

The Problem of Induced Radioactivity in the Use of High Energy Electrons (25 Mev) for Sterilization and Chemical Processing

By J. Ovadia,* F. Heinmets† and A. Herschman*

The ionizing radiations used until now for food preservation, sterilization of medical equipment and chemical processing have been mainly confined to the gamma rays emitted by cobalt-60 or spent fuel elements from reactors. It appears, however, that high energy electrons produced by accelerators may possess certain advantages over gamma rays from radioactive isotopes. In particular, accelerators have the basic advantage that the radiation can be turned on or off at will and high currents are available which provide a high dose rate.

High energy electrons produce a substantially uniform dose distribution in the irradiated sample, from the surface down to a depth determined by their energy and the density of the irradiated substance. The useful depth of penetration expressed in cm of unit density material is given numerically by dividing the energy of the electrons, expressed in Mev, by three. It therefore appears desirable to use electrons in the energy range of 20-30 Mev to provide uniform irradiation to objects 8 to 10 cm thick.

One of the problems inherent in the use of electrons with energy higher than approximately 10 Mev is the formation of radioactive isotopes in most elements. These are caused by various processes. (1) The gamma rays (bremsstrahlung) generated by the passage of the electron beam through the sample interact with nuclei causing them to disintegrate by neutron or proton emission; the probability of this process is described by the photodisintegration cross section of the element and it exhibits a resonance at gamma ray energies in the neighborhood of 20 Mev. (2) The electrons may interact directly with the nucleus, causing it to disintegrate. The electrodisintegration cross section of the target element describes the probability of this process, which exhibits no resonance and is two orders of magnitude smaller than the photodisintegration cross section. Higher order processes in which two or more neutrons, protons or even alpha particles are emitted become significant only at energies higher than those considered in this work.

In addition, the neutrons produced by photodisintegration, either in the sample or in the accelerator and associated structures, are thermalized in the sample and are themselves absorbed, producing radioactive isotopes by (γ , n) processes, as in a reactor.

This paper is concerned only with the first two processes of (γ , n) and (e, n) reactions. The amount of radioactivity produced in selected samples irradiated directly by a high intensity 25-Mev electron beam from a linear accelerator was measured. The experimental values were compared with calculations made on the assumption that the radioactivity arises from the photon spectrum produced in water by the high energy electron beam. The relatively good agreement between the experimental results and the calculated values permits an accurate prediction of the level of radioactivity to be expected in other elements not specifically investigated. The air dose resulting from the radioactivity of processed foods was evaluated for typical irradiation schedules, and the storage time required for the dose rate to fall below permissible levels was evaluated. These values apply also to the processing of organic substances, composed mainly of carbon, nitrogen, and oxygen. The health hazard from the radioactivity of ingested food irradiated with high energy electrons is considered last.

