TECHNICAL REPORT
FD-6

INDUCED RADIOACTIVITY IN FOOD AND ELECTRON STERILIZATION

by

Richard A. Meyer, Ph. D.
Radiation Sources Branch

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U. S. Army Materiel Command
U. S. ARMY NATICK LABORATORIES
Natick, Massachusetts

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Project Reference:
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FOREWORD

This report is a compendium on experimental work done on sterilization of food by high energy electrons to delineate those circumstances under which induced radioactivity is possible when energy above a threshold value is employed.

Several other related problems have been reviewed and analyzed in an effort to gain knowledge of the overall effects of food sterilization by high energy electrons. Among these are: Neutron fluxes present during irradiation, elements present in food in concentrations of parts per billion or less, radioactivity which is induced in the food containers during electron sterilization at energies above threshold, and efforts at theoretically predicting amounts of induced activity in food. It was possible to conclude that no significant amount of tritium is produced in irradiated foods during electron sterilization up to 30 MeV. Also, it was possible to negate isomer activation as a cause of radioactivity in electron sterilized food.

From the results of this report, it can be stated that no induced radioactivity can be measured in foods ten days after sterilization by electrons at energies below 12 MeV. Further research is under way to determine the threshold energy for the production of detectable activity.

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CONTENTS

CONTENTS (con't)

LIST OF FIGURES

LIST OF TABLES

LIST OF TABLES (con't)

ABSTRACT

An extensive survey has been made of the literature on activity induced in foods by the use of high energy electrons. This survey indicates that food irradiated with 24 MeV electrons contains less than a five percent increase in the radioactivity level over that present before irradiation. A compilation of the experimentally measured amount of activity induced in various foods as a function of initial electron energy is presented in various tables. An equation has been developed which will predict the average amount of induced activity as a function of initial electron energy, dosage, and elemental abundance. This equation yields results within the variation of trace elements in foods. To aid in the use of these equations, tables of trace element composition in various foods are presented. These were compiled for use with equations developed in the text due to a lack of any single source of food trace element composition in the literature.

That there is no tritium activity induced in foods has been found by several investigators. Even with the use of metallic targets, it was only possible to set an upper limit on the yield from the (γ,H^3) and (n, H^3) reactions. Isomer activation was also originally thought to be a possible source of activity induced in foods. However, isomer activity has not been detected in foods even from irradiation with high energy electrons. Only by resorting to pure elemental targets was it possible to set upper limits on the amount of isomer activity that might be produced. From the experimental data compiled, an empirical equation was developed to predict the amount of isomer activity produced in a particular food as a function of initial electron energy and dosage.

Although activity induced in cans is not a food consumption problem, the amount of activity present immediately after irradiation can produce a handling problem. It appears that the best solution to this problem may be the use of aluminum cans or an aluminized plastic packaging material.

In general it was found that sterilization of food with 24 MeV electrons will produce a slight increase in the activity level of the food. Such an increase is insignificant when compared to the natural activity in food or the two to ten fold increase in activity by the use of certain food additives. It can be stated that no radioactivity is induced in foods up to around 14 MeV. Current research is aimed at determining this threshold value.

INDUCED RADIOACTIVITY IN FOOD AND ELECTRON STERILIZATION

1. Introduction

Radiation sterilization of foods

Since very early times man has sought ways to store his food supply without spoilage for future consumption. As technology has developed, he has tried and applied many of his new found techniques to the preservation of food. One of the most recent techniques is radiation sterilization for the preservation of food. These are the use of gamma rays, from sources such as Co-60, and ionizing radiation such as electrons from linear accelerators. The former method is presently being used for the preservation of some foods while the use of electrons is fast approaching commercial feasibility. There are, however, some problems that must be overcome if electron sterilization is to be acceptable. One of these is the topic we are concerned with here, the radioactivity that might be induced in foods.

Before discussing the radioactivity that might be induced in foods as a consequence of high energy electron sterilization, we should consider several questions. These are: what type of food would be irradiated, how much irradiation would be necessary to sterilize food, and what sort of radioactivity level is present in food as a consequence of natural activity. The answers to these shall be discussed now and serve as a background to later discussion.

Foods that would be irradiated

To answer the question of what types of food will require radiation sterilization, consider the average intake of food as given in Table 1. Of the items listed, dairy products subjected to sterilizing doses of ionizing radiation have been found unacceptable to taste panelists; vegetables and fruits are generally subjected to pasteurization doses of radiation because of undesirable texture and flavor changes at sterilizing doses. It should be pointed out that the latter category has had its shelf life prolonged by light dosages of radiation, for example the inhibition of root formation on potatoes. The only major items left are meats, fish, and poultry. At present these items are the main concern; of course, this does not preclude use of radiation sterilization of the other food types.

TABLE 1

AVERAGE INTAKE OF FOOD PER DAY IN THE U.S.[a]

Component	Percent Total Weight
Water	33.09
Meat, Fish and Poultry	14.49
Milk Products	13.95
Root Vegetables	10.62
Leafy, Green, and Other Vegetables	8.90
Fruit	8.17
Cereal and Grain	7.28
Eggs	3.47
Beverages (tea and coffee)	0.03

[a] Calculated from data given in Reference 47.

Amounts of irradiation necessary to sterilize foods

The amount of radiation, or dose, given to a food item depends upon the extent of biological kill desired. For items upon which pasteurization effects are required, relatively low dosages are given; while for items in which sterilization is necessary, high dosages are required. The sterilization dosages presently used in meats to insure safety from *Clostridium botulinum* are on the order of two to five megarads*, while the dosage for pasteurization varies from 0.01 to 0.5 Mrads depending upon the product.

When electrons are used for sterilization, their penetration becomes a limiting factor since they rapidly lose their energy in matter of unit density such as food. The requirement of nearly uniform dosage within an irradiated food package further limits the thickness of package. In general, for irradiation from one side, the energy of the impinging electrons in MeV must be approximately three times the thickness of the matter to be irradiated in cm. Consequently, as larger and larger packages of food are irradiated, higher and higher electron energies are required. This is where radioactivity enters the picture, for as the energy of electrons increase, the possibility of nuclear reaction occurs.

* Recall that: A rad is equivalent to the deposition of 100 ergs per gram of absorber.

Problem of ingestion and of handling

As far as food irradiation is concerned, the activity that will be produced can be divided into two categories: Those long-lived activities that constitute a potential ingestion hazard and those short-lived activities that constitute a handling problem immediately after irradiation. The latter can be circumvented by automatic packaging and storage devices. It is on the former, that of long-lived activity, where the attention must be focused. The activities with half lives greater than a few weeks have the possibility of remaining in significant levels at the time of consumption. In general, the level of activity will be given as picocuries* per gram of food throughout this discussion.

Radiation levels of foods and additives from natural sources

Cognizance should be taken of the natural activity, i.e., the level of activity present in food without any kind of treatment, before considering the amount of activity induced by radiation. The two major sources of activity in foods are fallout and natural potassium-40 activity. Most of the fallout activities picked up by foods are short-lived enough so that they disappear from the foods soon after the nuclear detonation has occured. However Sr^{90} with a half life of 28 years and 30-year Cs^{137} remain in rather constant levels for very long periods of time. Typical values for levels of fallout activity are given in Table 2. That the level of activity of K^{40} is far greater than present levels of either Sr^{90} or Cs^{137} can be realized by comparing the values given in Table 3 with those given in Table 2. The level of K^{40} can be increased further to concentrations as much as tenfold by the use of certain food additives. Typical values for these additives are given in Table 4. From these Tables the average activity present in untreated food is 1 to 3 picocuries per gram of food while certain additives can increase this amount tenfold. Such levels, as will be shown, are far above levels added by irradiation, even with the highest energy electrons and the largest dosages, viz., food irradiated with 24 MeV electrons to a dose of 5 Mrads.

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* Recall that: 1 pc = 1 picocurie = 10^{-12} curie = 2.22 disintegrations per minute.

TABLE 2

FALLOUT ACTIVITY IN VARIOUS FOODS

(activity in pc/gm food)

Food	Cs^{137} ($x\ 10^2$)	Sr^{90} ($x\ 10^3$)
Meats	8.5	2.1
Sea Foods	7.0	1.4
Eggs	2.4	6.8
Fruits	3.7	2.9
Cereals	1.2	1.2
Dairy Products	0.2	3.9
Milk	8.8	26.2
Leafy Vegetables	2.2	14.3
Root Vegetables	2.2	7.4

Source: References 44-47, 49

TABLE 3

NATURAL POTASSIUM-40 LEVELS IN FOODS

(activity in pc/gm food)

Food	Activity	Food	Activity
Beef	2.5 to 3.5	Iodized Table Salt	0.02
Pork	2.1	Mustard	9.1
Ham	1.6	Pepper, Black	0.4
Chicken	1.6	Curry, Powder	.18
Shrimp	0.8	Ginger, Ground	8.7
Beans	1.4	Molasses	12.0
Peaches	0.9	Honey	0.5

Source: Reference 37

TABLE 4

NATURAL POTASSIUM-40 CONTENT IN FOOD ADDITIVES

Potassium Compound	pc K^{40}/gm	Process used in	Food used in	Increase in food activity (pc/gm)
Acid Tartrate	171	Acid Buffer	Baking Powder	0.43
Bicarbonate	322	Alkali	Cocoa Products	1.6
Carbonate	370	Coloring	Confections	9.7
Citrate	298	Emulsifier	Confections	3.2
Hydroxide	567	Coloring	Confections	2.8
Chloride	430	Brewing	Beer	0.028
Bisulfite	268	Preservative	Wine	0.0067

Source: Reference 37

MPC values of isotopes

The levels of activity that are presently accepted as the upper limit of allowable intake of radioactivity without probability of harm have been set by the National Committee on Radiation Protection. These quantities, called Maximum Permissible Concentrations (abbrev: MPC) are given in the National Bureau of Standards Handbook 69. A few of the more important activities that have a possibility of being found in irradiated foods are quoted in Table 5 for reference purposes. These values are for a 168-hour week and have been adjusted for non-radiation workers, viz., the general public.

TABLE 5

MPC VALUES FOR NON-RADIATION WORKERS AND A 168-HOUR WEEK

Isotope	Critical Organ	MPC in pc/gm*	
		Critical Organ Value	Total Body
Na^{22}	Total Body	40	40
Na^{24}	G.I.	200	400
P^{32}	Bone	20	90
Ca^{45}	Bone	9	70
Rb^{87}	Pancreas	100	200
Sr^{90}	Bone	0.1	0.4
I^{126}	Thyroid	2	200
Cs^{137}	Total Body	20	20

Source: Reference 15

* These were given originally as µc/gm of water in Reference 15.

Sensitivity of radioactivity detection techniques

While considering the levels of activity found in the various foods, one important factor should be kept in mind: the extreme sensitivity of detection for radioactivity over other means of analysis. In general, a careful analysis can detect activities as low as a fraction of a picocurie per gram of irradiated food. This is equivalent to being able to detect a few specific nuclei in the approximately 10^{25} nuclei which are present in a number 10 can of food. This is an incomparably greater sensitivity to any potentially carcinogenic additives than any other means such as chemical analysis which yields impurities in parts per 10^6 or spectroscopic means with a maximum sensitivity of approximately one part in 10^9.

2. Methods and procedures

Detection methods

For the detection of the radioactivity produced in foods, the two detection methods used were gamma ray spectroscopy and electron counting. Since the former method affords a more definite identification of the radioactivity, it was used more frequently. The latter method, electron counting, was employed by a few investigators to measure the amounts of beta decay products present.

Sample preparation: 4 methods

Several methods of sample preparation were used, depending upon the level of detectability of radioactivity per gram of food:

1. counting the food without modifying its physical state
2. post-irradiation ashing of the sample and counting of the residue
3. adding known amounts of elements to the food prior to irradiation
4. using aqueous solutions of the elements known to be trace constituents of food.

The first method yields a minimum detectability of a few picocuries per gram of food and the second yields as low as 0.003 pc per gram of food. The last two methods are used more for estimating upper limits of induced activity and yield a minimum detectability of 10^{-6} pc per gram of food.

Energy of electrons used

For detecting radioactivity induced by electron irradiation, the lowest energy electrons used by investigators were approximately 10 MeV. This is the energy region of the threshold of radioactivity production from the (γ,n) reaction. In most cases, the highest energy electrons used for irradiation of foods was 25 MeV; this is the energy at which the cross section has again become small after passing through a maximum (c.f. Figure 1).

The radioactivity in food does not even become detectable until the electron energy is above 15 MeV(1 to 9, 13). This is due to the slow rise of most cross sections with increasing energy as well as the bremsstrahlung distribution (see Figure 2). Theoretically, we would expect the radioactivity in food to increase sharply with increasing electron energy after the radioactivity becomes detectable, then more slowly at energies higher than point E in Figure 2. That such is the case experimentally is evident in the plot of radioactivity of Na^{22} versus electron energy as shown in Figure 3.

