivities and a computer program developed to calculate radioactivities produced in foods during sterilization with high energy electrons are given in Technical Report No. 11[10]. To demonstrate the validity of this computer program, the levels of induced activities following irradiation by 24-mev electrons were calculated and comparison was made with the experimental results from the preceding phase of the investigation. The agreement was very good, generally within experimental error.

In the second and present phase of the investigation, the major emphasis was placed on the experimental measurement of radioactivities induced in actual foods. The range of energies considered extended down to 6 mev; however, the energy range between 10 and 16 mev was emphasized. The number of foods considered was also greater than in Phase I. Using the program and methods developed in Report No. 11, additional computer calculations were carried out to predict the radioactivity levels which might be expected at these lower energies.

The present report is specifically concerned with the differences of approach and method pertinent to the present phase of the investigation and is intended to be supplemental to Reports No. 10 and No. 11. The general discussion of the processes leading to induced

activation, beam monitoring equipment, food element variations, etc. are therefore not repeated, but occasional specific references are made to data or discussions in Reports No. 10 and No. 11 which are of particular interest to the present work.

III PHASE II EXPERIMENTAL PROGRAM

A. Phase II Approach to the Problem

The approach followed in the second phase of the SRI investigation of induced radioactivities differed in three ways from the approach previously followed: (1) large samples of unaltered foods were irradiated and large counters were used for measuring radioactivity, (2) several kinds of foods were irradiated and (3) the range of electron energies was extended (6 to 25 mev).

At the request of the QMC, all foods irradiated in the present phase of the investigation were packaged in conventional No. 10 cans. This choice was appropriate because previous irradiation experience indicated that the No. 10 size sample was particularly convenient for use with accelerators having high current, narrow beam characteristics; and, at the higher electron energies, samples of this size were required to assure absorption of the electron beam in the sample. Further, the use of a uniform sample size permitted more efficient and economical irradiation and counting of successive samples and simplified data reduction.

To gain a greater net detection efficiency (and hence lower limits of detectability) of gamma radioactivities in these large samples, a large gamma scintillation detector was used. In general, the geometrical contribution to the radiation detection efficiency of the crystal was comparable to that achieved previously with small samples and small crystals. However, the increased probability of detecting a given gamma ray in the full energy photopeak and the relatively small increase in background in such large crystals greatly increased the net sensitivity for detecting and identifying radioactivities induced in the foods.

As indicated in Report No. 10, the elemental abundance may vary between different samples of the same food. Even greater variations may be expected between samples of different foods. It therefore seemed advisable to include in Phase II a greater variety of foods than previously used. Those chosen included beef, pork, ham, chicken,

shrimp, and green beans. Peaches were originally included and some measurements were made on peach samples; however, they were excluded from further irradiations after container deterioration became evident in some of the control samples. Samples of the QMC composite diet were also included. Prepared samples of the selected foods were supplied by the QMC. In the preparation process, large batches of each food were first homogenized; samples from the batch were then sealed in No. 10 cans and heat-sterilized prior to irradiation.

Most of the induced radioactivities previously observed in foods irradiated by high-energy electrons resulted from reactions leading to the emission of neutrons or protons from the nucleus. It was expected that the efficiency for promoting such reactions should decrease with decreasing electron beam energies. To demonstrate and evaluate the decreasing production of radioactivities, irradiations were carried out at energies down to 6 mev. The region between 10 and 16 mev, known to be the general threshold region for these reactions, was emphasized.

B. Irradiation of Samples

1. Irradiation Procedures. Since the object of the present investigation was not to optimize sterilization of the foods, no special care was taken to assure uniform dose distribution within the samples. Indeed, over the range of electron energies considered, it would have been impossible to achieve uniform dose distributions in a single sample size. This impossibility results from the characteristic manner in which electron beams deposit energy in matter: the relative dose deposited by the beam rises from some value at the surface of the material toward a peak value as the flux of dose-depositing, low-energy secondary electrons builds up; then as the primary electrons are absorbed, the dose falls off steeply (almost exponentially) at greater depths. The depth of the peak value and the extrapolated range of the distribution depend upon the energy of the electron beam with these quantities being greater for higher energy.

This non-uniformity of dose distribution leads to the concept of an "ideal" package depth corresponding to every electron energy--that package depth which, when cross-fired, gives the most uniform distribution. The ideal package depth varies from about 8 inches for 25-mev electrons down to about 2 inches for the 6-mev electrons used in the present work. The No. 10 can (7 inches thick) is an ideal package for electrons between 22 and 24 mev.

Since uniform dose distributions could not be achieved in No. 10 cans at all energies of interest in Phase II, a black-box method of irradiation was used. In this, a total amount of energy which was equivalent to that which would be required for a uniform irradiation to the chosen dose was delivered to each can by cross-firing. This method simplified the irradiation procedures; however, it might be suspected that the method could lead to an artificially high level of any induced radioactivities. The reason for such enhancement lies in the deviation from "ideal" sample size. In actual food sterilization the sample depth will be dictated by the electron energy to be used and the number of electrons of the given energy required to give the desired dose will be delivered to the sample. In general, some fraction of the total electron energy will be given up in bremsstrahlung (x-ray) production. It is this bremsstrahlung which reacts with the atomic nuclei to produce essentially all of the induced radioactivities. Bremsstrahlung of higher energy (that most capable of producing activities) will continue on through the package. Some will be scattered or absorbed by nuclei, thus inducing radioactivity; but much will pass out the back of the package. As indicated, there will be a definite relation, depending on experimental conditions, between the number of primary electrons and the deposited dose. There will also be a definite relation between the number of electrons (hence the number of x-rays produced) and the induced activities.

In the black-box method of irradiation, some number N of these ideal packages are, in effect, stacked to a thickness equal to that of a No. 10 can. If N times the number of electrons needed to give the

proper dose to the ideal package are delivered to the stack, a dose of N times the proper dose will be delivered to the first package depth while little or no dose is delivered at greater depths, but the average dose to the stack will be the proper dose. However, the activity-inducing x-rays formed in the first package depth will also increase by N times and this increased flux will continue through the other packages in the stack. The increase of x-rays at all depths will lead to enhancement of any activities produced by an amount approximately equal to the same factor N. The correction needed to account for this enhancement is small at higher energies but approaches a factor of 5 at energies near 6 mev. In Phase II this correction factor was in general smaller at each energy than the uncertainties resulting from other experimental parameters. However, since the failure to include it would result in the propagation of a systematic error, the correction factor was applied to the present results.

No experimental dosimetry was carried out during Phase II. Instead the integrated primary electron flux (microamp-min) required to give the desired average dose was determined using a simple calculation. The average dose rate resulting in a sample of mass M from an electron beam of energy E is given by

$$\text{Dose Rate} = \frac{E\left(\frac{\text{mev}}{\text{electron}}\right) \times 1 \left(\frac{\text{coulomb/sec}}{\text{amp}}\right) \times 10^{-6} \left(\frac{\text{amp}}{\mu\text{amp}}\right) \times 1.6 \times 10^{-6} \left(\frac{\text{ergs}}{\text{mev}}\right) \times 60 \left(\frac{\text{sec}}{\text{min}}\right)}{1.60 \times 10^{-19} \left(\frac{\text{coulomb}}{\text{electron}}\right) \times 10^{2} \left(\frac{\text{ergs/gram}}{\text{rad}}\right) \times M \text{ (gram)}}$$

$$= 6.0 \times 10^{6} \frac{E}{M} \quad \frac{\text{rads}}{\mu\text{amp.min}}$$

$$= 6.0 \frac{E}{M} \quad \frac{\text{Mrad}}{\mu\text{amp-min}}$$

Such dose calculations have been found to give results in good agreement with experimentally determined values, even though some of the

input energy is scattered out of the sides of the sample or lost as bremsstrahlung.

2. Irradiation Apparatus. Samples for Phase II were irradiated at two electron accelerator facilities: The General Atomic Electron Linear Accelerator was used for energies from 8 to 25 mev and the Midwest Service Irradiation Center accelerator at Rockford, Illinois, was used for energies from 6 to 10 mev. The experimental setup at General Atomic was the same as that previously described (Report No. 10). The cans were rotated at 96 rpm on a jarmill which was arranged so that the center of the beam struck one face of the can approximately one-half way between the center of the face and the upper rim. The rotation of the can then allowed electrons to enter all portions of the face. At the Rockford accelerator, which is a vertical machine, the cans were placed on a rotating horizontal turntable and were again oriented so that the axis of rotation (center of the can) was parallel to, but slightly displaced from, the beam centerline. At each installation polyvinylchloride films were fastened to the can faces during the irradiations to determine the beam position and to assure that the beam always entered the can as desired.

During most of the irradiations at both facilities, the average beam current was measured using a secondary emission monitor. Recorded curves of average beam current vs. time were integrated to give the integrated beam current for use in computing the dose. When such monitors were unavailable, pre- and post-irradiation measurements of the instantaneous beam pulse height were used to determine the average current per pulse during the irradiation. This average current, the accelerator duty cycle, and the total irradiation time were then used to estimate the integrated current for the runs. The beam energy spectrum was measured at General Atomic using magnetic beam analysis methods. At Rockford such analysis was unavailable and the maximum beam energy was estimated by the depth of darkening in stacks of microscope slides.