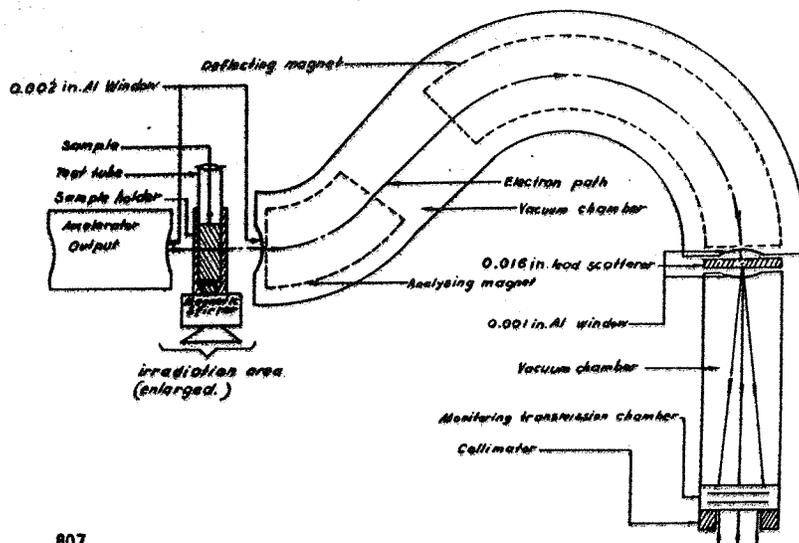
EXPERIMENTAL ARRANGEMENT

The source of the high energy electrons is a linear accelerator facility of the Tumor Clinic of Michael Reese Hospital in Chicago and has been previously described.^{1,2} The electron beam emerging from the accelerator is approximately 1 cm in diameter, has a non-uniform intensity distribution and delivers an average dose rate of the order of 10^6 rads/min of pulsed radiation. A magnet which analyzes the energy of the electrons in the beam and a scatterer used to increase its area attenuate the intensity by a factor of 10^3 to 10^4 ; no saturation problem is encountered in monitoring the electron beam with a transmission ionization chamber.

An irradiation site was set up at the end of the accelerator tube in order to utilize the high intensity available there. A schematic diagram of the experi-

* Tumor Clinic, Michael Reese Hospital, Chicago, Illinois.

† Quartermaster Food and Container Institute, Radiation Office, Chicago, Illinois.



807
Figure 1. Schematic diagram of the experimental arrangement used for irradiation

mental arrangement appears in Fig. 1. In order to simplify the analysis of the experimental data, 2-cm thick samples were used. The electron beam transmitted through the sample and analyzed by the magnet traverses the ionization chamber, whose current is a measure of the number of high energy electrons passing through the sample. A plot of activity induced in an ammonium iodide solution versus the integrated current read by the monitoring transmission chamber appears in Fig. 2. The fact that this plot is linear and goes through the origin means that the ionization chamber provides a good measure of the intensity of the high energy electron beam irradiating the sample and that no measurable radioactivity is produced by any contaminant high energy gamma rays in the background.

In order to compensate for the non-uniform intensity distribution of the beam, it was decided to irradiate magnetically stirred liquid samples whose cross-sectional area was larger than the beam. The dosimetry was performed with ceric sulfate solutions irradiated under identical geometric conditions. By thoroughly stirring the liquid samples and the ceric sulfate solution the non-uniform intensity of the beam is averaged over identical volumes and thus the average radioactivity in a sample can be compared with the average dose that produces it. No saturation effects arising from local depletion of target atoms can occur in the production of radioactivity. However, at the intensities used it was found essential to stir thoroughly the ceric sulfate dosimeter solution in order to avoid depletion of ceric ions in the central and most intense region of the beam.

The integrated transmission chamber current was calibrated against the corresponding quantity of ceric ion reduced, as measured with a spectrophotometer. A G value of 2.56 molecules/100 ev was used to convert the ceric sulfate data into rads.

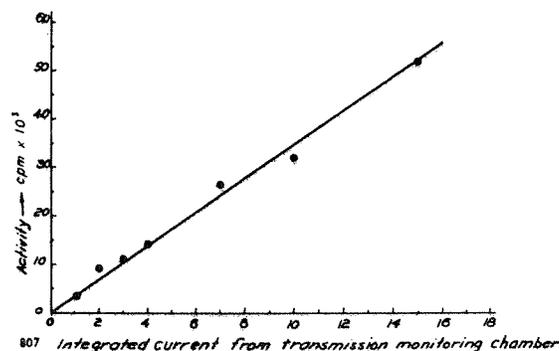
Elements leading to radioactivities with con-

veniently measured half lives and simple decay schemes were chosen to cover a wide region of the atomic table from an atomic number of 6 to an atomic number of 53. The following nuclides were measured (figures in parentheses represent half-lives): O^{15} in water (2 min), F^{18} in NaF solution (1.9 hr), Cl^{34} in NaCl solution (33 min) and I^{126} in NH_4I solution (13 d). In addition, samples of bacon and cheese were irradiated and the O^{15} and C^{11} (20 min) activities measured.

All the above elements decay by positron emission only, with the exception of Cl^{34} , where a gamma ray is also emitted, and I^{126} , which decays by emission of beta and gamma rays only. The annihilation radiations and the gamma rays were measured in a calibrated well-type sodium iodide scintillation counter, and the activity in μc per g of element was evaluated from the characteristics of the counter and the decay scheme of the radioactive isotope.