3. Isotopes found after induced radioactivity

Na-22, the dominant isotope, long-lived

The gamma ray spectra from electron-irradiated foods are dominated by one activity, Na^{22} produced from the reaction Na^{23} (γ,n) Na^{22}. The levels of this activity are shown in Figures 3 and 4. Glass and Smith (1) as well as Skaggs(5) have performed analysis by ashing methods on the induced activity from 25 MeV electrons in several foods. The only detectable gamma activity present several days after irradiation was Na^{22}. A summary of the values they found is presented in Table 6. The overall average for Na^{22} activity produced in beef with 24 MeV electrons is 0.21 ± 0.04 picocuries per gram of food per 5 Megarads; while at 14 MeV the Na^{22} activity could only be estimated at 0.003 ± 0.003 pc/gm food/5 Mrads(3). Rich(9) found no evidence of radioactivity when food was irradiated with electrons of 16 MeV and Meneely(13) found no detectable activity below 11.2 MeV.

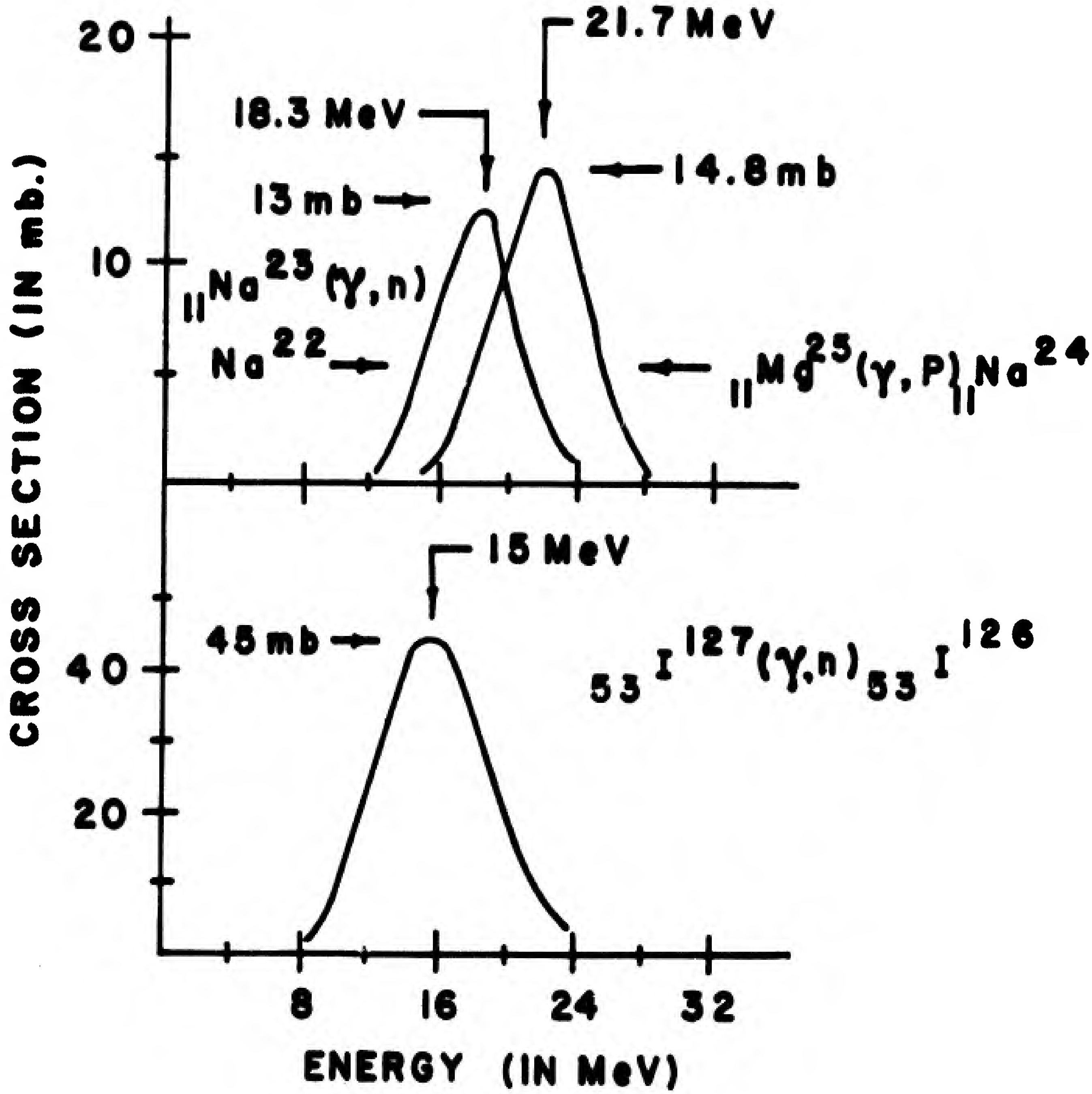

FIGURE 1

CROSS SECTIONS OF THE MORE COMMONLY ENCOUNTERED NUCLEAR REACTIONS IN FOOD STERILIZATION BY HIGH ENERGY ELECTRONS

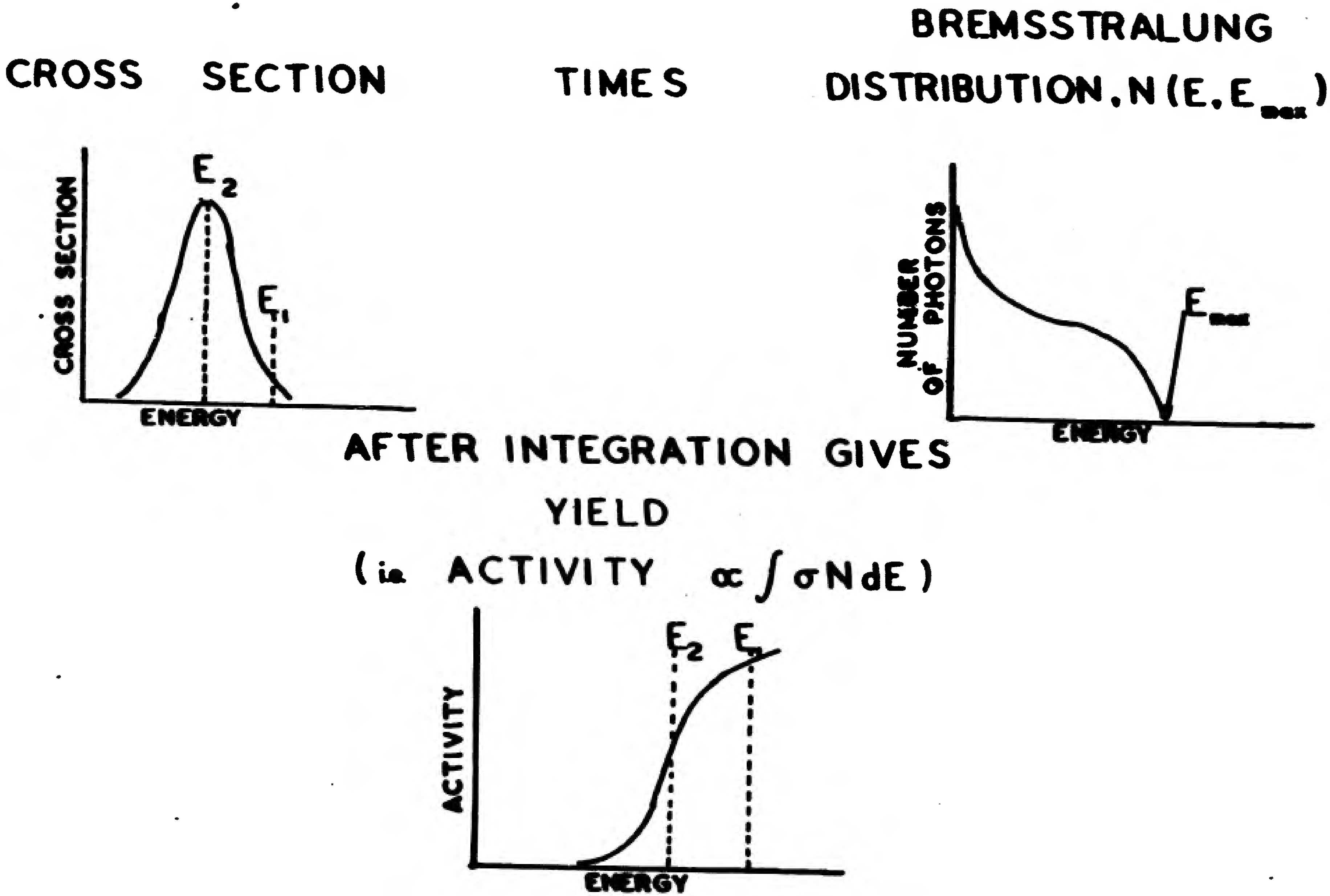

FIGURE 2

LEVEL OF RADIOACTIVITY AS A FUNCTION OF ELECTRON ENERGY AND ORIGIN OF THE YIELD CURVE

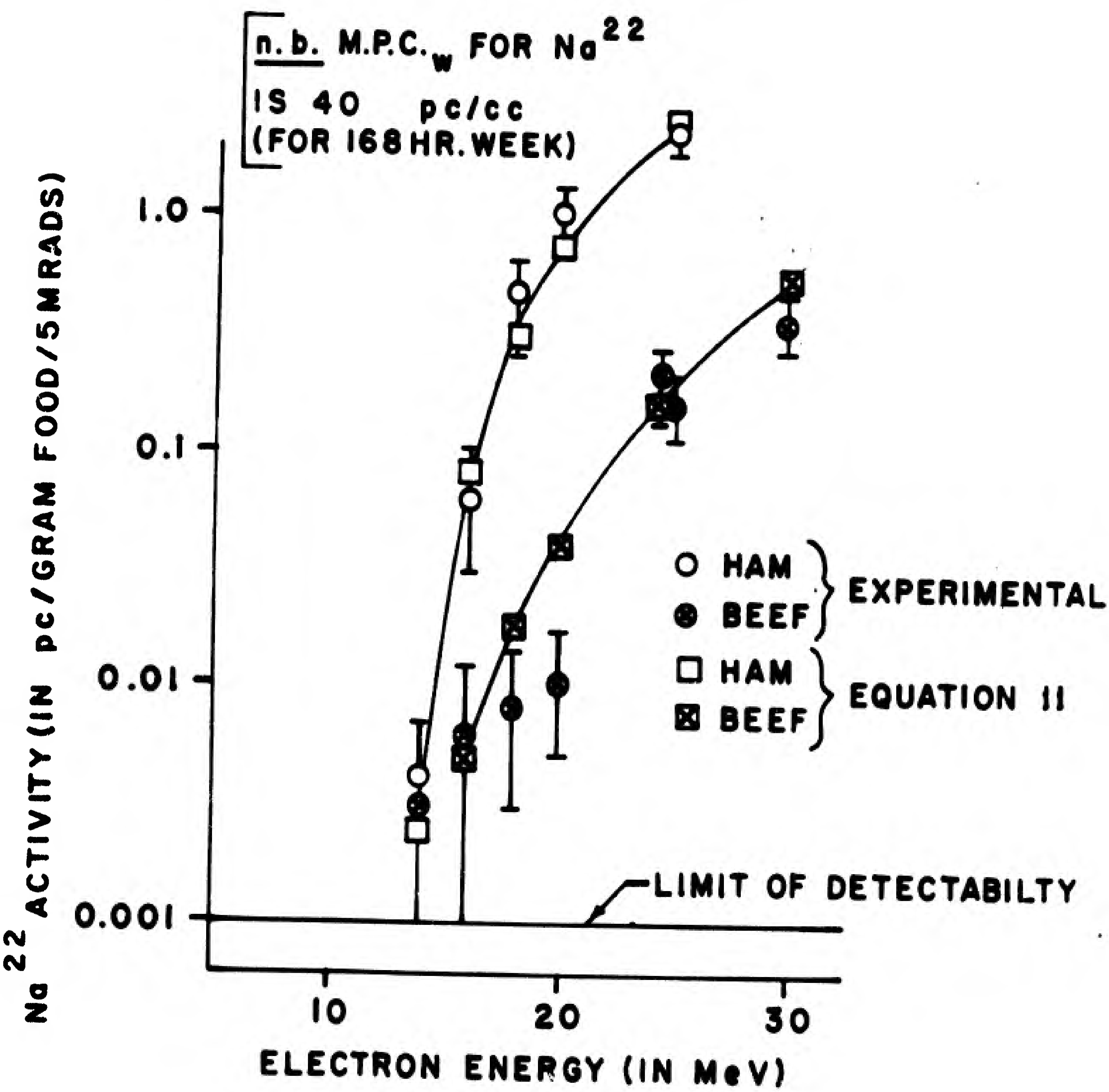

FIGURE 3

SODIUM-22 ACTIVITY INDUCED IN FOOD

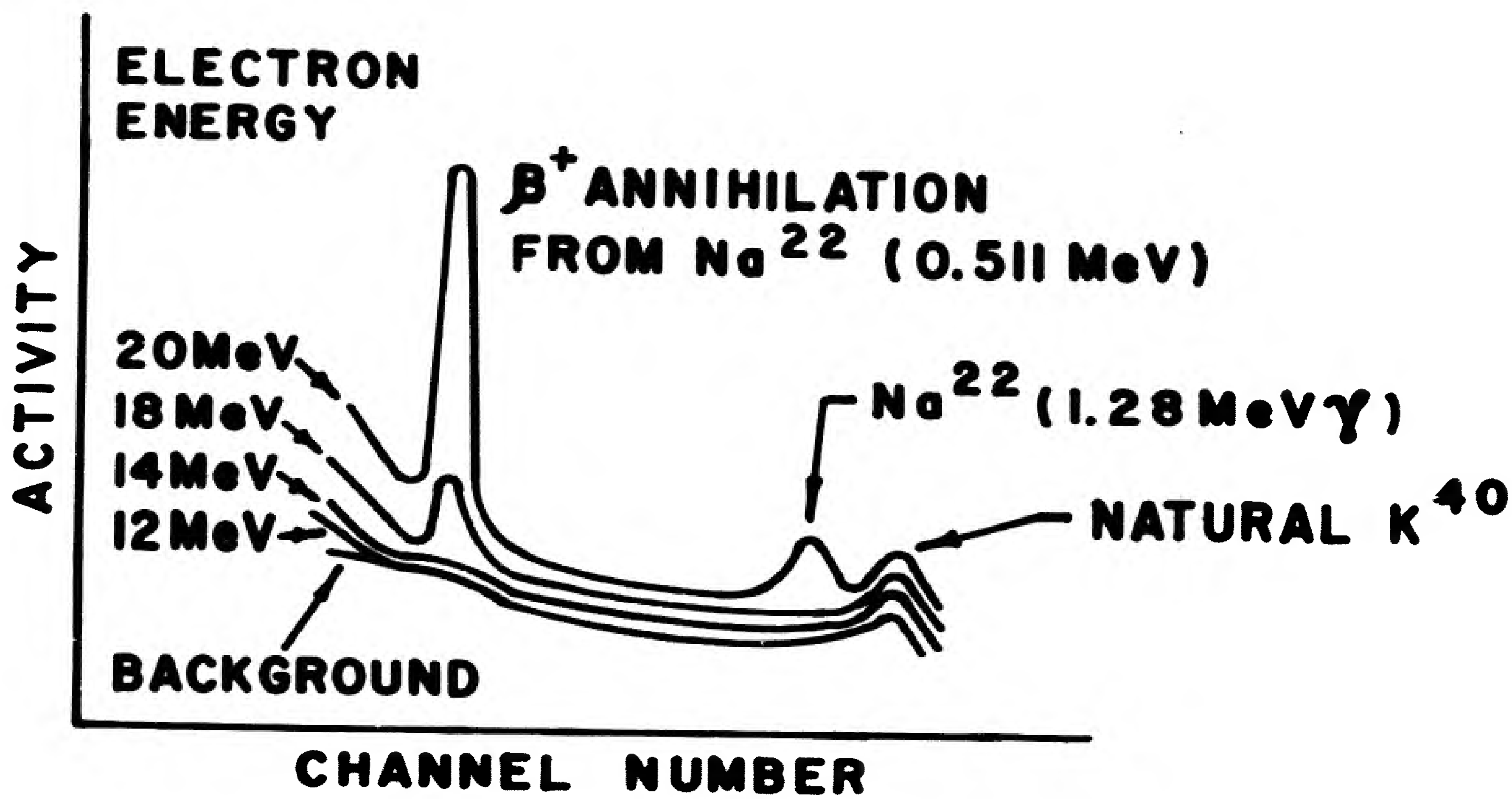

FIGURE 4

PHOTON SPECTRA FROM IRRADIATED FOODS

TABLE 6

SODIUM-22 ACTIVITY INDUCED IN FOODS

(Activity in pc/gm food/5 Mrads)

Food	Electron Energy (in MeV): 30	24	20	18	16	14	12
Beef	0.35	0.21	0.01	0.008	0.006	(0.003)[d]	None
Pork	a	0.16	0.026	0.015	b	b	None
Ham[c]	a	2.2	0.94	0.45	0.06	0.004	None
Chicken	a	0.15	0.03	b	b	b	None
Shrimp	a	0.16	0.026	b	(0.002)[d]	b	None
Beans	a	0.71	0.038	b	b	b	None
Composite Diet	a	a	0.03	0.018	0.008	(0.001)[d]	None

a No data availalbe.
b Below detectability, if present at all.
c Ham has approximately twelve times as much sodium content as beef.
d Limit of detectability is approximately 0.003 pc/gm food/5 Mrads.

Note: MPC for 2.6 year Na^{22} is 40 pc/cc.

P-32, Ca-45, relatively short-lived

All the beta decay activities induced in foods are relatively short-lived in comparison to the sodium activity produced. Fourteen-day P^{32} has been found to be the predominant beta activity for the first few months after irradiation(12,26,27). There is no detectable beta activity left one year after food irradiation. However, we can estimate an upper limit from Kruger's data that 160-day Ca^{45} is present at a level of 10^{-5} pc/gm food/5 Mrads. This was estimated by taking Kruger's data(26) for whole chicken irradiated with 24 MeV electrons to 5.6 Mrads and converting the activity at the end of irradiation for chicken meat irradiated to a 5 megarad dose.* These data are given in Table 7.