C. Counting Apparatus

1. Large Sample Gamma Counter. Following irradiation, the samples were generally returned immediately to SRI and transferred to clean unirradiated cans for gamma counting. However, the samples irradiated at Rockford were first counted using the Low-level Food Radioactivity Detector at the QMC F+CI in Chicago.[11]

For the routine counting of the food samples at SRI, a low background--but not an ultra-low background--gamma counter was assembled. While it was desirable to reduce to a minimum the cosmic and terrestrial radiation and that resulting from radioactivities inherent in the shielding material, the techniques now used to reduce the potassium-40 (K^{40}) background in scintillator assemblies (light piping, quartz-faced multiplier phototubes, etc.) were not required for the purposes of the present work. It was expected that the portion of the background which might have been reduced by these techniques was overshadowed by the gamma radiation resulting from the radioactivity normally occurring in food samples. Previous counting experiments with large NaI detectors (Report No. 10) had shown count rates of about 50 counts per minute for K^{40} contained in the foods. For comparison the K^{40} activity detected in large NaI crystals as a result of potassium occurring in the phenolic bases of the photomultiplier tubes in the glass of the tube envelopes, etc., might normally be of the order of 10 counts per min.

A Harshaw 8 inch by 4 inch NaI (Tl) crystal of their "Matched Window" design was selected as the nucleus of the gamma-detection system. Although this is a packaged detector, it more than adquately meets the experimental requirements for background and resolution. A detector of this design also can be easily modified as necessary for any future use in more refined or ultra-low background experiments. The large size of the crystal was chosen to take advantage of the greater net detection efficiency offered for counting large samples over that for detectors previously used at SRI.

The shielding for the crystal assembly (Fig. 1) was designed to satisfy present requirements and yet allow for later modifications which might be required for present or future investigations. Since the efficiency of detecting background radiation increases with the size of the detector, an attendant increase in the shielding is required to reduce the crystal background to a desired level. Previous experience had indicated that 3 to 4 inches of lead should reduce the background in an 8 inch x 4 inch crystal to a level sufficiently low for most applications. The indicated design was chosen to achieve an equivalent shielding. For the cylindrical portion of the shield a 3 inch thick lead annulus was cast between two concentric pieces of standard steel pipe, with the outer pipe 18 inches OD and the inner pipe 12 inches OD. Internal to this portion of the pipe is a 1 inch thick annular reservoir (formed with the 12 inch pipe and an additional 10 inch OD pipe) which was filled with mercury. The mercury served to increase the effective shield thickness for background radiation and also shielded against any radiation emanating from the lead. (Since several of the natural radioactivity decay chains pass through or terminate in lead isotopes, most commercially available lead has inherent radioactivity which sets a limit on its shielding capabilities. Mercury is, however, essentially free from such radioactive impurities.) In the mercury reservoir an additional annular region approximately 1 inch thick was left for possible inclusion of a "graded Z" shield to reduce the backscatter and luminous radiation or an anti-coincidence shield should the assembly later be modified for ultra-low background investigations.

The sample volume included within the shield is 8-1/4 inches in diameter and 8-1/4 inches in height (a No. 10 can is 6-3/16 inches in diameter and 7 inches in height). The sample volume is reached through a track-mounted, split-door in the top portion of the shield. (Details of this door are not indicated in the figure). A removable plug was included in the bottom portion of the shield to give access

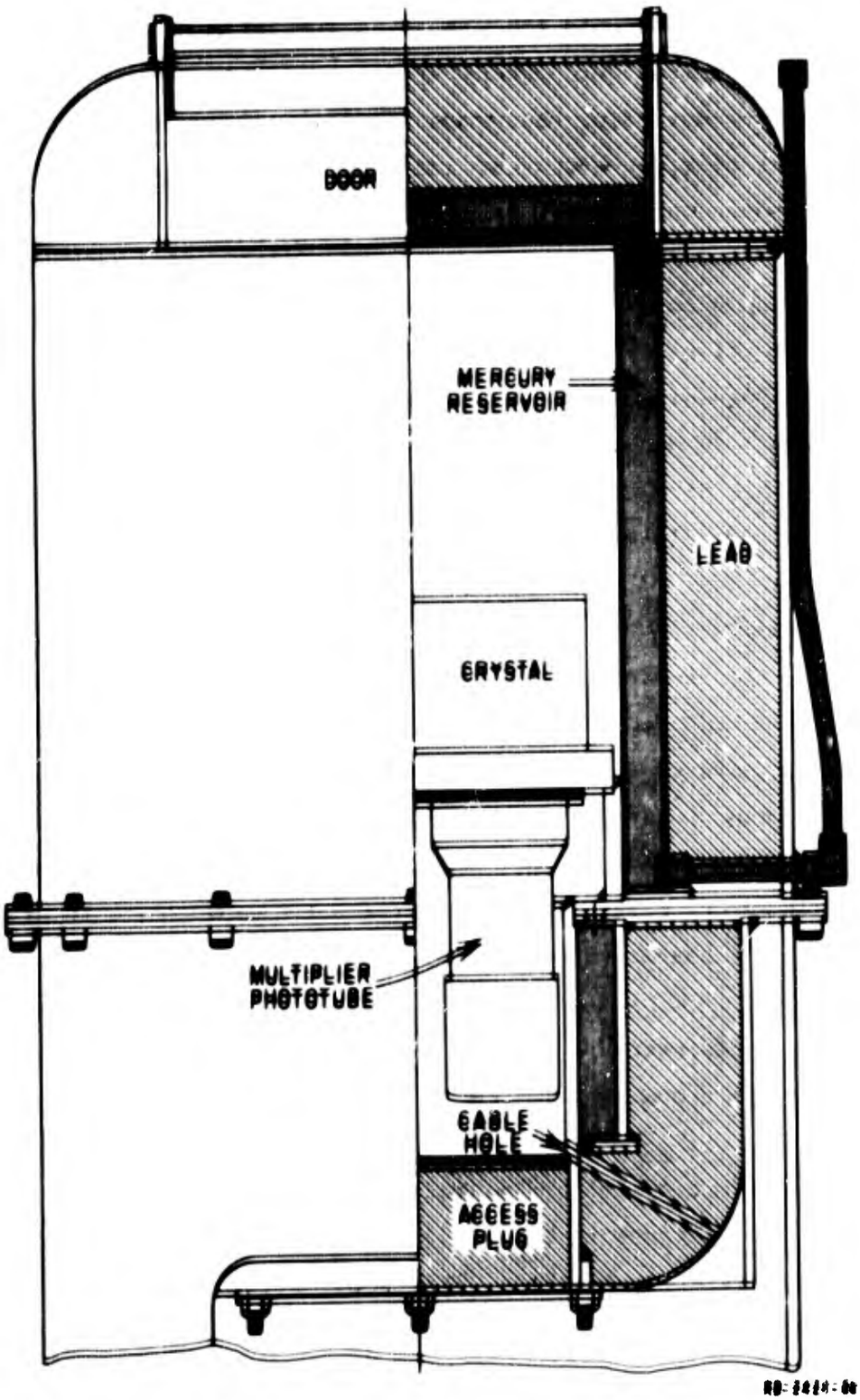

FIG. 1 GAMMA RAY DETECTOR ASSEMBLY SHOWN IN QUARTER SECTION

to the photomultiplier tubes for individual testing or adjustment. The top and bottom of the shield are similar to the main body except that the inner mercury shield is not included in the plug.

After the gamma detector was assembled it was necessary to determine the operational characteristics of the system, in particular the detection efficiencies and the resolution of the scintillator crystal. The efficiency of specific interest to the present investigation was that for detecting, in the full energy photopeak, the gamma rays emitted by the radioactivity uniformly distributed in a unit density sample packaged in a No. 10 can. The measured resolutions and efficiencies are given in Table I.

Table I.

Large Sample Gamma Detector Efficiency and Resolution

Source	Gamma Energy (mev)	Resolution	Efficiency
Positron annihilation (Na^{22})	0.511	0.105	--
Cs^{137}	0.662	0.095	0.047
Zn^{65}	1.114	0.082	0.042
Na^{22}	1.276	0.082	0.038
K^{40} (in KCl)	1.46	0.076	0.035

The resolution as given here is the ratio of the full width of the photopeak at half maximum height to the mean energy of the photopeak. The efficiency is the ratio of the gamma rays detected (under the conditions and restrictions described above) to the number of gamma rays emitted by the sample.

2. Beta Counters. It was originally anticipated that the counting of the samples for beta activities would similarly be done using a large-sample detector. However, the problems involved in the design of such a counter were not adequately solved during Phase II to justify construction. A discussion of the design considerations is given in Section IV. Lacking such a large-sample detector, the beta

counting was done using the 2π flow geiger counter and the low background geiger counter previously described (Report No. 10). Samples of the whole ash or chemically separated portions were counted.