THEORETICAL CALCULATION

The radioactivity to be expected in the various samples was calculated on the assumption that it



807
Figure 2. Variation of the saturation activity in an ammonium iodide solution with the dose monitored in a transmission ionization chamber

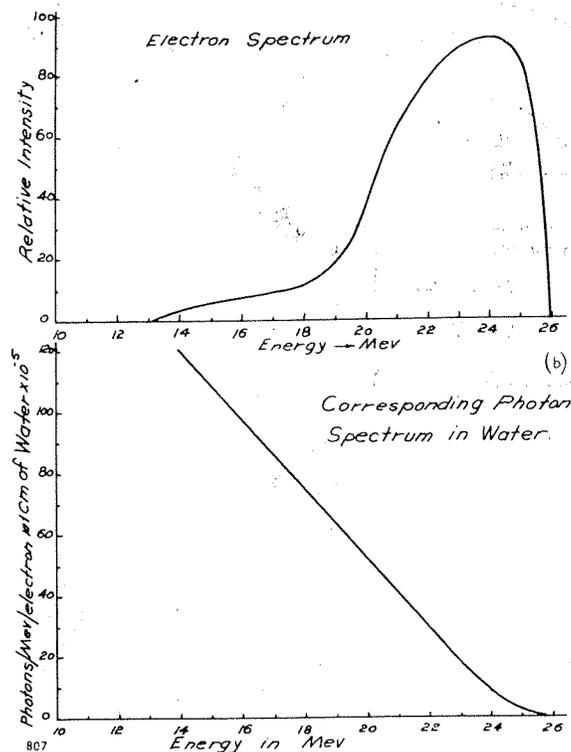


Figure 3. (a) Measured energy spectrum of the electrons emerging from the linear accelerator. (b) Calculated photon spectrum produced in 1 cm of water by the above electron spectrum

arises from bremsstrahlung generated in water by the high energy electron beam. A detailed analysis must take into account the scattering of the electron beam as it penetrates the sample and the simultaneous reduction of the electron energy. Although the theory of electron penetration is well understood, its application to thick samples leads to complex calculations which are beyond the scope of this work. By using a relatively thin sample, these effects were minimized. The gamma ray spectrum was calculated from the measured incident electron spectrum by using a theoretical expression applicable to thin targets. Figure 3(a) shows the measured electron spectrum and Fig. 3(b) is a plot of the corresponding photon spectrum in water calculated under the conditions described above. The absolute value of the number of photons per electron was obtained by taking the energy loss of electrons in the energy range of 20–30 Mev as 2 Mev per cm of water and using published values of the cross section for bremsstrahlung.³ Figure 4 shows the cross section for photodisintegration^{4,5} in carbon, oxygen, sodium and fluorine, superimposed on the photon spectrum. The radioactivity produced was obtained by integrating the photodisintegration cross sections of various elements of interest over the calculated photon spectrum. The production of radioactivity calculated by using this photon spectrum is an upper limit of the radioactivity

(a) Table 1. Radioactivity Produced by 10^6 rads of 25-Mev Electrons

Sample irradiated	Isotope produced	Half-life	Radioactivity, $\mu\text{c/g}$ of element	
			Experimental	Calculated
Water	O^{16}	2 min	1.24	1.8
NaCl solution	Cl^{34}	33 min	0.2	0.52
NaF solution	F^{18}	1.9 hr	0.124	0.14
NH_4I solution	I^{126}	13 d	0.006	0.008
Bacon	C^{11}	20 min	0.024	0.2
Cheese	O^{15}	2 min	0.4	1.8
	C^{13}	20 min	0.020	0.2
	O^{16}	2 min	0.3	1.8

to be expected, since in a thicker sample the spectrum present at greater depths is partially produced by electrons of reduced energy. Calculations for samples irradiated with electrons of energies higher than approximately 25 Mev must take into account a build-up of photons with energies lying within the photodisintegration resonance.