TABLE 7

ACTIVITY RESULTING FROM IRRADIATED WHOLE CHICKEN

Initial Activity	($t_{\frac{1}{2}}$)	pc/gm of Chicken/5 Mrads
Na^{24} ($+K^{42}$)	(15 hr.)	8.5 ± 1.7
P^{32}	(14.3 d)	0.14 ± 0.02[a]
(Ca^{45})	(160 d)	(0.02 ± 0.01)[a]
Na^{22}	(2.6 y)	0.20 ± 0.06

Source: Reference 26

a Obtained by radiochemical analysis of the chicken for calcium, followed by beta counting. This value is for homogenized whole chicken; hence it is higher than would be expected from chicken meat only, due to the high phosphorus and calcium content of the bones.

Na-24, short lived, therefore only a handling problem

As for handling the food following irradiation, the relatively short-lived radioactivities are of some interest. Food irradiated with 24 MeV electrons has been analyzed by several investigators(2,11,13,26,27). The predominant beta activity for periods up to one week after irradiation is Na^{24} (15 hour half life). This activity has been found to be produced to levels of 12 pc/gm beef/5 Mrads using 24 MeV electrons(1,26). The induced Na^{24} activity as a function of electron energy and food type is given in Table 8 and a plot of the Na^{24} activity induced in ham, beef, and pork is reproduced in Figure 5. As will be shown in a later section

* The ratio of calcium in Kruger's homogenized chicken to chicken meat was 1 to 4×10^{-4}

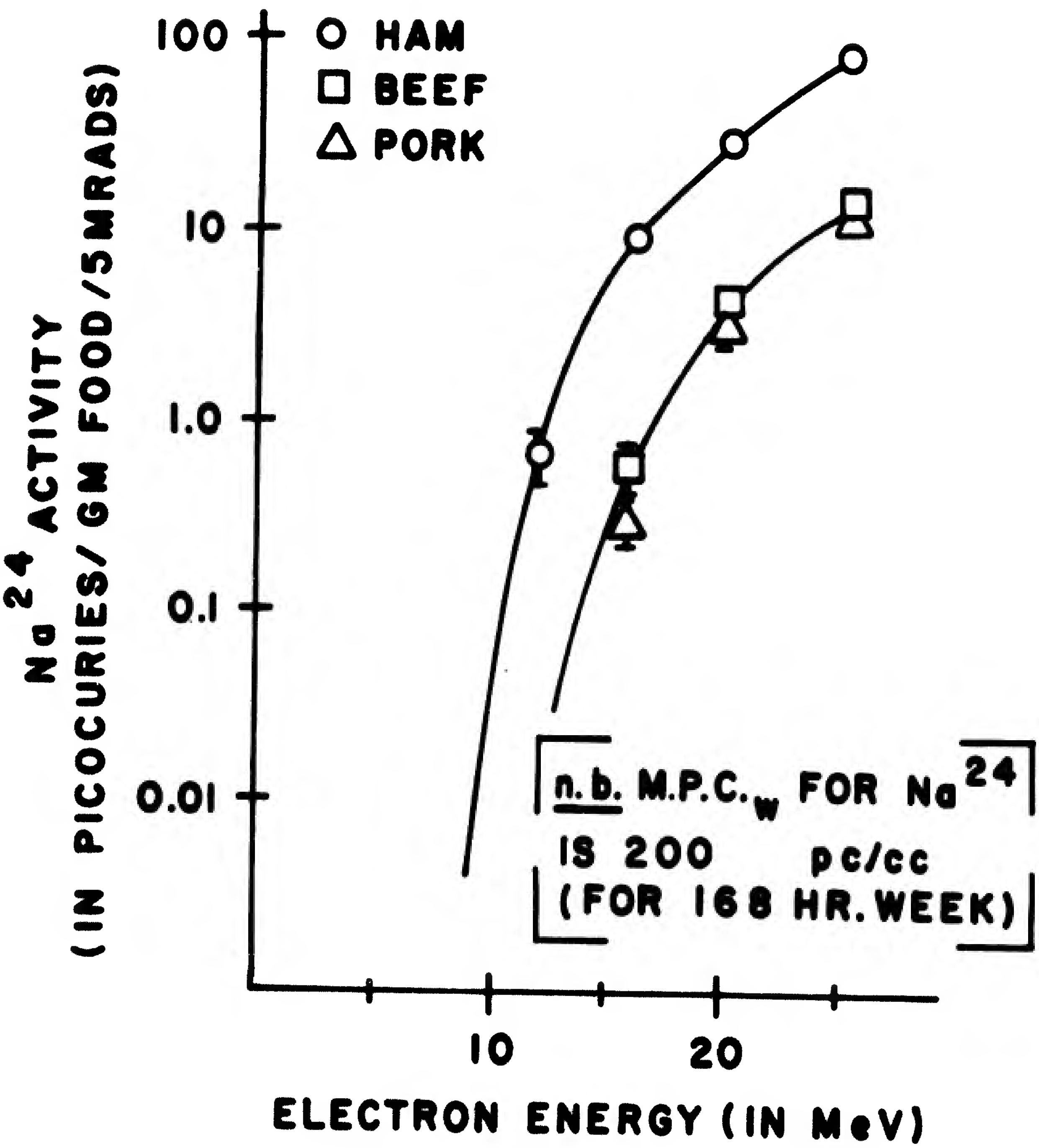

FIGURE 5

SODIUM-24 ACTIVITY INDUCED IN FOODS

at lower electron energies, the major mechanism for the production of beta unstable products is from the (n,γ) reaction and not a (γ,p) mechanism.

TABLE 8

LEVELS OF INDUCED SODIUM-24 ACTIVITY IN ELECTRON STERILIZED FOODS

(Activity in pc/gm food/5 Mrads)

Food	Electron Energy (MeV)				
	24	20	18	16	14
Beef	12.0 ± 1.3	4.2 ± 0.5	2.5 ± 0.3	0.57 ± 0.17	0.01
Pork	11 ± 1	3.9 ± 0.4	a	0.32 ± 0.06	0.01
Ham	85 ± 9	29 ± 3	20	8.9 ± 1.8	0.01
Chicken	8.2 ± 1.7	3.4	a	a	0.2
Shrimp	14	7.6	a	0.3	0.3
Beans	a	16	a	4.4	a

a Data not available.

Rb-87, random occurance of

The only other long-lived activity that has been detected without pre-irradiation chemical modification of the food is Rb^{87}. This, however, was found only in a specific sample of beef and at a level of 0.052 pc/gram beef/5 Mrads while other beef samples yielded no Rb^{87} activity after irradiation(3). We have been able to verify that such an activity is found in some samples of beef(28). This is just one indication of the wide range of trace elements present in the same type of food media. However, it has been shown that, on the average, the variation of a trace element in a given food is less than a factor of 10(3). The variation of a given element from food to food can be seen in Tables 22 to 25.