D. EXPERIMENTAL DATA

1. Data Analysis. The experimental determination of gamma radioactivity in the foods proceeded in the following manner: unirradiated control samples of each of the foods were counted in the large-sample gamma counter and the gamma spectra were examined for any inherent radioactivities resulting from naturally occurring radioisotopes or fallout products. The spectra of each of the irradiated samples were then examined and compared with appropriate control spectra to determine the nature and extent of any additional radiation-induced radioactivities.

Standard methods were used to extract gamma radioactivity data from the spectra. To obtain a relative measure of any radioactivities in a given sample, a spectrum was integrated (from about 0.10 to 1.50 mev) and compared with a similar integration of the background. Integrations from irradiated samples were further compared with integrations from appropriate controls to obtain an indication of the extent of any radiation-induced activities. These spectra were then scanned for any obvious photopeaks, which, when present were analyzed to identify the radioisotope or radioisotopes present in the samples. Each photopeak was then separately integrated, background was subtracted (including that resulting from other radioactivities in the sample), and, using the previously measured detection efficiencies for the energy region of interest, the absolute radioactivities for the indicated isotopes were calculated. Spectra from some irradiated samples were also integrated step-wise in approximately 100-kev energy groups and compared group by group with the controls to determine the presence of small, non-obvious amounts of any induced activities. Since the induced isotopes, Na^{22}, was apparent in the spectra of all foods irradiated at the higher electron energies, spectra of every irradiated sample were analyzed for this isotope, even when no

photopeaks were obvious. The results of these more specific analyses of the spectra were then compared with the total spectra integration results to verify that the sets of results were consistent and that no significant radioactivities had been overlooked.

To express any induced radioactivities in units which are of interest to the present investigation (micro-microcuries per gram food per 5 megarads) a number of steps were required. As previously indicated a sample activity A_s, a background activity A_b, and a control activity A_c were determined for an energy interval of interest. Then, for a sample of mass M_s, which had been irradiated to a dose of D megarads, the desired radioactivity level is given by

$$\frac{\mu\mu\text{curies}}{\text{gram food-5Mrads}} = \frac{\left[\frac{A_s - A_b}{M_s} - \frac{A_c - A_b}{M_c}\right] \frac{\text{cpm}}{\text{gram food}} \times 5 \frac{\text{Mrads}}{\text{5Mrads}}}{\epsilon \frac{\text{cpm}}{\text{dpm}} \times D \text{ Mrads} \times 2.22 \frac{\text{dpm}}{\mu\mu\text{curie}}}$$

where ϵ is the detection efficiency for the gamma rays in the energy region of interest and M_c is the mass of the control.

It is estimated for the conditions of Phase II that the lower limit of detectability for any induced radioactivity was about 0.001 micro-microcurie (0.002 dpm) per gram food per 5 megarads. This value corresponds to about 5 net counts per minute per can in the gamma photopeak over most energy regions of interest. This value is within the normal fluctuations observed in successive counts of given samples or background and is certainly within the differences observed between counts of different control samples of any foods. At this net count rate, the standard deviation of the difference of two routine one hour counts became several times larger than the difference itself so that the statistical significance of the data was small. While this limit might be lowered by making longer counts, irradiating to higher doses, or reducing the counting background, the limit seems to be consistent with limitations imposed by other experimental variables (sample inhomogeneity, etc.) and is therefore adequate for the present investigation.

The limits indicated for values are those appropriate to a 90% degree of confidence. They were determined from the statistical standard deviation which would be expected for single measurements of the observed quantities. Repeated measurements of radioactivity levels measured in presumably similar samples are consistent with this expected deviation; however, occasional inconsistent variations were observed between similar samples (probably as a result of sample or electron beam inhomogeneity). Since the intent of Phase II was to measure radioactivity induced in homogeneous samples irradiated under reproducible beam conditions, such variations were reported in the experimental data but were excluded in the error analysis. A summary of the error considerations used in the present report is given in Appendix A.

2. Natural Radioactivities. The most apparent gamma activity in unirradiated foods results, as would be expected, from the naturally occurring radioisotope K^{40}. The spectra of each food showed a prominent photopeak at 1.46 mev, which is characteristic of this isotope. The extent of this radioactivity as determined for each of the eight foods is given in Table II. Using these measured radioactivities and the specific activity of K^{40} (micro-microcuries of K^{40} per gram K) the percent abundance of potassium was calculated for each food. These values are compared, where possible, with the average value and with the range of values for potassium abundances given in Report No. 10. While the present values are generally within or close to the previously reported range of values, the several exceptions (notably shrimp) indicate that wider variations of trace element abundances in foods may be encountered than previously suspected.

Table II

NATURAL POTASSIUM LEVELS IN FOODS

Food	K^{40} μμcuries gram food	Potassium Abundance in Food (%)		
		Present Data	Report No. 10	
			Average	Range
Beef	2.52 ± 0.29	0.296 ± 0.034	0.336	0.290-0.382
Pork	2.09 ± 0.24	0.246 ± 0.028	0.326	0.260-0.415
Ham	1.63 ± 0.24	0.192 ± 0.028	0.210	0.110-0.281
Chicken	1.56 ± 0.23	0.182 ± 0.027	0.367	0.250-0.402
Shrimp	0.81 ± 0.13	0.095 ± 0.015	0.447	0.220-0.760
Composite Diet	1.60 ± 0.24	0.188 ± 0.028		
Beans	1.36 ± 0.21	0.160 ± 0.025	0.263	0.186-0.300
Peaches	0.93 ± 0.15	0.109 ± 0.018		

There is some indication that the fallout product cesium-137 (Cs^{137}) is present in the food spectra; however, the levels appear to be very low. In every food the activity of this radioisotope is well below the value of 0.36 micro-microcurie per gram food previously measured in large crystal counting of other beef samples (Report No. 10). The highest level observed in the present foods is that of green beans which is approximately 0.08 micro-microcurie per gram beans. The reduced Cs^{137} levels result from the greater time that elapsed between atmosphere nuclear tests and the preparation of the foods.

3. Induced Radioactivities. Following irradiation, each sample can was opened, thoroughly mixed to assure uniform distribution of any induced radioactivities, and replaced in clean, unirradiated cans for gamma counting. Removal to fresh cans was necessary to prevent any radioactivities induced in the container materials from interfering with the determination of those induced in the foods. Such container radioactivities, in particular manganese-54 and tin-123, have been observed in the cans of samples irradiated at energies at least as low

as 16 mev. Since these radioactivities would not in general be transferred into the foods or could in any case be reduced or completely eliminated by container modification, they were not of interest to and were purposely excluded from Phase II considerations.

Gamma spectra typical of irradiated ham samples are shown in Figure 2. As indicated, the superimposed spectra are those of samples irradiated with electrons of several energies. Experimental points are indicated only for the sample irradiated at 24 mev and the background. The observed photopeaks are characteristic of two radioisotopes: radiation induced Na^{22} (0.511 mev and 1.28 mev) and naturally occurring K^{40} (1.46 mev). Comparison of the relative heights of the two photopeaks attributed to Na^{22} with similar peaks for known Na^{22} samples indicates that no other positron emitting isotopes were present in these samples. It is apparent in the figure that Na^{22} radioactivity levels decrease sharply with decreasing electron energy.

Besides that Na^{22} which was observed in some samples of every type of food, only two other gamma radioactivities were observed. The shorter lived sodium product, Na^{24}, was also observed in some samples of every food and Rb^{84} was observed in one special beef sample irradiated at 25 mev. The measured Na^{22} and Na^{24} levels as a function of the irradiating electron energy was given in Tables III and IV. The "less than" signs indicate measured values which were so low that the upper confidence limit was at or below the assumed detectibility limit of 0.001 micro-microcurie and the measured values were therefore considered to be statistically insignificant. For several of these samples, the actual value measured for the induced activity was equal to or less than zero.

As indicated in Report No. 10, an additional induced radioactivity (characterized by a gamma energy of 0.88 mev and a half life of about 30 days) was previously observed in large samples of beef irradiated at 24 mev. From its radiation characteristics it was identified as Rb^{84}. Samples of all foods irradiated in Phase II were carefully examined for this isotope, but it was not present at detectable levels. However, when the one remaining control sample from the beef used in the earlier study was irradiated at 25 mev, the