EXPERIMENTAL RESULTS AND COMPARISON WITH CALCULATED VALUES

Table 1 summarizes the experimental results and the theoretical calculations for 10^6 rads of 25-Mev electrons. The radioactivity produced is expressed in μc per g of element present in the irradiated sample. In the calculations, bacon was considered to consist of 22% oxygen and 59% carbon by weight, whereas cheese was taken as 45% oxygen and 36% carbon. It is noted that for water and the various salt solutions the experimental values agree with the calculated values to within a factor of 2, which is considered excellent in view of the drastic simplification made in the calculations. In the case of bacon and cheese, the experimental carbon yields are one order of magnitude smaller than the calculated value. This discrepancy is

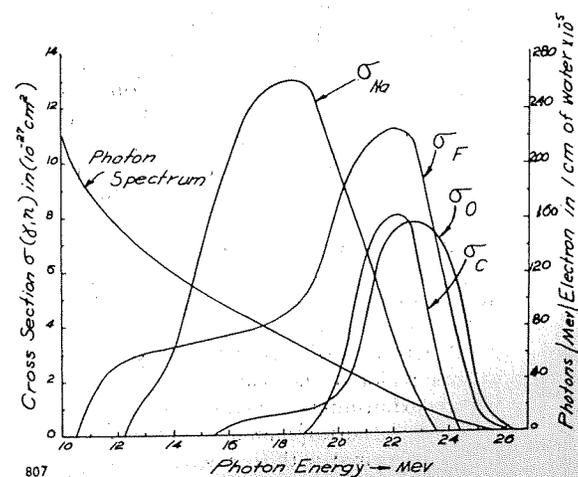


Figure 4. Photodisintegration cross section of C, O, Na and F superimposed on photon spectrum produced in 1 cm of water by electron spectrum of Fig. 3

Table 2. Activities in 1 Ton of Processed Food

Isotope	Half-life	Concentration in food where element is prevalent ^a	Maximum dose rate (mr/hr)	Necessary storage time	Safety factor
C ¹¹	20 min	67% (many foods)	2400	4.5 hr	—
F ¹⁸	1.9 hr	(10 ⁻³)% (sardines)	2.4 × 10 ⁻²	—	8
Cl ³⁴	33 min	1.2% (bacon)	230	5.5 hr	—
Na ²²	2.6 yr	1.6% (cheese)	6 × 10 ⁻³	—	30
Cu ⁶⁴	12.9 hr	2 × 10 ⁻³ % (liver)	7 × 10 ⁻³	—	30
I ¹²⁶	13 d	3 × 10 ⁻⁴ % (cheese)	1.3 × 10 ⁻²	—	15

attributed to the high value of the threshold for photo-disintegration in carbon, and the yield depends to a critical degree on the details of the high energy end of the photon spectrum. A search was made for Na²² (half-life, 2.6 yr) but no long-lived activity was detected in the sodium chloride solution. Taking into account the background and statistics, this means that less than 2 × 10⁻⁴ μc of Na²² is present per g of sodium per Megarad of 25-Mev electrons. Since the information appearing in Table 1 is used to evaluate radiation hazards, the calculated values of radioactive yield, which are always found to exceed the experimental values, are used in the subsequent work. If the total radioactivity of a thick sample is calculated by multiplying the radioactivity per g, applicable to a thin sample, by the total weight, the total radioactivity thus calculated is an overestimate of the actual value. This procedure was followed, however, since it provides an additional safety factor.

RADIOACTIVITY AT IRRADIATION SITE

The radiation hazard presented by irradiated products was calculated for the following conditions:

- (1) A sterilizing dose of 5 × 10⁶ rads is delivered.
- (2) The production schedule is 3 tons per hr of unit density material.
- (3) The irradiated food is stored in cubic stacks of 1 m side.
- (4) The external dose rate is due only to gamma rays and annihilation radiation and is calculated for an effective distance of 50 cm. The beta radiations are absorbed within the sample and do not contribute appreciably to the external dose.
- (5) In calculating the activity of various elements present in small amounts in some foods, the particular food in which that element is prevalent is used for the calculations.