C-11, N-13, O-15, very short lived: handling problem

The very short-lived activities(for these purposes those with a half-life less than one hour) form a separate group as they are capable of producing a handling hazard immediately after irradiation rather than a consumption hazard. These activities which are C^{11}, N^{13}, and O^{15} have been measured using food irradiated with 24 MeV electrons; the results are presented in Table 9. It should be pointed out that 6 hours after irradiation, these activities are below detection level (i.e., less than 10^{-3} pc/gm beef/5 Mrads). A graphic example of this decay is reproduced in Figure 7.

TABLE 9

ACTIVITIES WITH VERY SHORT HALF LIVES

INDUCED IN FOODS WITH 24 MEV ELECTRONS

Isotope ($t_{\frac{1}{2}}$)		(pc/gm beef/5 Mrads x 10^6)
C^{11}	(20 m)	0.36
N^{13}	(10 m)	0.16
O^{15}	(2 m)	0.44

Results from Method 3: raising level of detectability

In order to gain more knowledge of the level of activity from trace elements other than sodium, some investigators have used a technique of chemical concentration. In this type of experiment, an element is added to the food prior to irradiation so that the element's concentration is increased by a factor which varies from 10^2 to 10^7. In this way, activity normally below detection level can be produced at detectable levels in samples of beef. The activities found and their calculated activity per gram of beef are given in Tables 11 and 12. An extension of this technique is to irradiate aqueous solutions of the food element; however, this tends to give values that are higher than those from irradiated foods(5). In regard to the effects of changing media on the activity induced in foods, it should be pointed out that the amount of activity induced does not change with a change in food media(4), if the trace element concentration remains constant.

Table of average activity in four foods

For comparison of all experimentally measured activities induced in foods, data were collected from the literature and converted to a standard form of picocuries per gram of food per 5 megarads. This included activities detected by all the techniques mentioned before. These values are average values for four food categories and are presented in Table 10. The activities listed are for isotopes with half lives greater than one month, as can be seen from Figure 6, while those predominant at short times after the end of irradiation are shown in Figure 7.

Before leaving this subject, one more series of papers should be mentioned. These are the Meneely reports on induced activities(13,35). The data presented there do not discriminate between activity in the food and activity in the can. However these papers have been very useful in showing that below 11.2 MeV there is no detectable radioactivity present in electron sterilized food.

TABLE 10

AVERAGE ACTIVITY IN FOOD AS DETERMINED BY EXPERIMENTAL METHODS*

Initial Activity (in pc/gm food/5 Mrads)

	Activity($t_{\frac{1}{2}}$)	Meat	Fish	Vegetables	Fruit
24 MeV Electrons	Na^{22} (2.6 y)	0.19	0.5	0.008	0.03
	P^{32} (14.3 d)	0.14	-	-	-
	P^{33} (25 d)	0.3	0.09	0.22	0.003
	S^{35} (87 d)	0.003	0.01	0.001	0.0002
	Ar^{37} (35 d)	0.09	0.08	0.07	0.08
	Ca^{45} (164 d)	2×10^{-5}	12×10^{-5}	6×10^{-5}	0.9×10^{-5}
	V^{48} (16 d)	-	-	-	-
	Cr^{51} (27 d)	3×10^{-5}	-	-	-
	Mn^{54} (291 d)	0.0004	0.0016	0.0017	0.0001
	Fe^{55} (2.7 y)	0.0014	0.011	0.004	0.0014
	Fe^{59} (45 d)	2×10^{-6}	-	-	-
	Zn^{65} (245 d)	0.06	0.002	0.01	0.003
	I^{126} (13.3d)	0.007	-	-	-
Natural	K^{40} (1.3×10^{9} y)	3.0	2.6	3.0	0.8
18 MeV Electrons	Na^{22}	0.012	0.01	0.01	-
	K^{40} & Zn^{65}	Same as data given for 24 MeV.			
	All others	Absent or below detection.			
12 MeV Electrons	Na^{22}		Not present.		
	All others		Same as 18 MeV.		
8 MeV Electrons	All		Same as 12 MeV.		

---

* All activities with levels less than 0.003 pc per gram of food are below detection limits by ordinary means (see text). Also note that those values are the average from all possible reference sources.

TABLE 11

CALCULATED ACTIVITY IN AN AVERAGE BEEF ITEM FROM STERILIZATION WITH 24 MEV ELECTRONS

Isotope ($t_{\frac{1}{2}}$)		Calculated Initial Activity* (pc/gm food/5 Mrads)
C^{11}	(20 m)	3.1×10^5
N^{13}	(10 m)	7.5×10^4
O^{15}	(2 m)	1.0×10^7
Na^{22}	(2.6 y)	1.3×10^{-1}
Na^{24}	(15 h)	2.5
P^{32}	(14 d)	1.1×10^{-1}
P^{33}	(25 d)	3.0×10^{-1}
Cl^{34m}	(32 m)	3.0×10^3
K^{43}	(22 h)	5.6×10^{-2}
Fe^{53}	(8 m)	1.2×10
Cu^{62}	(10 m)	3.0
Cu^{64}	(12.9 h)	6.0×10^{-2}
Cu^{67}	(5 m)	5.0×10^{-2}
Zn^{62}	(9 h)	1.4×10^{-1}
Zn^{63}	(38 m)	1.4×10^2
Zn^{65}	(245 d)	1.2×10^{-2}
Zn^{69m}	(14 h)	1.3
I^{126}	(13 d)	4.0×10^{-3}

* See note at bottom of Table 12.

TABLE 12

INDUCED ACTIVITY FOUND IN BEEF FROM STERILIZATION WITH 24 MEV ELECTRONS

Isotope ($t_{\frac{1}{2}}$)		Initial Activity (pc/gm food/5 Mrads)*
S^{35}	(87 d)	0.003
Mn^{54}	(291 d)	0.0004
Fe^{55}	(2.9 y)	0.011
Zn^{65}	(245 d)	0.004
I^{126}	(13.3 d)	0.007

* These values obtained by taking the activities from the chemically concentrated samples (pc/gm element/5 Mrads) and multiplying them by the appropriate elemental abundance in the food.

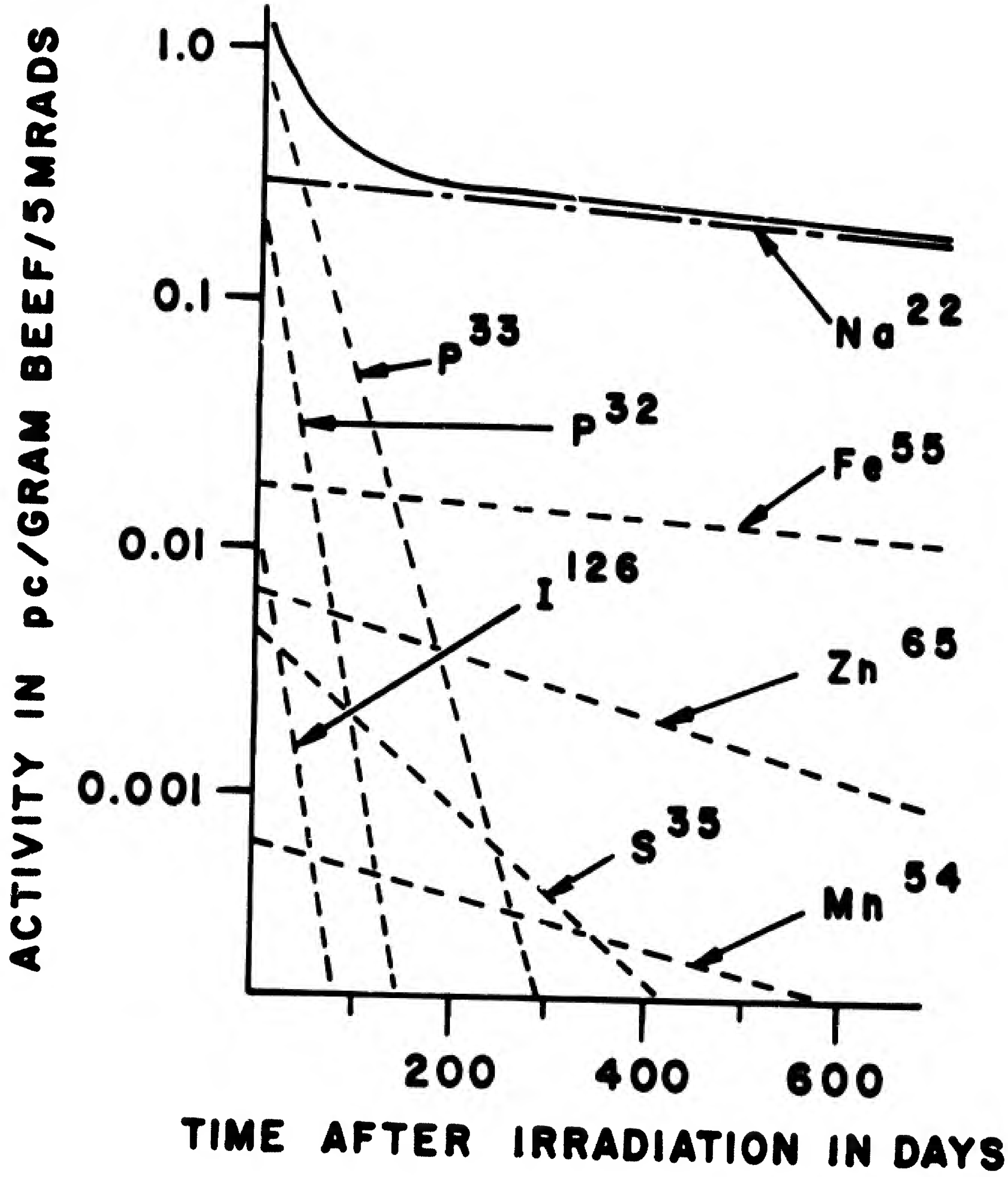

FIGURE 6

DECAY OF ACTIVITY INDUCED IN BEEF BY 24 MEV ELECTRONS

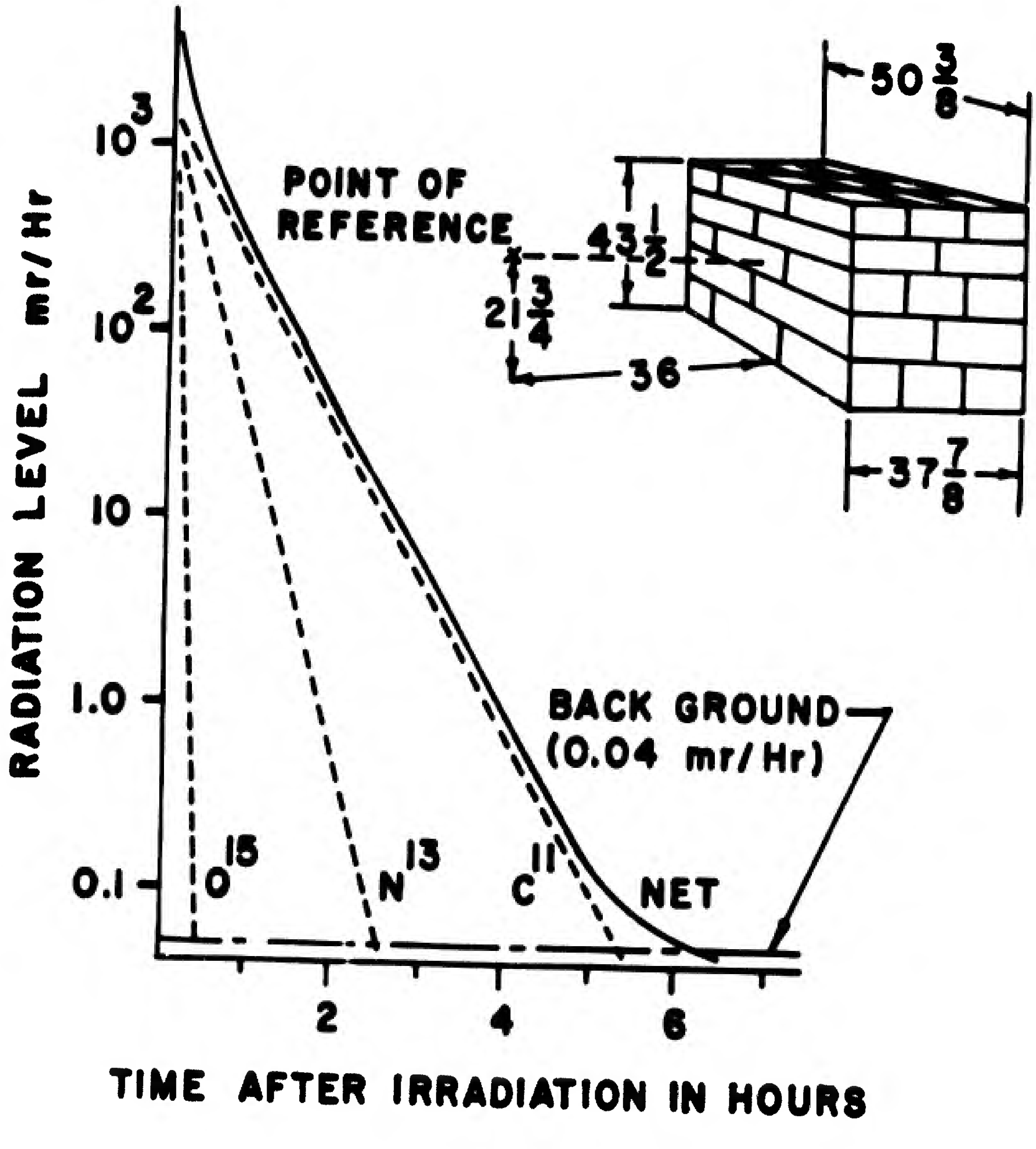

FIGURE 7

CALCULATED RADIATION LEVEL FROM A STANDARD PALLET

OF IRRADIATED NUMBER 10 CANS

Containers as a handling problem: activity & solution

Activities that are induced in the food packaging material can present a handling problem but not one of consumption. Immediately following the irradiation of food in tin-plate containers, the radiation level is high. However, six hours after the food is irradiated an entire standard pallet of number ten cans yields a lower radiation level than there is in the background. This has been shown in a calculation by Glass and Smith(5); their results are reproduced in Figure 7. The activity found in tin-plate cans after being irradiated to 5 Megarads with 24 MeV electrons is given in Table 13.