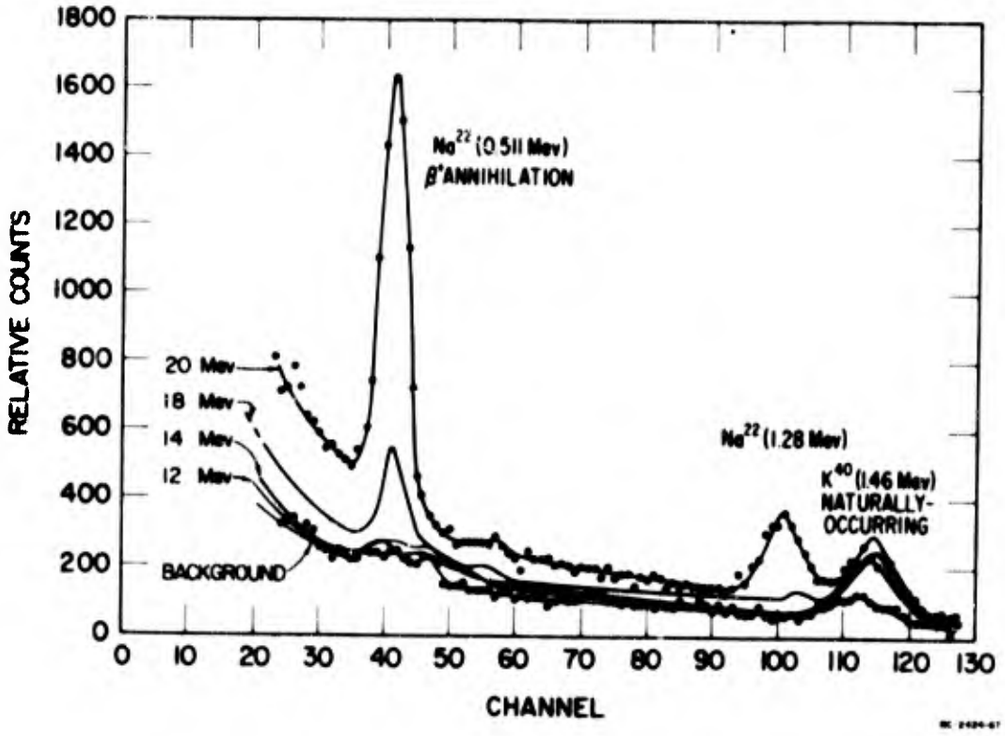

FIG. 2 GAMMA SPECTRA OF IRRADIATED HAM

TABLE III

INDUCED Na^{22} RADIOACTIVITY IN FOODS

Induced Na^{22} Radioactivity ($\frac{\mu curies}{gram\ food\text{-}5Mrads}$)

Food \ Electron Energy (mev)	25	20	18	16	14	12	10	8
Beef	0.17	0.013	0.008	0.004	0.003	<	<	<
	0.13	0.006		0.007		<	<	<
				0.007		<	<	
Average	0.15±0.039	0.010±0.006	0.008±0.005	0.006±0.006	0.003±0.003			
Pork	0.016	0.015	0.015	<	<	<	<	<
		0.036		<		<		<
Average	0.016±0.042	0.026±0.012	0.015±0.010					
Ham	2.2	1.7	0.45	0.043	0.004	<		<
		0.18		0.086		<		<
				0.064				
Average	2.2±0.45	0.94±0.291	0.45±0.185	0.064±0.036	0.004±0.003			
Chicken	0.09	0.011		<	<	<	<	<
		0.046				<	<	<
Average	0.09±0.023	0.028±0.013						
Shrimp	0.16	0.026		0.002	<	<		<
								<
Average	0.16±0.062	0.026±0.015		0.002±0.002				
Beans	0.71	0.034			<	<	<	<
		0.042				<	<	<
Average	0.71±0.163	0.038±0.015						
Peaches				0.003			<	
Average				0.003±0.003				
Composite Diet		0.031	0.018	0.008	0.001	<	<	<
		0.029					<	
Average		0.030±0.014	0.018±0.012	0.008±0.007	0.001±0.001			

TABLE IV

INDUCED Na^{24} RADIOACTIVITY IN FOODS

Induced Na^{24} Radioactivity ($\frac{\mu\mu curies}{gram\ food\text{-}5Mrads}$)

Food \ Electron Energy (mev)	25	20	18	16	14	12	10
Beef	12.0	4.2		0.58			
	12.0			0.55			
Average	12.0±1.32	4.2±0.46		0.57±0.170			
Pork	11.0	3.9		0.43			
				0.31			
				0.23			
Average	11.0±1.21	3.9±0.043		0.32±0.100			
Ham	85.0	29.0		8.9		0.68	
Average	85.0±9.35	29.0±3.19		8.9±1.78		0.68±0.238	
Chicken	7.8	3.4				0.23	
Average	7.8±0.86	3.4±0.37				0.23±0.092	
Shrimp	14.0	7.6		0.52		0.28	
				0.19			
Average	14.0±1.54	7.6±0.84		0.32±0.096		0.28±0.112	
Beans		16.0		4.4			
Average		16.0±1.76		4.4±0.88			
Composite Diet							0.017
Average							0.017±0.008

activity was again observed. The measured value for the induced activity of Rb^{84} of 0.008 micro-microcuries per gram beef per 5 megarads is in reasonable agreement with the previously measured value of 0.005 for the sample irradiated at 24 mev.

Following gamma counting, the samples were dry ashed by heating them at successively increased temperatures up to 600°C. Some of the samples were separated chemically, using the radiochemical procedures presented in Report No. 10. Thin beta-counting samples were then prepared using material from both the chemical fractions and the whole ash.

The results from the beta counting of these samples was at best inconclusive, even though several different approaches were used in the data analysis. It now appears that the major problem was achieving reproducible ashing of the samples, since some differences were apparent in the ash color and in the ratio of recovered ash to whole food for the samples. Since the chemical separation and counting were both referenced to ash weights, inhomogeneity of the ash could cause the observed scatter in the counting results.

While the actual beta counting data are inconsistent, several general statements may be made about the observed results. When extrapolated to a total-radioactivity-per-can basis, the net activity for each whole ash sample was close to or below the value which would be expected for the Na^{22} activity previously determined for the sample. In the chemical fraction samples, the count rate per gram for the control phosphorous and sulfur fractions was always greater (sometimes much greater) than that of the irradiated foods. Finally, no decay of the measured activity level was observed for any ash or fraction sample even though repeated counts were made over a period of several months.

No additional induced activities were observed in the beta counting of the samples.

IV LARGE-SAMPLE BETA COUNTER

The original planning for the Phase II had included the design and construction of a plastic scintillator beta detector capable of counting with reasonable efficiency the entire contents of No. 10 cans (approximately 200 cu. in.). The assembly then envisioned included two large sheets of plastic scintillator which were to be cast or machined to contain a sample volume of the desired dimensions. Since the ranges in unit density material of the beta particles emitted by radioisotopes of interest in the present investigation are so short (generally about 1/8 to 1/4 inch), an average sample thickness of less than 1/4 inch would seem indicated for an efficient detector. To achieve this sample thickness a detector area of about 900 square inches would have been required for each of the scintillator sheets. Scintillator sheets of this size (30 inches x 30 inches) would present obvious problems in fabrication, shielding, and use.

In subsequent consideration of the problems involved in such a large sample beta counter, a unique design was developed for a counter which would lend itself readily to routine beta counting operations. The general configuration for this counter includes a series of parallel slabs of scintillator material, rigidly mounted in such a way that they could be inserted into the material to be examined. There would be two advantages of such a system. Regardless of the system used--sample between two scintillator sheets or a series of scintillators inserted into the sample--the surface area of the scintillation material would, for a given sample volume, be exactly determined by the maximum sample thickness allowed. For the design presently proposed in which the detectors are inserted into the sample, both surfaces of the scintillator material would be used, so that the total amount of scintillator required would be reduced by about a factor of two compared to a flat sheet assembly. The reduced scintillator volume, as well as the increased flexibility in the scintillator geometry, would also simplify attendant shielding considerations. The other advantage of such a system would be the increased facility visualized in its use for routine counting operations.

In the proposed design, each slab would act as its own light pipe carrying the scintillations to a common detection surface on one end. The major problem encountered in such a design is that of securing efficient optical coupling of the series of slabs to the detector (photomultiplier). In general, if normal light piping or direct coupling of the detector to the slabs is attempted, the "effective" surface of the scintillator group at this common end must include the open areas between the scintillator end surfaces. Since this area would be approximately equal to that of the actual scintillator light-pipe surfaces, the average output intensity (photons/cm^2) of the scintillator group would be reduced to about half that of the individual scintillators at this junction. Further, since the light of the scintillators cannot be funneled to increase intensity, the intensity might be further reduced by the ratio of detector surface area to "effective" surface area of the scintillator group, if the detector is smaller than the scintillator. Even at best, light piping or direct coupling would prove to be very inefficient in coupling a detector to this scintillator geometry.

An alternative method proposed by Garwin[18] seems to have some promise for a system of this geometry. He proposed that a slab of some scintillator which has an emission spectrum centered about some wavelength, λ_1, be butted into and optically coupled with a slab of a second scintillator. If the second slab is transparent to the scintillations of the first, photons arriving at the interface would be transmitted essentially unchanged across the second slab and presumably out the opposite side since they would be incident almost normally on the opposite surface. However, if the second scintillator were to strongly absorb the primary scintillations of wavelength λ_1 and reemit the light at some longer wavelength, λ_2, the fluorescent (reemitted) light would be isotropic. Some portion of it (typically approximately one-quarter) would be trapped and carried down the second slab which would now act as a second light pipe. The surface area of the first slab would obviously be the product of its thickness and its width, that of the second slab would be the product of

its thickness and the same width. In practice, the thickness of the second slab would be determined by the thickness of the material needed to absorb some desired fraction of light of wavelength λ_1. If this width could be made sufficiently small and a major portion of the incident light could still be absorbed, the intensity (photons/cm^2) of the reemitted light in the second slab might be near to or even greater than that of the first slab.

The application of the scheme to the proposed beta detector would seem to be beneficial in two ways. The diluting effect of the open areas between the primary scintillators would be removed since the only areas of importance are the cross sectional areas of the individual slabs at each interface. The second slab could also be designed to have a cross section more readily adaptable to existing detectors.

For efficient application of this method of photon energy degredation and intensification it would seem necessary to have strong absorption of the primary wavelength and efficient fluorescence (low self-absorption) in the second slab. To assure these conditions a rather large difference between the primary and secondary wavelength is almost certainly indicated. Since, in the proposed application, photons of the primary wavelength would be generated in and would travel for some distance through the initial plastic detector slabs, a further restriction exists in that the primary photon wavelength must be longer than that of the strong absorption edge exhibited in the ultraviolet by the solvent (or body plastics) more commonly used in plastic scintillators (polystyrene and polyvinyltoluene). These two combined restrictions tend to indicate that efficient application of the proposed scheme is probably not compatible with the S-11 response demonstrated by the more common photomultiplier tubes, and that a detector sensitive at longer wavelengths would be desirable.

One readily available detector type which demonstrates such longer wavelength response is the silicon photodiode. While the overall sensitivity of this type of detector is considerably less than that for photomultiplier devices, it has several characteristics which would

make it more desirable in the present application: very low cost per unit, minimal and simplified accessory electronics, broad and relatively flat spectral response, small size, etc. In practice the difference in sensitivity per unit might be partially made up by using a relatively larger number of the silicon diodes fastened in parallel so that their signals would be additive. The smaller size and greater geometrical flexibility of diodes should also allow more efficient placement of the detectors in the fluorescent material.

The problem to be solved then before the applicability of this scheme can be tested is the selection of a scintillator-wavelength shifter pair with the proper responses. It would seem that one of the organic scintillators normally used in the liquid or plastic scintillation materials would be appropriate for use in the primary detector slabs since their fluorescent radiations are only slightly absorbed in the usual plastic solvent. It is a considerably more difficult problem to locate fluorescent materials for use as a wavelength shifter in the secondary or coupling slabs of the proposed detector. Relatively few investigations have been carried out concerning materials which fluoresce in the wavelength region of interest (7000A to 10,000A), but acridine has shown fluorescence in the region 5000A to 8000A and chlorophyll-b is known to have a strong emission at about 8600A. Other possibilities include materials in the azuline group.

The exact processes involved in energy absorption and fluorescence by materials is apparently very complex and not well known. However, the immediate environment around the fluorescent molecule is known to be involved in these processes. It is important therefore, even after finding compounds with the proper fluorescent wavelength, to determine if they will fluoresce efficiently under the conditions of the proposed detector.

To answer experimentally some of these basic problems, sample slabs of some appropriate material have been ordered from Nuclear Enterprises, Ltd. of Canada. These slabs (4x4x1/4 inches) are of clear polyvinyltoluene in which small amounts of the compounds of interest have been dissolved. For the primary scintillators, samples

have been obtained containing 2-1/2% of p-terphenyl or 1-1/2% of 1, 1, 4, 4-tetraphenylbutadiene. These materials fluoresce much as expected. The commercially available plastic scintillator NE-102 should also work very well as a primary scintillator. For use in the secondary or coupling slabs, samples of polyvinyltoluene have been ordered which contain various percentages of acridine or chlorophyll-b. To now, only the acridine samples have been obtained. Examination of these samples revealed no fluorescence regardless of the exciting wavelength. (It now appears possible that a "fluorescent grade" acridine might not have been used in the preparation of the samples which might help to explain the negative results.)

While no transparent red fluorescing materials have yet been located and no verification of the wavelength shifting scheme has been obtained, the extreme applicability of a detector of the proposed design in the routine beta counting of large samples together with the many other possible applications of the scheme and of red scintillating or fluorescing materials in the fields of basic or applied nuclear physics, would seem to be strong justification for further work in this area.

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V. FURTHER THEORETICAL RESULTS

Several additional computer runs were made using the previously developed theoretical program (Report No. 11) to calculate expected activity levels for electrons of 18 and 12 mev (an energy of 13 mev was used for Na^{22}, since 12 mev is below the energetic threshold for the Na^{22} (γ,n) reaction). A summary of the calculated induced levels of several isotopes in irradiated beef is given in Table V. Previously calculated levels for 24-mev electrons are also included. Figure 3 shows the agreement between computed and experimental levels of induced Na^{22} in beef. The previously noted good agreement (Report No. 11) of the computed activities with experimental values for all isotopes at 24 mev and the present acceptable agreement for Na^{22} at the lower energies would seem to justify some measure of confidence in the theoretical program and in its application as a guide in any further investigations.

TABLE V

SUMMARY OF COMPUTED RADIOACTIVITY LEVELS IN IRRADIATED BEEF

Energy (mev)	Induced Radioactivity ($\frac{\mu curies}{gram\ beef - 5\ Mrads}$)					
	Product Isotope					
	C^{11}	O^{15}	Na^{22}	Mn^{54}	Fe^{55}	I^{126}
24	1.0×10^{6}	3.5×10^{7}	0.17	7.2×10^{4}	2.1×10^{-2}	4.4×10^{-3}
18	— —	5.9×10^{5}	6.3×10^{-2}	2.4×10^{-4}	9.6×10^{-3}	2.5×10^{-3}
12	— —	———	6×10^{-4}	1.3×10^{-6}	1.4×10^{-4}	2.3×10^{-4}

A further application of the computer program was made in an attempt to reproduce the experimental measurements of activities induced in aqueous solutions of food elements by x-rays. For the experimental investigations (Report No. 10) electrons of several successive energies between 12 and 24 mev were stopped in a 3/8-inch thick lead plate. The "forward scattered" bremsstrahlung (those proceeding in the direction of the original electron beam) were then used to irradiate the solution

and the induced activity levels were measured. To apply the theoretical program to this experiment, several steps were required. The transfer and removal coefficients (Code No. 1) and bremsstrahlung source strengths (Code No. 2) were first calculated for the electron beam traversing the lead plate. These results were divided by 10 to account for the density difference between the lead and the usual food medium for which the program was written. These reduced outputs were used (with Code 3) to compute the x-ray spectrum at each depth in the plate. The spectrum at the depth corresponding to the plate thickness was then extracted from the output for this calculation. The transfer and removal coefficients characteristic of aqueous solution mediums were computed (Code No. 1) and, using the x-ray spectrum from the previous calculation, the x-ray spectrum at successive depths in the solutions was determined. (Since all of the high energy electrons were presumed to have been stopped in the lead plate, it was unnecessary to compute the bremsstrahlung source strengths (Code No. 2) for the solutions.) Using the x-ray spectra from the first calculation, target nuclide densities, and reaction cross-section, the induced activities were then computed (Code No. 4). The results of these calculations and the experimental values are compared in Table VI.

TABLE VI

RADIOACTIVITIES INDUCED IN AQUEOUS SOLUTIONS
BY X-RAYS OF VARIOUS ENERGIES

Isotope	Energy (mev)	Induced Radioactivity μμcuries/gram elem-μampmin		Ratio Theoretical / Experimental
		Theoretical	Experimental	
Zn^{63}	18	3.5	0.15	23
Zn^{63}	12	1.4×10^{-3}	2.7×10^{-4}	5.2
I^{126}	18	6.0×10^{-2}	4.2×10^{-3}	14
I^{126}	12	4.2×10^{-3}	8.2×10^{-4}	5.1

The agreement is sufficiently good to be encouraging, since a number of assumptions, some obvious and some subtle, were inherent in the present application of the theoretical program. The major reason for the differences in calculated and experimental values probably results from

the inability to account for the "wide-angle scattering" of x-rays which occurs in the lead plate. Since in this program we assume that nearly all of the x-rays produced in the plate proceed directly forward into the solution, the predicted levels are higher than those actually observed. Even the present application, however, might be of some use in establishing expected upper limits for any induced activities in any proposed x-ray irradiations of foods.

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VI. DISCUSSION OF RESULTS

The experimental data obtained during Phase II establish the approximate levels of radioactivities induced in a number of foods irradiated with electrons of various energies. This information should be generally applicable as one of the basic inputs in designing an irradiation sterilization program for it will aid in the evaluation of any potential health hazard which might be presented by these induced activities. The experimental information further serves to verify in foods and under sterilization conditions the applicability of some simple theoretical considerations of the processes leading to the induced radioactivity.