TABLE 13

AVERAGE ACTIVITY INDUCED IN TIN-PLATE CONTAINERS BY 24 MEV ELECTRONS[c]

Isotope ($t_{1/2}$)	Initial Activity[a] (pc/cm^2/5 Mrads)	t_n[b]
Long Lived Beta Activity ($t_{1/2} > 15$ d)	0.6 ± 0.2	---
Sn^{117m} (14 d)	1.2×10	190 days
Ni^{57} (36 hr)	9.9×10	24 days
Cu^{64} (12.8 hr)	2.7×10^2	7.5 days
Mn^{56} (2.6 hr)	5.6×10^3	3 days
Sn^{123g} (40 m)	6.1×10^2	9 hours
Fe^{53} (8 m)	1.4×10^5	3 hours

a Total area of a number ten can is 924 cm^2 (9, 31)
b t_n is the time required for the given activity to fall to a level below detectability (0.001 pc/gm).
Source: Taken in part from References 5 and 12.

The data of Meneely(13) should be discussed at this point as they provide information on the longer-lived activities in tin-plate containers. It should be pointed out that the report(13) mis-assigns a number of activities as well as includes a few errors in data reduction. As a consequence, the raw data presented in the report were taken, re-analyzed, and re-calculated. The corrigenda are given in Table 14. As can be seen, the activities Sn^{117m}, Ni^{57} agree with the activities found in Table 13. Also, one would expect to find Sn^{123m} and Mn^{54} on the basis of Table 13. Due to the discrepancies in the data on induced activities in tin-plate containers, there is a definite need for further work in this area.

TABLE 14

ACTIVITY FOUND IN TIN-PLATE CONTAINERS -- CORRIGENDA OF REFERENCE 13

Isotope	($t_{\frac{1}{2}}$)	Original Assignment	Initial Activity (pc/cm^2/5 Mrads)
Sn^{117m}	(14 d)	(Sc^{47})	11.2
Sn^{123m}	(125 d)	(Sc^{46})	2.1
Ni^{57}	(36 h)	(Sc^{48})	28
Mn^{54}	(278 d)	(Mn^{54})	17.0
β^+ Component	(80 ± 30 d)	(Te^{121})	27.3
260 ± KeV Photon Component	(50 ± 10 h)	(Xe^{133m})	200 ± 50

There are two other classes of containers -- aluminum cans and plastic packaging. Both of these categories have been investigated for activities induced in them (28,5). The activity induced in the former is strongly dependent upon the type used. For example, type 2S (high purity) aluminum has almost no activity produced in it, while less pure types may have up to a thousand times as much activity present. The data obtained by various investigators are presented in Table 15.

TABLE 15

ACTIVITY INDUCED IN ALUMINUM BY 24 MEV ELECTRONS

(Initial Activity in pc/cm^2/5 Mrads)

Isotope ($t_{\frac{1}{2}}$)	Type Aluminum 2S	24S	75S
Long Lived ($t_{\frac{1}{2}} > 1$ d)	None	None	None
Cu^{64} (12.9 h)	160	5,700	1,500
Cu^{62} (10 m)	8,000	5.3×10^5	7.0×10^4

Plastics present a different case since they may have more than one major component, such as saran which has carbon, hydrogen, and chlorine present. However, the initial activity present is C^{11} from the C^{12} (γ,n) C^{11} reaction. The average value for C^{11} activity is 53,000 pc/cm^2 for plastic material irradiated to five megarads by 24 MeV electrons. Values for various types of plastics are given in Table 16 and are taken from Glass and Smith(5). Long-lived activities were not found in any of the plastics tested.

TABLE 16

ACTIVITIES PRODUCED IN PLASTIC CONTAINERS

IRRADIATED WITH 24 MEV ELECTRONS

(Activity in pc/cm^2 x 10^6)

Plastic	C^{11} (20 m)	Cl^{34m} (32 m)	F^{18} (1.7 h)
PVC	0.057	0.088	None
Saran	0.059	0.054	None
Fluorothane	0.013	0.079	0.032
Others*	0.053	None	None

* These include Mylar, Polyethylene, Nylon, and Polystyrene.

4. Neutron fluxes in foods

Results with simulated food

Since it appears that residual radioactivity can be induced at lower energy by neutron activation than by direct photonuclear reaction (see text later), attention must be paid to the amount of neutron flux that is present during food sterilization. Neutron fluxes have been measured only with simulated food such as press-board or wet wood. Kruger and Wilson(26) have measured the neutron flux present at electron energies of two to eight MeV as well as at 30 MeV. The values they obtained, presented in Table 17, were arrived at by use of various foils imbedded within a block of wet wood the size of a number ten can. They also present data on neutron fluxes present at various gamma irradiation facilities.

TABLE 17

INTEGRATED NEUTRON FLUX PRESENT DURING
ELECTRON IRRADIATION OF SIMULATED FOODS
(flux in n/cm^2/5 Mrads)*

Neutron Energy(MeV)	Electron Energy (in MeV)				
	2	3.5	6	8	30
Thermal	5.5	240	1.1×10^3	1.2×10^3	1.8×10^7
2.5	-	-	3.0×10^5	-	1.9×10^9
4.6	-	-	6.0×10^5	-	1.7×10^{10}
8.1	-	-	1.2×10^6	-	2.6×10^8
11.5	-	-	1.2×10^6	-	3.1×10^8

* The error in these figures is ± 30%

Effect of beam stopper behind irradiated food

The only other work performed on the measurement of neutron fluxes at food irradiation facilities was by Haddad et al.(27). Their experimental arrangement was such that they measured the effect of a beam stopper directly behind the irradiated food. This is useful is pointing out the need for proper beam stopping devices. The data they obtained are given in Table 18.

TABLE 18

NEUTRON FLUXES ATTENDANT WITH ALUMINUM AND LEAD BEAM STOPPERS

(flux in $n/cm^2/5$ Mrads)[a]

Electron Energy (MeV)	Al Beam Stopper[b]		Pb Beam Stopper	
	Thermal	Fast	Thermal	Fast
13	2.1×10^7	2.5×10^7	5.4×10^9	4.3×10^8
11	1.1×10^7	6.1×10^6	2.0×10^9	6.2×10^7
9	3.6×10^6	3.6×10^6	3.4×10^8	3.6×10^6

[a] All figures quoted have a ten percent accuracy.
[b] Slow neutron shielding was also used.

Two estimates of neutron flux

Although no work has been done on the neutron flux within food during irradiation, we can arrive at an upper estimate of the integrated thermal neutron flux present by use of the activities present in food arising from the (n,γ) reaction. According to Glass and Smith(5), the ^{24}Na activity is due almost entirely to the ^{23}Na (n,γ) ^{24}Na reaction.* We know that the ratio of the slow neutron cross section to the fast neutron cross section for this process is large in comparison to the presently known ratio of slow to fast neutrons in food irradiation facilities(26). Using the data for ham given in Tables 8 and 24, we obtain Table 19.

TABLE 19

ESTIMATED NEUTRON FLUX

(flux in $n/cm^2/5$ Mrads)

	Electron Energy (MeV)				
	24	20	18	16	14
Integrated Thermal Neutron Flux:	24×10^8	8.2×10^8	5.7×10^8	2.5×10^8	3×10^5

(If these were calculated as if the entire activity arose from fast neutrons, the values would be of the order of 10^{14} $n/cm^2/5$ Mrads)

* This is true for energies below 20 MeV, but may not be true for energies greater than 20 MeV (see later text).

Another source that can be used to estimate the integrated neutron flux is the data of Peterson, et al.(30). Their data on activity produced in various foods after being subjected to thermal neutrons are given in Table 20 in modified form:

TABLE 20

INTEGRATED NEUTRON FLUX IN SEVERAL FOODS

Food	Integrated Flux (n/cm^2/gm food)
Ham	2.02×10^5
Beans (Baked)	6.94×10^5
Bread	1.15×10^5
Butter	1.13×10^5
Fish (Tuna)	5.71×10^5
Egg	5.78×10^5
Spinach	5.38×10^5

Using their data in conjunction with the data given in Table 8, Table 21 is obtained:

TABLE 21

CALCULATED NEUTRON FLUX IN FOOD

(n/cm^2/5 Mrads)

at Electron Energy (MeV):

24	20	18	16	14
22.3×10^9	7.5×10^9	5.2×10^9	2.3×10^9	3×10^6

Upon comparison of Tables 19 and 21, it appears that the predicted integrated neutron flux should be between 10^8 and 10^{10} n/cm^2/5 Mrads, if all the Na^{24} were due to the ^{23}Na (n,γ) ^{24}Na reaction. However, these values are high in comparison to the data of Kruger(26). Furthermore, these values must be taken with reservation, particularly at higher energies ($E > 20$ MeV), since it appears that the ^{25}Mg (γ,p) ^{24}Na reaction may contribute significantly to the ^{24}Na activity produced during food irradiation. The amount of ^{24}Na activity that might be expected from such a reaction can be estimated from the Na^{22} activity produced. This can be done if we assume the cross sections of the two processes, ^{25}Mg (γ,p) ^{24}Na and ^{23}Na (γ,n) ^{22}Na have the same relative shape; as can be seen in Figure 1, they do except for a translation in the energy axis of 3.4 MeV. Also, the approximation that the bremsstrahlung distribution is approximately the same for electrons of initial energy E and E + 3.4 MeV must be made. With the use of these assumptions and Tables 8 and 23 to 25, the calculated values for Na^{24} activity were obtained and are presented together with the measured values in Table 22:

TABLE 22

CALCULATED Mg^{25} (γ,p) Na^{24} ACTIVITY AND MEASURED Na^{24} ACTIVITY IN FOOD

(Activity in pc/gm food/5 Mrads)

		Electron Energy (MeV):									
Na^{24} Activity		33.4	27.4	24	23.4	21.4	20	19.4	18	17.4	16
Beef	Calculated*	32	19	-	9.2	0.7	-	0.5	-	0.3	-
Beef	Measured	-	-	12	-	-	4.2	-	2.5	-	0.57
Ham	Calculated*	-	212	-	90	43	-	2.5	-	0.4	-
Ham	Measured	-	-	85	-	-	29	-	20	-	8.9

* Calculated from A_1 (^{24}Na at E + 3.4) = BA_2 (Na^{22} at E) $n_1T_2/n_2 T_1$, B is the ratio of cross section height, n is the isotopic abundance, and T is the half life.

Summary: no data on neutron flux within irradiated food

From the foregoing we can only conclude that the induced activity is produced by neutron reactions at the lower energies, while at the higher irradiation energies photonuclear reactions predominate in contributing to, at least, the Na^{24} activity. As a consequence of this, the neutron fluxes cannot be determined from the Na^{24} activity found in food; hence, other means must be employed. Due to the lack of data on neutron fluxes within electron irradiated foods, experiments are in progress in this Laboratory to measure the neutron flux as a function of electron energy, food media, and irradiation conditions.

5. Elemental composition of foods

If we desire an a priori knowledge of what radioactivity is going to be produced in the foods upon high energy electron sterilization, then one of the much needed pieces of information is the fractional abundance of elements, particularly those present in trace quantities. Some of this information has been made available as a by-product of one of the programs to measure neutron fluxes in electron irradiated foods via the activation analysis of various meats for trace element composition(32,51). The data obtained for beef, ham, pork, and chicken are given in Table 23. For sake of completeness, data were compiled on the variation of sodium, phosphorus, potassium, and iodine in a number of foods as well as the abundance of the more common elements in several foods. These values are given in Tables 24 and 25.