The major isotope observed in the present work was Na^{22}, which was produced by the (γ,n) reaction on Na^{23}. This isotope was observed in all of the foods at the higher electron energies verifying the general occurrence of sodium as one of the major trace elements in foods. The average induced level of 0.15 (±0.039) micro-microcurie per gram food per 5 megarads measured in beef at 25 mev is in reasonable agreement with the value of 0.26 (±0.13) micro-microcurie previously measured at 24 mev. The sharp decrease in the induced levels of this isotope with decreasing electron energy is indicated in Fig. 3 for ham, pork, and beef. The difference between the level for this isotope measured in ham and that measured in pork and beef is consistent with reported relative abundances of sodium in these foods (Report No. 10). Also indicated in the figure are (1) the Na^{22} levels predicted in beef using the theoretical computer program (Report No. 11) and (2) the reported energetic threshold for the Na^{23} $(\gamma,n$ reaction. It is apparent that the predicted level for 18 mev is above the actual measured level in beef. However, the general shape of a curve through the theoretical values is more consistent with that through the measured ham values. All Na^{22} levels measured in the present investigation are consistent with reported threshold of 12.1 mev, since no Na^{22} activity was observed for irradiations with electrons of 12 mev or below. Reported thresholds for other isotopes may, therefore, be

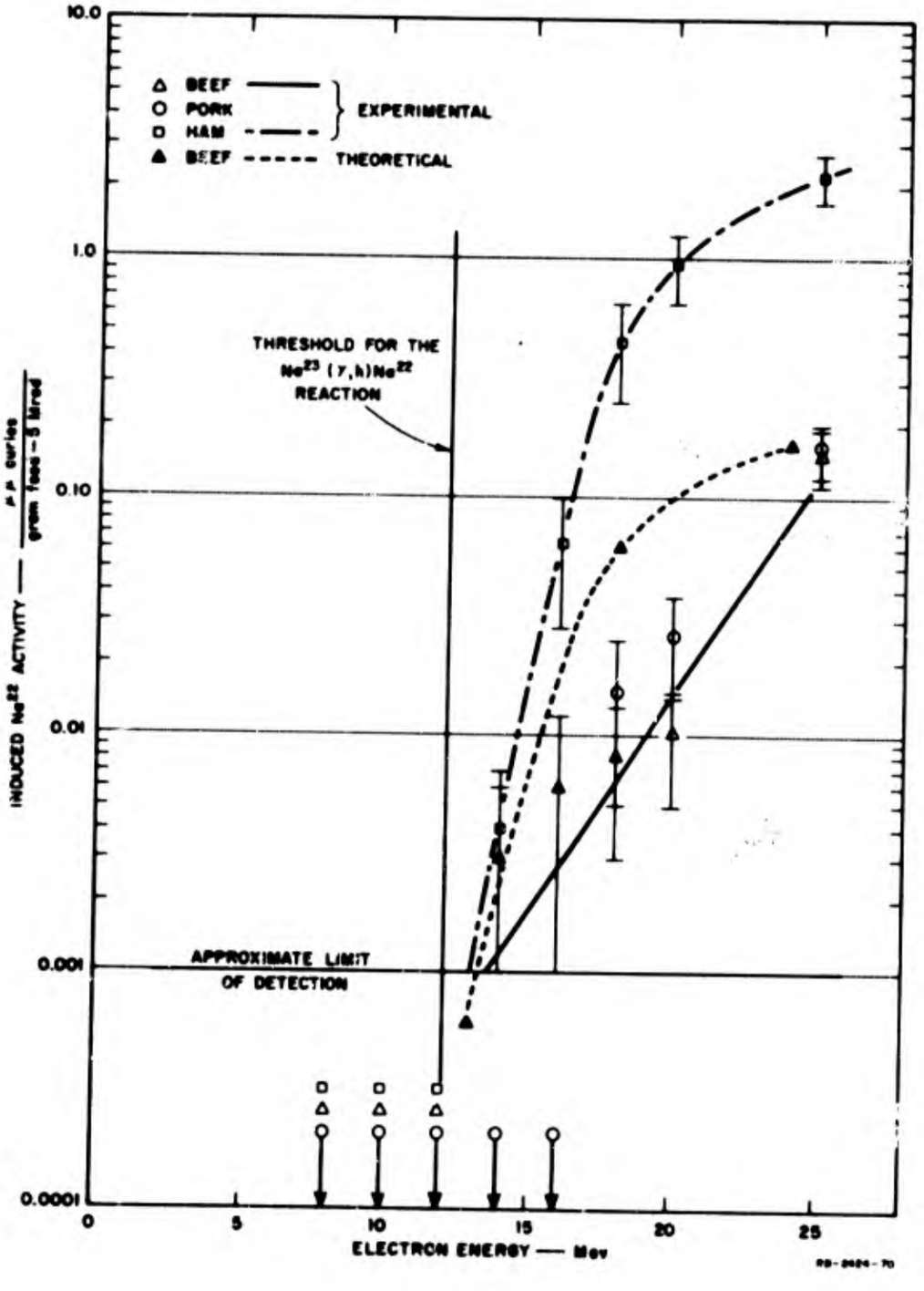

FIG. 3 INDUCED Na^{22} RADIOACTIVITY IN THREE FOODS

applied with some confidence to any other direct-product activities induced in foods. The observed variation in the Na^{22} levels between samples of the same food is probably due, at least in part, to variations in sodium abundance in the samples. A possible explanation is that the water soluble sodium salts remain more in the meats and juices than in the fat portion. If the fat and meat portions were allowed to separate slightly after "homogenizing" and before samples were taken from the large batch for canning, variations in the meat-to-fat content (and the relative sodium abundance) might then occur. Visual observation did indicate a somewhat greater fat layer in some samples than in others.

The other important induced activity observed in the present investigation was Na^{24}. The variation of the measured levels of this isotope with energy is indicated in Fig. 4 for ham, pork, and beef. (The indicated values were obtained by extrapolating measured values back to the end of irradiation, using a 15.1 hour half life.) Also indicated is the value for pork given in a preliminary report on the Vanderbilt University-Tracerlab study of induced radioactivity in radiation sterilized foods[7]

This isotope was not observed directly in foods during Phase I, but it was predicted to be in the foods irradiated at 24 mev as the product of two different reactions. The major portion was expected to result from the (γ,p) reaction in which the bremsstrahlung interact with a magnesium nucleus to cause emission of a proton. This reaction has a threshold of about 12 mev. The induced activity levels predicted as a result of this reaction in beef irradiated at 24 mev would be 2.9 micro-microcuries per gram per 5 Mrads. The other reaction resulting in Na^{24} production is the (n,γ) reaction in which a sodium nucleus absorbs a neutron produced elsewhere in the irradiated medium. Since this is a secondary reaction, the threshold for its occurrence and the level of the induced Na^{24} are not immediately defined, but depend directly upon the source or sources of the activating neutrons.

The relatively high level of the measured Na^{24} activity induced at 25 mev compared to that predicted as a result of (γ,p) reaction

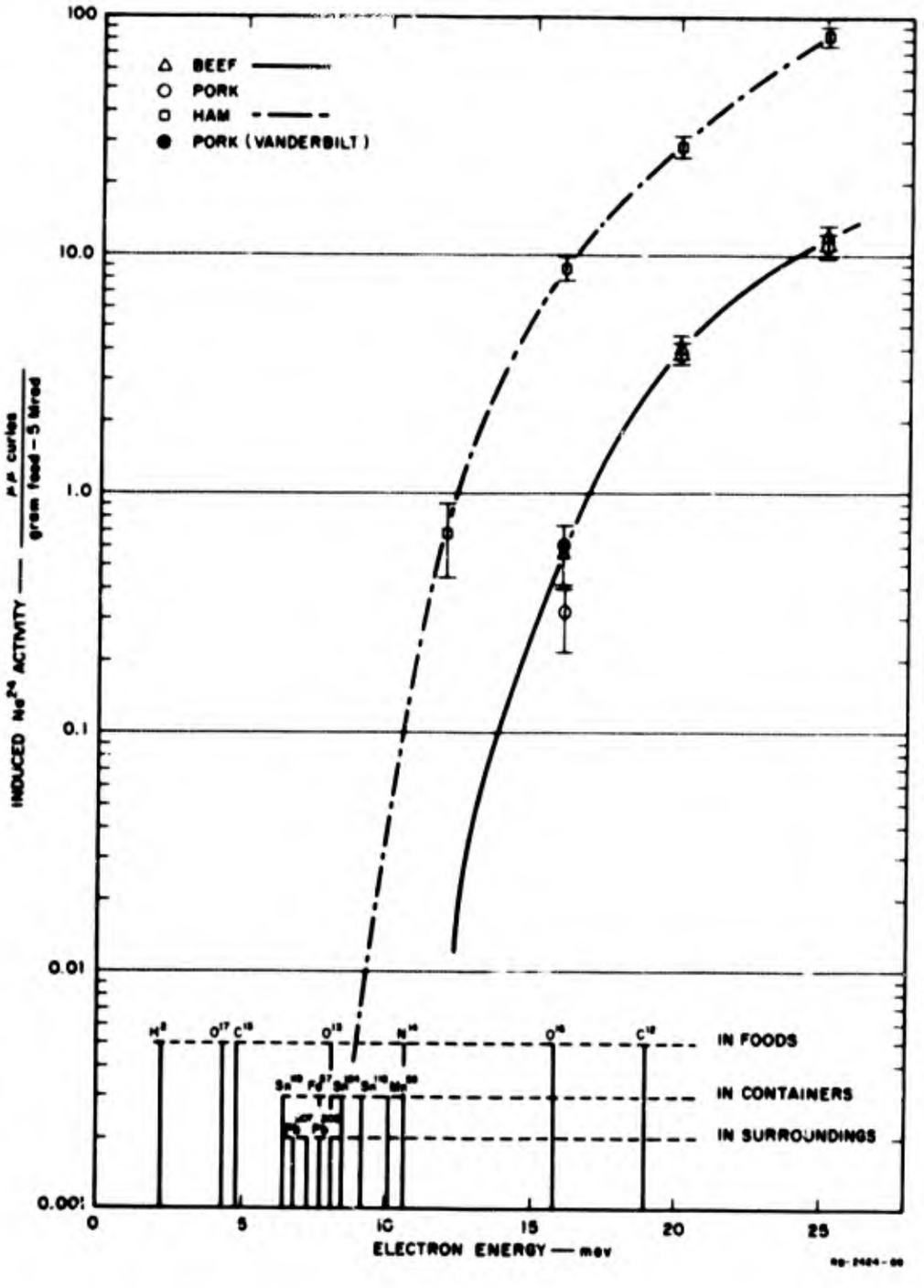

FIG. 4 INDUCED Na^{24} RADIOACTIVITY IN THREE FOODS

would indicate that a major portion of the observed activity resulted from the (n,γ) reaction. (Some of the difference might also be attributed to a difference in the magnesium abundance in the present foods). Further evidence for the (n,γ) reaction is the existence of measurable Na^{24} activity in ham irradiated at 12 mev, the threshold for the (γ,p) reaction. It is impossible from the results of Phase II to determine with certainty the origin of the activating neutrons. However, it is probable that the neutrons are produced in (γ,n) reactions involving several of the elements present in the foods, the containers, or possibly in the immediate surroundings. Reported thresholds for this reaction for some of these isotopes are indicated on the abscissa of Fig. 4. It is apparent that the theoretical absolute threshold for activation may be determined by the deuterium (2.22mev) or the carbon-13 (4.95 mev) which occur naturally in the foods. However, as a result of their relatively small abundance, the practical threshold for detection may be considerably higher. From the present data it is indicated that the induced levels decrease sharply below 12 mev and only one occurrence of Na^{24} production was observed at energies below 12 mev (that in a composite diet sample at 10 mev).

The final induced isotope observed in the present investigation was the Rb^{84} in the one special beef sample. This activity is produced by the (γ,n) reaction. Attempts were made to observe this isotope in irradiated samples of each of the foods supplied by the Quartermaster Food and Container Institute for the present investigation, but it was never present at detectible levels. This isotope was observed, however, in the sample of beef which had been prepared for a previous investigation at SRI. The absence of rubidium activity in the present foods is a further indication of the variation of trace element abundance which must be expected between samples of different foods and even between different samples of the same food. The level measured following irradiation by 25-mev electrons of 0.008 micro-microcurie per gram beef per 5 Mrads is in good agreement with the value of 0.005 micro-microcurie previously measured in the samples irradiated at 24 mev. The reported threshold for the production of

this isotope is 9.3 mev. Using the measured induced levels in the previously developed empirical expression for predicting induced levels (Report No. 10), it is possible to estimate the rubidium abundance in the special beef sample which would have been required to give the measured activity level. The abundance thus estimated is 4 parts rubidium per million parts food. Koch[13] determined a rubidium concentration of 1.3 parts per million in other beef by neutron activation analysis.

On the basis of the results from Phase I, observable levels of the beta-emitting radioisotopes phosphorous-32 (P^{32}) and phosphorous-33 (P^{33}) were anticipated for the whole ash or chemically separated fraction of the foods irradiated at 25 mev; but as previously indicated, neither was apparent in the beta counting data. Attempts to identify these and other radioisotopes by relatively high observed count rates or by decay of the observed radioactivity levels of the appropriate samples were equally unsuccessful.

These radioisotopes were expected as the products of probable (γ,p) reactions in the foods. The P^{32} activity might also result from a (n,γ) reaction. It might be expected, therefore, that the approximate level of these and other possible induced activities could be predicted on the basis of measured Na^{24} levels. The decay of radioactive nuclei is a purely random process but for any specific radioisotope the average decay rate (radioactivity level) is directly proportional to the total number of such nuclei present and inversely proportional to the half life characteristic of the isotope ($t_{1/2}$). The total number of nuclei of any specific type which is produced in a food should, however, be proportional to (1) the abundance of the appropriate parent element in the food (A_f), (2) the abundance of the target isotope in the element (A_e) and, (3) the cross section for the reaction (σ). The induced level of any activity in the food should therefore be proportional to $A_c A_e \sigma / t_{1/2}$. For different activities which are produced by the same general reaction and which have reasonably similar reaction cross sections, the expected relative level (R) in foods irradiated under similar radiation conditions would be given

by

$$R_1 \sim \frac{(A_f A_e \sigma)_1}{(A_f A_e \sigma)_2} \cdot \frac{(t_{1/2})_2}{(t_{1/2})_1} R_2$$

In general the reaction cross section is strongly dependent upon the energy of the interacting photon or particle and the cross section for a given reaction will differ for different elements or isotopes. However, the assumption of a similar energy dependence of the cross section (or even the assumption in some cases of an exactly similar total cross section) is probably sufficiently accurate in the present application to allow approximation of relative induced activity level.

If this method is used to predict the radioactivity induced in beef by the (γ,p) reaction using the trace elements and average abundances given in Report No. 10, only 3 isotopes (besides Na^{24}) are found to give significant expected levels. These isotopes and their predicted activity relative to Na^{24} are P^{32} (3%), P^{33} (9%), and K^{43} (5%). The only significant activities predicted for products of the (n,γ) reaction are K^{42} (70% of Na^{24}) and P^{32} (4%). The two potassium isotopes, K^{42} and K^{43}, each have prominent gamma rays among their characteristic decay radiation and either should be observable at relatively low levels in the gamma spectra. A recheck of the beef gamma spectra specifically for these isotopes indicated that neither was present at detectable levels. The absence of these activities may indicate that the predicted levels actually represent approximate upper limits for these other induced activities. Further information forthcoming from the extensive beta counting portion of the Vanderbilt University-Tracerlab study[7] may help define this portion of the induced radioactivities problem.

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VII. RECOMMENDATIONS AND CONCLUSIONS

The levels of radioactivities induced in irradiated foods are now well outlined for a variety of irradiation conditions. The information obtained in the investigation reported here should be useful in evaluating any potential health hazard presented by the induced activities and in designing a food sterilization program. Further information may be required eventually on the occurrence and variation of some trace elements in the foods (such as the variation observed in rubidium in beef). This information could be obtained by activating the trace elements with either electrons or neutrons. It may also be desirable at some time to establish more exactly the levels of the induced beta-emitting isotopes and to determine the relative significance of the (γ,p) and the (n,γ) reaction in the production of radioactivities.

As the reader considers the specific results of this investigation, he should remember the extreme sensitivity of current gamma detecting apparatus and techniques. It has been possible in this investigation to detect and measure gamma radioactivities at levels as low as a few disintegrations per minute in the entire contents of the No. 10 can. By allowing longer counting times or by counting sample ash or chemically separated fractions, it should be easily possible to lower this sensitivity to fractions of a disintegration per minute. This sensitivity is equivalent to detecting and identifying a few specific nuclei in the approximately 10^{25} nuclei which are present in a typical No. 10 can of food and is, therefore, almost incomparably greater than that of the methods commonly used to identify other potentially carcenogenic additives in foods.

The determination of the biological significance of these measured induced radioactivities and the eventual establishment of maximum acceptable levels of radioactivities in foods are very difficult problems, the solutions of which must properly be left to workers in the various life sciences. These solutions require evaluation of interactions between many complex factors such as the physical and biological half life of the radioisotope, its biological fate in the

body metabolism, and the mode and energy of its decay process. Also to be considered are the relative effects of other radioactivities normally occurring in unirradiated foods. These activities include naturally occurring carbon-14 (1.5 micro-microcuries per gram food), potassium-40 (2.0 micro-microcuries per gram food), tritium (0.001 micro-microcurie per gram food), and smaller trace amounts of such heavy element radioisotopes as radium-226.[14] Some foods also contain fallout activities cesium-137 (0.16 micro-microcurie), strontium-90 (0.001 micro-microcurie), and zinc-65 (0.02 micro-microcurie).[15]

At least one group, The National Committee on Radiation Protection and Measurements, has been organized to consider these problems. This group, which was organized by the National Bureau of Standards, includes authorities from both the medical and physical science fields, as well as representatives from the various professional societies and government agencies. Their original recommendations of maximum permissible concentrations (MPC) of radionuclides in air and water were given in National Bureau of Standards Handbook 52 (1953). These recommendations have recently been revised and reissued in Handbook 69(1959). While these values carry no legal implications and are subject to revision as more is learned of the biological effect of ingested radioactivity, the values represent one of the few attempts to establish such limits based on a reasonable interpretation of the best information currently available. It is of some interest therefore to note the MPC values which are recommended for the drinking water of the population at large for the isotopes measured in the present investigation. The recommended value for Na^{22} is 400 micro-microcuries per gram and the value for Na^{24} is 2,000 micro-microcuries per gram. No value is reported for Rb^{84} but a provisional maximum permissible concentration for unidentified radionuclides in water is set at 0.1 micro-microcurie per gram. It may be seen that these MPC values are at least a factor of 20 higher than the highest level measured in foods for the shortlived Na^{24} and Rb^{84}. For Na^{22} the highest measured level is approximately a factor of 200 below recommended MPC value. For foods irradiated at lower electron energies, the measured value decreased

even further below the MPC values. The MPC values for Na^{24} and Na^{22} are factors of 1000 and 100,000 higher respectively than the highest levels measured for these isotopes in foods irradiated at 14 mev.