TABLE 23

TRACE ELEMENT ABUNDANCE IN FOUR MEATS[c]

(n.b. A x 10^B is quoted as A(B))

Food Element	Beef	Pork	Ham	Chicken
Sc	5.5(-9)	2.6(-9)	2.4(-9)	7.3(-9)
Ir	7(-12)	5(-12)	2.0(-11)	6(-12)
Se	2.7(-10)	7.8(-11)	2.6(-10)	4.1(-11)
Rb	1.3(-6)	1.3(-6)	1.5(-5)	2.4(-5)
Sr	1.8(-7)	9.2(-8)	3.8(-7)	2.2(-7)
Zr	2.0(-7)	1(-7)	1(-7)	1(-7)
Mo	1.5(-8)	--	--	--
In	9.2(-9)	2.2(-9)	1.1(-8)	4.7(-9)
Ag	1.4(-8)	--	--	--
Tm	7(-12)	5(-12)	2(-11)	6(-12)
Sn	2(-9)	--	--	--
Te	5(-9)	3(-9)	2(-9)	6(-9)
Cs	9.2(-9)	6(-9)	6.6(-9)	1.1(-8)
Ba	3.5(-6)	2.1(-7)	3.8(-7)	3.3(-7)
Ce	3.8(-10)	1.8(-11)	1.5(-11)	1.1(-11)
Hf	2.0(-8)	1.9(-8)	1.2(-8)	1.9(-8)
Pb	1.0(-8)	--	--	--
U^{235}	2.7(-11)	1.5(-11)	3.0(-11)	3.6(-11)
U^{238}	3.5(-9)	3.4(-9)	3.8(-9)	6.8(-9)
Rare Earths[a]	7(-11)	5(-11)	2(-10)	6(-11)
Pt Metals[b]	7(-10)	5(-10)	2(-9)	6(-10)

a The same value applies for Yb, Lu, and Eu.
b The same value applies for Ru, Pb, and Pt.
c This is a compendium of data in References 5, 16 to 26, 30, 50, and 51.

TABLE 24

AVERAGE ABUNDANCE OF SELECTED ISOTOPES IN VARIOUS FOODS

(Abundance in mg/100 gm food)

	Na^{23}	P^{31}	K^{40}	I^{127}
Beef	49.5	194	336	0.009
Ham	736	149	254	--
Bacon	750	100	210	0.016
Pork	70	221	316	0.008
Chicken	81	202	337	--
Eggs	116	187	126	--
Cod Fish	73	191	350	0.031
Tuna Fish	800	294	240	0.031
Halibut	84	211	304	0.050
Shrimp	140	318	424	0.036
Oysters	471	204	143	0.10
Wheat Flour	a	93	370	--
Cereal	25	330	340	--
Bread	465	25	110	--
White Potatoes	16	50	506	0.004
Sweet Potatoes	21	47	452	0.002
Carrots	58	32	320	0.004
Beans, Green	b	40	229	0.007
Beans, Baked	490	88	255	--
Peas	300	41	57	--
Cabbage	18	24	260	0.002
Spinach	513	14	163	--
Apples	2	12	125	--
Oranges	5.8	26	182	0.001
Peaches	15	15	256	0.01
Milk, Powder	275	950	1,116	0.032
Milk, Whole	50	92	145	0.004
Butter, (Salted)	626	10	22	--
Chocolate	9	27	165	--
Cheese	490	66	60	--
Boullion	23,000	600	1,400	--
Beer	5	17	26	--

Source: Taken in part from 14, 16-26 (References).

a = Range is 0.23 to 23
b = Range is 3 to 160.

TABLE 25

AVERAGE ELEMENTAL ABUNDANCE IN FOODS
(n.b. A x 10^B is given as A(B))

Element	Fractional Abundance			
	Beef	Pork	Chicken	Fish(Cod)
Na	5 (-4)	7.0(-4)	8.1(-4)	7.3(-4)
Mg	2.7(-4)	2.5(-4)	3.7(-4)	2.2(-4)
Al	1 (-6)	--	--	--
Si	1 (-6)	--	--	--
P	1.8(-3)	2.2(-3)	1.9(-3)	5.0(-3)
S	2.2(-3)	2.1(-3)	2.7(-3)	2.0(-3)
Cl	5.6(-4)	5.5(-4)	5.6(-4)	1.5(-3)
K	3.9(-3)	3.2(-3)	3.4(-3)	3.5(-3)
Ca	2.0(-4)	1.2(-4)	1.5(-4)	1.2(-4)
Cr	3.0(-8)	--	--	--
Mn	2.0(-7)	1.2(-6)	5.0(-7)	3.0(-7)
Fe	3.7(-5)	3.1(-5)	1.8(-5)	8.5(-6)
Co	2.0(-9)	--	--	--
Ni	3.0(-9)	--	--	--
Cu	5.0(-7)	5.0(-6)	5.4(-6)	2.6(-6)
Zn	1.5(-5)	1.4(-5)	5.0(-6)	5.0(-6)
I	9.0(-8)	8.0(-8)	--	3.1(-7)

Element	Potatoes	Green Beans	Carrots	Oranges	Milk
N	3.4(-3)	1.9(-3)	1.1(-3)	1.3(-2)	5.3(-3)
Na	1.6(-4)	1.0(-3)	5.8(-4)	5.8(-5)	5.0(-3)
Mg	2.6(-4)	2.7(-4)	1.9(-4)	1.1(-4)	1.5(-4)
Al	--	1.0(-7)	5.0(-7)	--	--
P	5.0(-3)	4.0(-4)	3.2(-4)	2.6(-4)	9.2(-4)
S	3.3(-4)	2.3(-4)	1.6(-4)	9.0(-5)	3.1(-4)
Cl	5.4(-4)	3.0(-4)	4.8(-4)	4.0(-5)	1.1(-3)
K	5.1(-3)	2.3(-3)	3.2(-3)	1.8(-4)	1.5(-3)
Ca	1.2(-4)	5.4(-4)	4.2(-4)	3.4(-4)	1.2(-3)
Mn	2.1(-6)	4.6(-6)	4.0(-6)	3.0(-7)	3.0(-7)
Fe	1.1(-5)	9.0(-6)	6.4(-6)	4.3(-6)	1.6(-6)
Co	6.0(-8)	3.5(-7)	2.0(-4)	--	1.0(-9)
Ni	2.5(-7)	--	--	--	4.0(-9)
Cu	1.6(-6)	1.2(-6)	1.1(-6)	1.1(-6)	2.6(-7)
Zn	4.0(-6)	1.0(-6)	5.0(-6)	1.3(-6)	3.2(-6)
I	4.0(-8)	7.0(-8)	4.0(-8)	1.0(-8)	4.0(-8)

Taken in part from References 5, 16 to 26, and 30.

6. Tritium activity

During the early phases of the study of radioactivity induced in foods by high energy electrons, it was postulated that tritium might be produced at high levels in irradiated foods. However, this has been disproved by several investigators who could not detect any increase in the tritium activity level over the naturally occuring levels even from irradiation with 24 MeV electrons(1,5,27,35).

Haddad et al. found that the tritium content of food is less after irradiation than the level of natural tritium content before irradiation. This phenomenon, however, probably can be explained by the migration of natural tritium out of the food during irradiation. This is quite plausible due to the large migration effect of tritium ions(36) and the temperature rise found during electron irradiation of food(6).

There are two reaction types that could lead to tritium production: namely, the $(\gamma, {}^3H)$ and the $(n, {}^3H)$ reaction. That the latter does not contribute to tritium production was shown by Haddad et al. who varied the thermal neutron flux by a factor of 200 and the fast neutron flux by a factor of 2 through the use of aluminum and lead beam stoppers. The former reaction type, the $(\gamma, {}^3H)$ was investigated by Glass and Smith(5) who attempted to set an upper limit on the tritium activity that would be produced in foods by irradiating aluminum and copper foils with 24 MeV electrons. Their results, in modified form, are given in Table 26.

TABLE 26

TRITIUM ACTIVITY PRODUCED BY 24 MEV ELECTRONS

Target	Activity (pc/gm element/5 Mrads)	Activity (pc/gm beef/5 Mrads)*
Aluminum	6.9×10^{-5}	6.9×10^{-11}
Copper	1.4×10^{-4}	7.0×10^{-9}

(Arrived at by use of values in Table 25)

* The average temperature rise in food irradiated with 24 MeV electrons is 2.2°C/Mrad(6,43).

7. Isomer activation

No radioactive isomers in food

Isomer activation is a process which could produce radioactivity in foods at energies as low as one MeV. At the lower irradiation energies, below particle emission threshold, a long-lived metastable state must be produced from an isotope with a stable ground state. In general, the elements whose isotopes are capable of being excited to metastable states are not present in food or, if present, are found in concentration of a few parts per billion, or less. Consequently, it is to be expected that these should not produce radioactivity at detectable levels in foods. Glass and Smith could find no radioactive isomers present in foods irradiated to five megarads with 4, 8, 16, and 24 MeV X-rays*, as well as with three gamma sources (Cs^{137}, Co-60, and fuel element)(34).

Since it is of interest to know to what extent isomers are produced by five megarad doses, two sets of investigators have irradiated elements (in their elemental and chemical form) known to yield isomeric states in order to determine the upper limits of the amount of activity that could be produced (12,34).

Equation to predict isomer activity produced from irradiation with electrons

It is desirable to have an equation that will predict the activity produced from irradiation with electrons of energy E. Such an equation will take the form:

$$\text{Activity}^{**} = C\ f(E)\ f(a,i)\ t^{-1} \qquad (1)$$

where:

C = a constant
f(E) = a function dependent upon the energy of irradiation
f(a,i) = elemental and isotopic abundance of parent isotope
t = half life of isomer

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* Among the foods irradiated were: Beef, Bacon, Shrimp, Chicken, and Green Beans.

** This assumes a short irradiation time (viz., $\frac{\text{irradiation time}}{\text{isomer half life}} \ll 1$)

By using data given in the literature(34), we can determine f(E) and C:

$$R = K_2\ f_1 E^5 t^{-1} \text{ (in } \mu\text{c/gm } \underline{\text{element}}\text{/5 Mrads)} \qquad (2)$$

or

$$R = K_3\ f_e E^5 t^{-1} \text{ (in pc/gm } \underline{\text{food}}\text{/5 Mrads)} \qquad (3)$$

where:

R = Activity from the reaction $A(\gamma,\gamma')\ A^m$ for the energy range 3 to 30 MeV.
$K_2 = (8 \pm 1.4) \times 10^{+1}$ and $K_3 = (8 \pm 1.4) \times 10^{-5}$
f_1 = Fractional abundance of parent isotope in the natural <u>element.</u>
f_e = Fractional abundance of element in the given <u>food</u> under consideration.
E = Energy of the electrons(MeV).
t = Half life of product isomer (hours).

Equations 2 and 3 are not of the functional form that might be expected for such a process; however, they do yield a fairly good estimate of the activity produced, as can be verified by comparison of tables 27 and 28 or by inspection of Figure 8 where several calculated and experimental values are plotted. It should be noted that Glass and Smith present a relationship for estimating the activity produced from isomer activation but only at specific energies(34).

From an extensive survey on nuclear isomers, Glass was able to conclude that "no significant undiscovered isomers exist"(34). Consequently, with this and the foregoing experimental work, it is concluded that isomer production is not a problem in the electron irradiation of food.