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APPENDIX A

ERROR ANALYSIS

In addition to the continuous decrease in the average emission rate of the radioactive sample with a half life that is appropriate to the isotope or isotopes contained in the sample, the instantaneous rate is continuously fluctuating as a result of the random nature of the disintegration process. Since the phenomena of nuclear decay consists of a number of individual disintegrations which are completely independent of one another, the laws of probability may be applied to the process to insure a more correct interpretation of any results obtained in counting experiments.

If the same sample or identical samples are counted many times, resulting measurements of counting rate will, in general, all be different and will be randomly distributed about an average value according to some distribution function. From the distribution, it is possible to make some deductions about the consistency of the data and the probable error. Similarly, if a sample is counted only once, a distribution function can be derived which would indicate the probability that the correct value differs from that measured in the experiment by a given amount. For each case, the distribution would in general follow the familiar bell shape of the "normal" distribution. In measurements for which the statistics were relatively poor, the distribution would be low and broad. For measurements with better statistics the curve would be taller and more narrow.

In counting experiments the probability function would be best approximated by the Poisson distribution:

$$P(n) = \frac{e^{-A} A^{n}}{n!}$$

where n is the number of counts measured in time t, and A is the true average count for time t. When A is relatively large (greater than 100) the Poisson distribution may in turn be approximated by the Normal or Gaussian distribution given by

$$G(n) = \frac{1}{(2\pi)^{1/2}\sigma} \; e^{-\frac{(n-A)^2}{2\sigma}}$$

The constant quantity preceding the exponential is a factor to normalize the integral of the expression (between $-\infty$ and $+\infty$) to unity. The quantity σ in this expression is the standard deviation and is related to the relative broadness of the distribution curve. It happens in counting experiments that the true average value, A, and the standard deviation, σ, are not independent ($\sigma = A^{1/2}$) so that the standard deviation may be determined directly from the counting data.

While the standard deviation is, therefore, inherent in a counting measurement, it is not generally directly given as an expression of the accuracy of the measurement. Instead, the convention of establishing "confidence limits" has been adopted. For a series of measurements on the same sample, these limits define an interval centered about the true average value which should contain a given percentage (equal to the degree of confidence) of the measured values. For a single measurement on a sample, the limits define an interval centered about the measured value in which there is the indicated confidence of finding the true average value. The degree of confidence given by specific limits may be determined by integrating the previously given expression for a normalized Gaussian distribution within the specified limits. As would be expected, then, confidence limits and standard deviation are closely related. The confidence limits which characterize a given degree of confidence G_c are given by $G_c = \pm u\,\sigma$. The values of u appropriate to given confidence limits are given in Table A.1

Table A.1

Confidence Limits Related To Standard Deviation For Normal Distribution

u	0.6745	1.000	1.645	1.960	2.576	2.807
G_c	0.500	0.683	0.90	0.95	0.99	0.995

The final result of interest in most counting experiments is not usually just the observed counting rates and must, instead, be computed from a set of such experimental measurements. It is, therefore, of some importance to determine how the errors in the measured quantities propagate to give the error in the computed quantity. This information is generally obtained by performing the partial differentiation of the computational expression with respect to the contributing variable, the partial deriviatives in each case indicating the dependence of the computed quantity on a specific variable. For sum and difference equations, the differentiation immediately indicates the independence of the errors in the meas d quantities and the direct manner in which the errors propagate. Fo oducts, quotients, and other complex expressions the partial derivatives for each variable will contain the measured value of the other variables, indicating a dependence of the error in one variable on the values of the other variables. This dependence may be clarified by dividing these partial derivitives by the original expression; the results then indicate that for such expressions the fractional errors and not the actual errors are propagated. Since positive and negative errors in the measured quantities are usually equally probable, a more accurate approximation to the propagated error is obtained as a square root of the sum of the squares of the contributing errors. A summary of the error propagation for expressions of interest in the present work is given in Table A.2. More complete discussion of Statistical analysis may be found in the literature.[16,17]

TABLE A.2

ERROR PROPAGATION IN COMMON FUNCTIONS

Function	Standard Deviation
$A = B \pm C$	$\sigma_A^2 = \sigma_B^2 + \sigma_C^2$
$A = \frac{BC}{D}$	$\frac{\sigma_A^2}{A^2} \sim \frac{\sigma_B^2}{B^2} + \frac{\sigma_C^2}{C^2} + \frac{\sigma_D^2}{D^2}$
$A = aB + C$	$\sigma \sim a^2\sigma_B^2$

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REFERENCES

1. Meinke, W. W. Does Irradiation Induced Radioactivity in Food. Nucleonics 12 (10), 37 (1954)

2. Kruger, P. Determination of Neutron Dosages by Food Irradiation Devices, U. S. Army Quartermaster Research and Engineering Command, Quartermaster Food and Container Institute, Report No. 12 (Final)on Contract DA 19-129-QM-741, May 1960 prepared by Nuclear Science and Engineering Corp.

3. Skaggs, L. S. Radioactive Products Produced in Foods Sterilized by Electrons in the Energy Range Above 10 mev. U. S. Army Quartermaster Research and Engineering Command, Quartermaster Food and Container Institute, Report No. 5 (Annual) on Requisition 49-106-134-55 prepared by The Argonne Cancer Research Hospital, University of Chicago.

4. Herschman, A. An Estimate of the Maximum Induced Radioactivity Caused by Electron Irradiation of Foods, U. S. Army Quartermaster Research and Engineering Command, Quartermaster Food and Container Institute, Report No. 1 on Project 7-84-01-001 (Internal) File No. 0203a0 October 1956

5. Rich, M. Effect of 18-mev Electrons on Foods. Ph.D. Thesis, Massachusetts Institute of Technology. June 1951.

6. Ovadia, J., F. Heinmetz, and A. Herschman. The Problems of Induced Radioactivity in the Use of High Energy Electrons (25 mev) for Sterilization and Chemical Processing. Proc. 2nd United Nations Int'l. Conf on Peaceful Uses of Atomic Energy, 27, (Isotopes in Agriculture) paper P/807 USA, p. 418, September 1958

7. Meneely, G. R. Radioisotopes in Radiation Processed Foods. U. S. Army Medical Research and Development Command, Office of the Surgeon General, Report No. 1 on Project DA-49-193-MD-2101, March, 1961, prepared by Vanderbilt University.

8. Glass, R. A. and H. D. Smith. Radioactive Isomer Production in Foods by Gamma Rays,prepared by Stanford Research Institute for U. S. Army Quartermaster Research and Engineering Command, Quartermaster Food and Container Institute, Report No. 3 (Final) on Contract DA-19-129-QM-1511, October, 1960.

9. Glass, R. A. and H. D. Smith. Radioactivities Produced in Foods by High Energy Electrons, prepared by Stanford Research

Institute for U. S. Army Quartermaster Research and Engineering Command, Quartermaster Food and Container Institute, Report No. 10 on Project DA-19-129-QM-1100, January 1960

10. Newkirk, W. H., H. D. Smith, and R. A. Glass. Radioactivities Produced in Foods by High Energy Electrons: Theoretical Analysis and Computer Calculations, prepared by Stanford Research Institute for U. S. Army Quartermaster Research and Engineering Command, Quartermaster Food and Container Institute, Report No. 11 on Project DA-19-129-QM-1100, April 1960.

11. Snyder, O. P., Jr. The Design of a Low Level Food Radioactivity Detector, U. S. Army Quartermaster Research and Engineering Command, Quartermaster Food and Container Institute, Report No. 1 on Project 7-84-01-002 (Internal), June, 1960.

12. Garwin, R. L. Rev. Sci. Instr., 31, 1010, (1960)

13. Koch, R. C. Activation Analysis of Foods for Characterization of the Induced Radioactivities, prepared by Nuclear Science and Engineering Corp. for U. S. Army Research and Engineering Command, Quartermaster Food and Container Institute, Report No. 3 (Final) on Project DA-19-129-QM-1601, February 1961.

14. Lowder, W. M. and L. R. Solon. Background Radiation--A Literature Search. NYO--4712, U. S. Atomic Energy Commission, New York Operations Office, Health and Safety Laboratory, July 1956.

15. Van Dilla, M. A. Zinc-65 and Zirconin-95 in Foods, Science, 131, 659, March 1960.

16. Jarret, A. A. Statistical Methods Used In The Management of Radioactivity With Some Useful Graphs and Nomographs. AECU-262, U. S. Atomic Energy Commission, Oak Ridge National Laboratory, June 1946.

17. Jaffey, A. H. Statistical Tests for Counting, Nucleonics, 18, (11), 180-184, November 1960.