TABLE 27

EXPERIMENTALLY DETERMINED RADIOACTIVITY PRODUCED FROM ISOMER ACTIVATION

(Activity in pc/gm beef/5 Mrads)

Isomer	($t_{\frac{1}{2}}$)	X-Ray Energy (MeV): 4	8	16	24
Sr^{87m}	(2.8h)	3×10^{-5}	5×10^{-4}	0.8	1
Nb^{93m}	(12 y)	--	--	--	--
In^{115m}	(4.5h)	5×10^{-5}	9×10^{-3}	0.08	0.14
Te^{123m}	(104d)	--	--	8.8×10^{-10}	--
Te^{125m}	(58 d)	--	--	--	2×10^{-8}
Ba^{135m}	(28.7h)	2×10^{-6}	2×10^{-4}	7×10^{-3}	0.02
Pb^{204m}	(67 m)	2×10^{-7}	--	0.03	0.03
Hf^{160m}	(5.5h)	5×10^{-7}	6×10^{-8}	--	--
Pt^{195m}	(3.5d)	--	--	--	--
Hg^{199m}	(42 m)	1.8×10^{-9}	3×10^{-2}	--	--

Electron Energy (in MeV):	3	8	30
In^{115m} (pc/gm element/5Mrads)	1.1×10^{4}	1.0×10^{6}	7.4×10^{8}

TABLE 28

CALCULATED ACTIVITY AT END OF IRRADIATION FROM ISOMER ACTIVATION

(Activity in beef in pc/gm/Beef/5 Mrads)

Isotope	($t_{\frac{1}{2}}$, f_i)	Electron Energy (MeV): 6	12	24
Nb^{93m}	(12 y, 1.0)	5.9×10^{-10}	1.9×10^{-8}	6.1×10^{-7}
Cd^{113}	(5.1y, 0.12)	1.6×10^{-10}	5×10^{-9}	1.7×10^{-7}
Te^{123m}	(104d, 0.009)	1.1×10^{-8}	3.7×10^{-7}	1.1×10^{-5}
Te^{125m}	(58 d, 0.07)	1.6×10^{-7}	4.9×10^{-6}	1.6×10^{-4}
Pt^{195m}	(3.5d, 0.34)	1×10^{-6}	3.2×10^{-5}	1×10^{-3}
Ba^{135m}	(28.7h, 0.07)	5×10^{-4}	1.6×10^{-2}	0.5
Hf^{180m}	(5.5h, 0.35)	8×10^{-4}	2.5×10^{-2}	0.8
In^{115m}	(4.5 h, 0.96)	1.2×10^{-3}	3.9×10^{-2}	1.3
Sr^{87m}	(170m, 0.07)	3×10^{-3}	9×10^{-2}	2.9
Pb^{204m}	(67m, 0.015)	8.4×10^{-3}	0.27	8.6
Cd^{111m}	(49m, 0.13)	10^{-5}	3×10^{-4}	10^{-2}
Hg^{199m}	(42m, 0.17)	1.5×10^{-5}	4.8×10^{-4}	1.5×10^{-7}

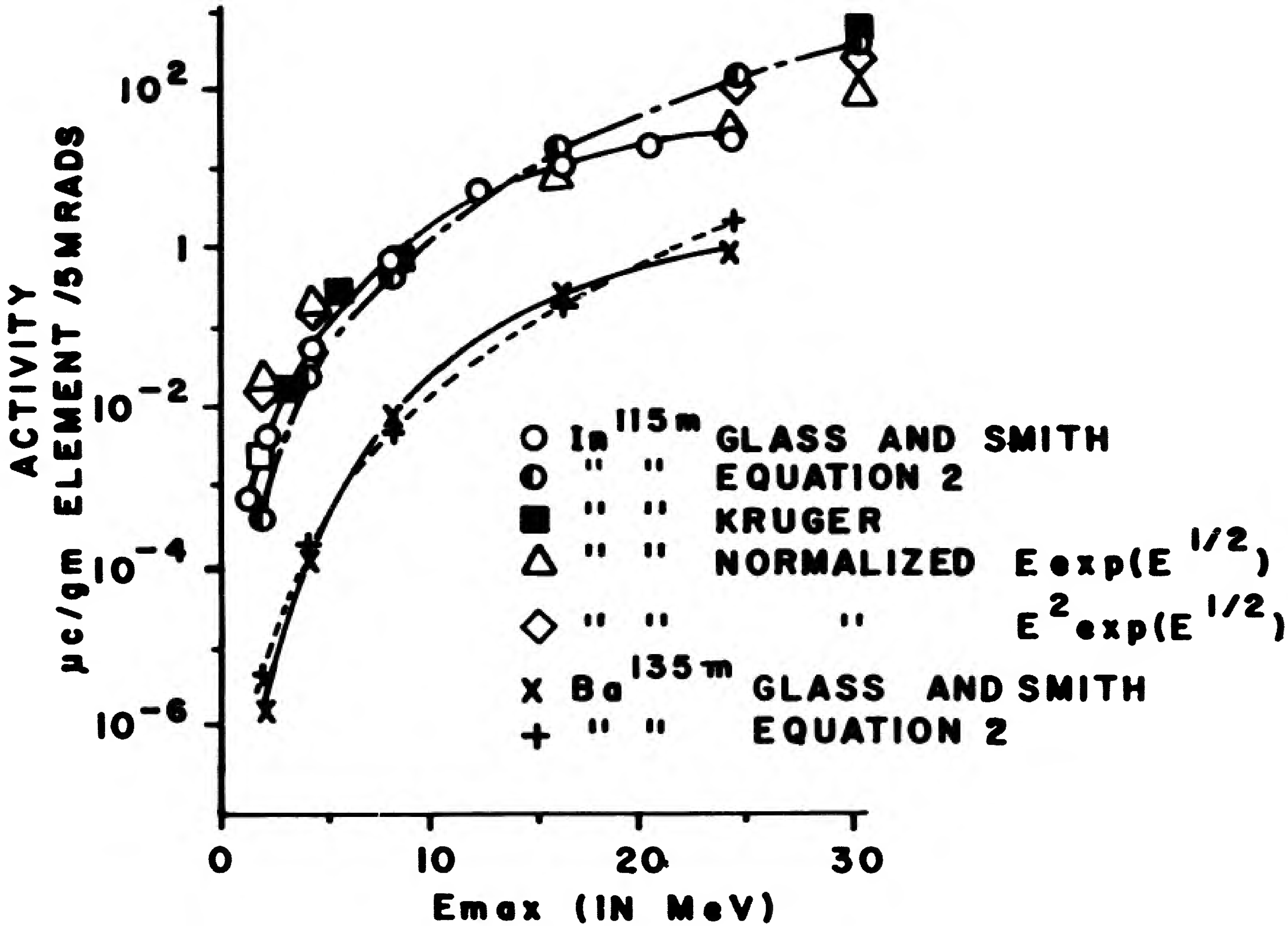

FIGURE 8. RADIOACTIVITY ARISING FROM ISOMER ACTIVATION

8. Equations for predicting average amount of induced activity

The problem of reducing "yield term" to analytical form

The activity produced in foods by electron irradiation could easily be calculated accurately if the expression for its production was in simple analytical form. Unfortunately, this is not the case. The production of activity can be put in the general form:

$$R_s = k\,G \sum_j \int_0^{E_0} N_j\,\sigma_j\;dE \qquad (4)$$

where:

R_s = activity at saturation produced in a given media subjected to electrons of energy E_0

k = constant (to give R_s in appropriate units)

G = a term dependent on the media

σ_j = cross section for the process (j,λ) producing the activity

Nj = the energy distribution (and flux) of j

The last term $\int_0^{E_0} N\sigma de$, the integrated cross section or yield, gives rise to the complication in calculation as well as inhibits a solution in simple form. Of the several equations that have been reported for calculating activities, the main difference in their development is the method of reducing the yield term to analytical form. For electron irradiations there will be two or more terms in the sum, one for the (e, e′n) process and one for the (γ,n) process as well as, in certain cases, terms for the (γ,p) and (n,γ) processes. More complicated processes will not compete in the energy ranges used in food sterilization. In general the (e, e′n) probability of occurrence is so much smaller than the (γ,n) process that it can be considered negligible, leaving only one term for the yield. For activities produced from the (γ,p) process, the same procedure is used as in the (γ,n) process. However, for neutron-induced activities a complicating factor is that the neutron energy distribution (and flux) arises from (γ,n) reactions in the food and surroundings, hence does not lend itself to purely theoretical calculation, but instead must be calculated by empirical means. One more simplification can be made by realizing that the time involved in electron sterilization will be very short with respect to any half lives of interest. Consequently, we can reduce the activity at saturation to that produced during a given dose by:

$$R = 0.693\ R_s\ t_e\ T^{-1} \qquad (5)$$

where:

R = the activity produced from an irradiation of n Megarads

t_e = time required to obtain an n Megarad dose

T = half life of activity under consideration

With these restrictions, the activity produced takes the general form:

$$R \propto D\ I\ T^{-1} \qquad (6)$$

where:

D = dose given to the food

T = half life of product activity

I = yield or integrated cross section (viz. $I = \int_{e}^{E} N\sigma dE$)

<u>Hershman's equation</u>

The simplest relationship is given by Hershman(38) who performs a cursory integration of the yield term to arrive at the relationship:

$$R = 30\ D\ E\ n\ T^{-1} \qquad (7)$$

where:

R = Activity (in pc)
D = Dose (in Mrads)
E = Initial electron energy
n = amount of material per gram of food
T = half life in years

An equation such as Hershman's should be used with caution, for it can easily exaggerate the amount of activity produced due to the energy dependent term, as will be shown later. Actually this relationship should be limited to the range $E_m - \Gamma$ to $E_m + \Gamma$ where E_m is the energy at which the maximum in the cross section occurs and Γ is the half width at half maximum of the nuclear reaction cross section curve. In general this would be from 15 to 21 MeV, or in the range normally encountered in food irradiation. Further, Glass and Smith(1) suggest that k be set at about 7.5 rather than 30. However, even this limitation appears not to be sufficient, as will be shown later.

Skaggs' equation

Another relationship is that of Skaggs(11) who presents a more detailed development of the bremsstrahlung production and electron energy loss within the sample, as well as the amount of activity produced at a given depth within the irradiated sample. To obtain an expression for the integrated cross section, Skaggs used Jones and Terwilliger's(40) empirical relationship for the integrated cross section to 27.5 MeV:

$$\int_0^{27.5} \sigma_n (E)\, dE = 5.2 \times 10^{-4} A^{1.8} \text{ MeV - barns} \qquad (8)$$

This yields the relationship for the activity set at saturation to be:

$$R_s \approx k\, n\, A\, F_e\, Y(L) \qquad (9)$$

where:

- R_s = activity at saturation in pc/gm food
- K = 2.0×10^{-4} constant
- n = fractional amount of parent isotope in food
- A = mass number of target isotope
- F_e = electron flux through the sample volume in electrons/cm^2 sec.
- L = depth of section below target surface in cm.

Y(L):

If $0 < L < 5$ cm
$Y(L) = 1.013 - \exp(-0.02L)$

If $L > 5$ cm
$Y(L) = 0.105 \exp(-0.02L)$

However, for food irradiation to megarad doses the irradiation time is short and, in general, the target thickness in cm is roughly one third E, where E is given in MeV. For a volume element in the middle of a food sample irradiated with 30 MeV electrons, Skaggs' equation can be reduced to the form:

$$R \approx k\, A\, n\, D\, T^{-1} \qquad (10)$$

where:

- R = activity in pc/gm food/D Mrads
- k = 15.4
- n = fractional amount of parent isotope in food
- A = atomic number of parent isotope
- D = dose in megarads
- T = half life of activity in years

Newkirk's technique (computer)

A totally different approach was used by Newkirk et al.(4) in their computer calculation. They calculated the radioactivity as a function of the depth of penetration an electron beam made into a sample of a given media. To calculate the yield term, they employed electron energy loss graphs, theoretical relationships for production and energy loss of X-rays, given by Heitler(39) and photonuclear cross sections. The sub-programs generated the production, gain, loss, and net spectra of X-rays as well as the density of radioactivity produced by X-rays and electrons within 19 energy bins in the range E_o to E = 6 MeV for 17 space points along the beam path within an irradiated sample. Using this program, they were able to confirm experimental results that there is no variation in the amount of induced activity due to a change in media if the parent material was present in constant amount.* Also they found, as would be expected, that the maximum activity produced shifts to greater depths as the energy increases and that for 24 MeV electrons the (e, e′n) reaction creates about one percent of the activity at a depth of one cm and becomes insignificant at four cm. Activities of several isotopes that are produced by irradiation of foods were also calculated. Unfortunately, their program is in their machine code form and not in a tractable form.

Evaluation of Hershman and Skaggs' equations

If we calculate the activity predicted by the Hershman Equation (equation 7), for example by Na^{22} production in beef, we obtain values that are not realistic. Also Skaggs' Equation, (equation 9) can only be evaluated for 27.5 MeV electrons or by the use of known integrated cross sections. Consequently there appears to be no simple relation to predict activities produced in foods irradiated with electrons of various energies that agrees with experimental results.

Development of Equation for predicting induced activity

With this in mind, a semi-empirical equation has been developed to calculate the activity induced by irradiating food with electrons of energy E. Before proceeding further, however, it should be pointed out that such an equation can be expected to yield only the activity produced within 20 to 50 percent, since one of the dominant terms must always be the fractional abundance of the parent isotope in a given food. To arrive at such a relationship, we must recall that the amount of activity produced is expected to be a function of the dose and the abundance of the target isotope. Also, it should be dependent upon the integrated cross section for the reaction producing the activity. This should

* This was for media such as Beef, Green Beans, and Water.

produce a net dependence on A, the mass number of the target nuclide, as well as some functional dependence on the initial electron energy. For the range of electron energies used in food irradiation, the form can be approximated by $(E - E_o)^n$ where E is the initial energy of the electrons and E_o is the threshold of the nuclear reaction producing the activity. The value of E_o may be obtained from any number of standard references such as the Nuclear Data Tables(42) or Hunt et al.(41). For convenience, a number of the more commonly encountered reactions and their threshold energies are given in Table 29. With these and the use of data given in Table 6, we arrive at:

$$R = K\,A\,n\,D\,T^{-1}\,(E - E_o)^3 \qquad (11)$$

where:

R = activity in pc/gm food/D Mrads
$K = 4 \times 10^{-3}$
A = atomic number of the target isotope
n = fractional abundance of the target isotope in the food*
D = dose in megarads
T = half life of product activity in years
E = initial electron energy in MeV
E_o= threshold energy for the reaction producing the product activity

Comparison of results with Equation 11 and those of Hershman and Newkirk

The results of Equation 11, Hershman and Newkirk, for the production of Na^{22} in beef and ham irradiated to 5 megarads with electrons of energy E were compared with the experimental values given in Table 6 and are presented in Table 30 and plotted in Figure 3. Upon inspection of Table 30, it appears that the average amount of activity produced in a given food can be predicted by Equation 11 to within the known variance of those elements in foods, and that it yields values closer to experimentally measured activity than do previous relationships.

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* Hence $n = f_i f_e$ of Equation 3.

TABLE 29

THRESHOLD ENERGIES AND PRODUCT HALF LIVES OF THE MORE COMMONLY ENCOUNTERED NUCLEAR REACTIONS

Reaction	Product Half Life	Threshold(E_γ in MeV)
$C^{12}(\gamma,n)C^{11}$	20 m	18.7
$N^{14}(\gamma,n)N^{13}$	10 m	10.6
$O^{16}(\gamma,n)O^{15}$	2.1 m	15.7
$Na^{23}(\gamma,n)Na^{22}$	2.6 y	12.4
$Mg^{25}(\gamma,p)Na^{24}$	15.4 h	12.1
$S^{34}(\gamma,p)P^{33}$	25 d	10.9
$S^{33}(\gamma,p)P^{32}$	14 d	18.8
$Cl^{37}(\gamma,np)S^{35}$	87 d	16.1
$Fe^{56}(\gamma,n)Fe^{55}$	2.6 y	11.2
$Zn^{66}(\gamma,n)Zn^{65}$	245 d	11.0
$Rb^{85}(\gamma,n)Rb^{84}$	33 d	10.5
$I^{127}(\gamma,n)I^{126}$	13 d	9.2
$Cs^{133}(\gamma,n)Cs^{132}$	6.5 d	9.0

TABLE 30

SODIUM-22 ACTIVITY IN HAM AND BEEF

(Activity in pc/gm food/5 Mrads)

Source	at Electron Energy (MeV):			
	24	20	18	16
HAM				
Experimental	2.2	0.94	0.45	0.06
Equation 11	2.3	0.67	0.29	0.08
Hershman(Eq. 7)	10.2	8.5	7.7	6.8
BEEF				
Experimental	0.21	0.01	0.008	0.006
Equation 11	0.16	0.04	0.018	0.005
Hershman, et al.	0.70	0.58	0.52	0.46
Newkirk, et al.	0.17	----	0.063	-----

For the production of I^{126}, Equation 11 predicts an activity of 0.01 pc/gm beef/5 Mrads while the experimental value is 0.007 pc/gm beef/ 5 Mrads(5). Also it is interesting to note that Equation 11 predicts 5.8 pc/gm beef/5 Mrads for the Na^{24} activity produced by the Mg^{25} $(\gamma,p)Na^{24}$ reaction in beef irradiated to 5 Mrads with 24 MeV electrons. This is to be contrasted to the value of approximately 14 pc/gm beef/5 Mrads predicted(50) and the experimental value of 12 pc/gm beef/5 Mrads. However, in this case it must be recalled that at least part of the Na^{24} activity should arise from the $Na^{22}(n,\gamma)Na^{23}$ reaction.

It should be pointed out that Newkirk et al. computer program is for detailed study of the activity produced as a function of depth into the irradiated food sample, while Equation 11 is for the average activity induced in a given sample. Consequently, with the use of Equation 3 and Equation 11, a fairly complete estimate of the activity levels in foods should be possible to a reasonably accurate degree.

9. Summary and discussion

Activities found

The major question of what, if any, long-lived activity is produced during food irradiation with electrons seems to be settled(1 to 11). The only long-lived detectable activity produced is that of Na^{22}, and at the highest electron energies used in food irradiation to date (24 MeV), the level of Na^{22} activity is only five parts in one thousand of the 168-hour week maximum permissible concentration(MPC) set by the National Committee on Radiation Protection for non-radiation workers(15). All other activities that are produced either have half lives of less than a day so that they do not constitute an ingestion problem, viz., they decay within a short time after irradiation or, if present, exist at levels below detectability. Other activities that might be mentioned here are: First, those of phosphorus which are beta emitters but present at levels just above detectability. For example, food irradiated with 24 MeV electrons to 5 Mrad doses will have P^{32} (14-day half life) and P^{33} (25-day half life) present at levels less than three parts in ten thousand of their MPC values. The only other activity that has been detected in an irradiated food is Rb^{87} which was found at a level of 0.05 pc/gm beef/ 5 Mrads or less than thirty parts in a million of its MPC value. This latter activity, which has been found in only two cases of many irradiations of beef, may be an example of over concentration in a very small number of cases of what would normally be a trace element. However, that such a variation can occur leads to the necessity of further experimentation to see if the only known samples (two) to produce this activity upon irradiation are oddities or truly random occurrences. Also it would be well to have further data on the activity induced at various energies in several food types to corroborate the existing data, not to mention the need to rectify the present lack of reliable information on beta activity levels in various foods.

Comparison of results with MPC values or with natural activities

Since MPC values are prescribed by a Committee, it might be useful to find a second method of comparison, such as the activity already present in food before irradiation. The exact pre-irradiation level of activity will depend on the level of fallout activity present at a given time. However the K^{40} activity* present from natural sources constitutes the predominant portion of the total natural activity present. Consequently the K^{40} activity level in a given food appears to be the most reasonable to be employed as a basis for comparison. In order to obtain an idea of the decay of the total activity in relation to K^{40} activity, the induced activity was calculated for several of the longer-lived activities at a number of time intervals after the end of irradiation. The data from earlier sections were used to calculate the values given in Table 31 and are for beef irradiated to 5 Mrads with 24 MeV electrons. From this can be calculated the ratio of total activity to natural activity present at various times after irradiation by using the value for K^{40} content in beef as given previously. The results of such a calculation are given in Table 32.

TABLE 31

INDUCED ACTIVITY FROM 24 MEV ELECTRON IRRADIATION OF BEEF

(Activity in pc/gm beef/5 Mrads)

Isotope ($t_{1/2}$)	Time after irradiation (in months) 0	1	2	6	12
Na^{22} (2.6 y)	0.19	0.19	0.18	0.16	0.15
P^{32} (14.3 d)	0.17	0.04	0.008	0.0003	3×10^{-9}
P^{33} (25 d)	0.60	0.25	0.11	0.0048	2×10^{-5}
S^{35} (87 d)	0.003	0.002	0.002	0.0008	2×10^{-5}
Mn^{54} (291 d)	0.004	0.0004	0.0003	0.0002	1.6×10^{-4}
Fe^{55} (2.7 y)	0.011	0.011	0.011	0.010	9×10^{-3}
Zn^{65} (245 d)	0.004	0.004	0.003	0.002	10^{-3}
I^{126} (13.3 d)	0.007	0.001	0.003	10^{-6}	10^{-10}

* K^{40} has a half life of 1.27×10^9 years and an abundance of 0.0118 percent in natural potassium.

TABLE 32

RATIO OF INDUCED LONG-LIVED ACTIVITY TO NATURAL ACTIVITY IN

BEEF IRRADIATED TO 5 MRADS WITH 24 MEV ELECTRONS

	Time After Irradiation (Months)				
	0	1	2	6	12
Total Gamma/K^{40}:	0.054	0.053	0.049	0.044	0.040
Total Beta/K^{40}:	0.18	0.079	0.035	0.0046	0.0025

From inspection of the table, it is evident that the long-lived activity produced, even at 24 MeV, is only a fraction of the activity already present in food. Further, it must be recalled that certain additives can increase the natural activity present by as much as a factor of ten, consequently reducing the induced activity to less than one half a percent of the natural activity. Use of certain high potassium content condiments can decrease this ratio even further. At the same time the variation in amount of trace elements that give rise to induced activity should be noted. For example, although the sodium content of beef is 0.5 parts per thousand, it can be as much as seven parts per thousand in meats such as ham. However, the variance is from food type to food type and not within a given food item. This was shown by Glass and Smith who could find only a ten percent variation in the amount of sodium in beef from various sources and cuts, while the calcium content was found to vary by a factor of two.* In general it is reasonable to estimate that the long-lived activity induced in foods will not exceed the amount of activity present without irradiation and in most cases will be insignificant in comparison to the natural activity.

Neutron fluxes, a cause of radioactivity: no data

At the lower energies used in electron sterilization of foods, neutrons appear to be the cause of a large portion of the induced activity. Consequently there exists a need for accurate information on the neutron fluxes that arise within the food; for example, from (γ,n) reactions within the food and can, as well as from external sources. At present there are only limited data on this subject, making neutron flux measurement one of the areas most in need of investigation.

* The contradiction between these results and that of rubidium certainly calls for a more detailed study.

Tritium and nuclear isomers: none found

Two possible sources of activity induced in foods that have conclusively been shown to contribute no detectable activity are: tritium and nuclear isomers. The former could not be found above levels already present as natural activity in foods(5,27). Further, by direct experimental search for evidence of the (γ,H^3) reaction, only an upper limit could be set on the amount of tritium that might be produced(5). The latter, that of isomer activation, is possible at electron energies above one MeV. However, no isomer activity has been found in foods. Only by using elemental targets was it possible to detect how much activity was produced per gram of element. In combination with known elemental abundances, the amount of activity that could be present was calculated. These calculations show that with 24 MeV electrons, the highest level of a long-lived isomer activity, that of Te^{125m}, is 10^{-4} pc/gm food/5 Mrads which corresponds to one part in a million of its MPC value.

Predictive equations developed

From the data compiled in this paper, it has been possible to arrive at two equations of simple form to predict the activity induced in food by electrons as well as by isomer activation (Equations 11 and 3 respectively). These Equations can be used to predict with some degree of reliability the activity to be expected in a given food. Their main use is intended for those who desire to know the average levels to be expected from a given isotope, and not for any purely theoretical use. If the latter type of information is desired, then the complexity of the process demands the use of a computer. However, no readily available program has been reported to date, although Newkirk et al.(4) have described a program they designed for following the physical processes subsequent to 24 MeV electrons impinging and penetrating a simulated food package six inches long. The main results they report are the variation of activity as a function of depth of penetration of the electron beam within the package, and that the activity induced does not change upon change in the food media. Even with the results of Newkirk, there is a need for further work, particularly in producing a more tractable program. One matter that does become obvious from the use of the Equations just mentioned is the need for a more accurate knowledge of trace element composition in various foods and their variation within different food types.

The problem of handling irradiated material

Besides the problem of induced activity and its consumption, there is the problem of the handling of food items in their containers and short-lived activity induced therein during early periods following irradiation with high energy electrons (energy greater than 20 to 24 MeV). If tin-plate cans are to be used for containers in food irradiation with high energy electrons, then automatic handling and storage procedures and devices will most likely have to be developed due to the high level of activity induced in the tin-plate containers(5,29,31,33,50). A more promising container material appears to be high purity aluminum, for it yields negligible short-lived activity after irradiation, whereas the tin-plate containers have residual activity for a relatively long time. The third choice for a packaging material is plastics, which have no activity present within a few hours after irradiation with 24 MeV electrons. This material is perhaps the most nearly ideal from an energy loss standpoint as it degrades the electron beam very little. The major drawback is that plastic may become enbrittled during irradiation as well as being partially permeable. However it appears that laminates of aluminum foil and plastic may provide the ideal packaging if such can be developed satisfactorily and economically. While considering the problem of handling, it must be remembered that immediately following high energy electron irradiation the level of activity within the food itself will be high, requiring some sort of transferral device. However, this activity will all decay away in less than five hours, as is evident from Figure 7.

Conclusion

In general it appears that electron irradiation of food does not produce a significant amount of induced activity. Indeed the threshold appears to be at 14 or 15 MeV and even at the highest energy investigated, 24 MeV, the induced activity level in most foods is found to be less than 5% of the natural activity already present before irradiation. This is insignificant in comparison to the tenfold increase in activity possible with use of certain food additives and condiments.

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