(6) The permissible dose rate is taken as 0.25 mr/hr corresponding to a weekly dose of 10 mr. This is a value of permissible exposure level applicable to the general population.⁶

The results of the calculations appear in Table 2. The concentration of iodine was obtained by assuming that iodized salt containing 0.01% potassium iodide is used. The required storage time and the safety factor were calculated by comparing the dose rate from the irradiated food with the value of 0.25 mr/hr considered permissible.

It is noted that C¹¹ (half-life, 20 min) and in rare instances Cl³⁴ (half-life, 33 min) create a health hazard. This means that the irradiated products cannot be handled directly by personnel immediately after processing, but must be placed in storage spaces by mechanical equipment. A storage space in which irradiated food can be stored for approximately 5 hr must be available.

The principle of the above calculation can be applied directly to processing of organic substances, since the concentration of carbon in them is of the same order of magnitude as in food and since it would be the only isotope creating any handling or storage problem.

The problem of the continuous irradiation of metallic structural elements has not been considered. Aluminium appears an excellent metal to use since only short-lived radioactivities are produced. The use of iron or brass in the direct path of the electron beam may create serious problems in maintenance arising from the production of the long-lived activities of Fe⁵⁵ and Zn⁶⁵.

HEALTH HAZARD FROM INGESTED FOODS

The dose absorbed by various organs exposed to the radiations from ingested radioactive foods was calculated for the following conditions:

Table 3. Maximum Radioactivity of Ingested Irradiated Food

Element	Amount ingested g/day	Radioactive isotope of interest	Effective half-life	Allowable specific activity μc/g	Activity produced μc/g	Safety factor
Carbon	300	C ¹¹	20 min	7 × 10 ⁻³	0.1	—
Fluorine	5 × 10 ⁻³	F ¹⁸	1.9 hr	340	0.5	7 × 10 ²
Sodium	4	Na ²²	29 d	6 × 10 ⁻³	5.7 × 10 ⁻⁵	10 ²
Chlorine	6.7	Cl ³⁴	33 min	6 × 10 ⁻²	0.84	—
Calcium	0.8	Ca ⁴⁵	152 d	300	4 × 10 ⁻⁶	10 ¹⁰
Calcium	0.8	Ca ⁴⁷	4.8 d	6	8 × 10 ⁻⁶	10 ⁶
Iron	1.2 × 10 ⁻²	Fe ⁵⁵	65 d	26	3 × 10 ⁻⁴	10 ⁶
Copper	2 × 10 ⁻³	Cu ⁶⁴	12 hr	350	0.2	1.7 × 10 ³
Iodine	2 × 10 ⁻⁴	I ¹²⁶	13 d	2.2	3.8 × 10 ⁻⁴	6 × 10 ³

(1) All the food consumed has been irradiated with a sterilizing dose of 5×10^6 rads.

(2) Values for the daily ingestion rate of various elements, the specification of the nature, the dimensions and the weight of the critical organ for a particular element, and the biological half-life of the isotope in that organ were obtained from the British Journal of Radiology.⁷

(3) The allowable weekly dose rate to the critical organ is 10 mr.

(4) The stomach is considered the critical organ for all activities with a half-life smaller than 3 hours, and the upper colon is considered the critical organ for isotopes having a half-life in the neighborhood of 12 hours.

The results of the calculations appear in Table 3.

It is again observed that only the short-lived activities of carbon and chlorine would be objectionable. However, the storage time required in processing would eliminate this problem, and, of course, one would not consider irradiating food intended for consumption immediately. The safety factors of all the long-lived isotopes are considerable, even under the quite low exposure rates considered permissible.

ACKNOWLEDGEMENT

The authors wish to thank Mr. John Duplex, the accelerator engineer whose help and assistance in constructing various mechanical devices and equipment, and in performing the irradiations, was most valuable.

REFERENCES

1. E. M. Uhlmann, C. L. Hsieh and C. L. Lootens, *The Linear Accelerator as a Source of Fast Electrons for Cancer Therapy*, Radiology, 66, 859-70 (1956).
2. C. L. Hsieh and E. M. Uhlmann, *Physical Characteristics of a 45 Mev Medical Linear Accelerator*, Radiology, 67, 263-72 (1956).
3. W. Heitler, *The Quantum Theory of Radiation*, Oxford University Press, Oxford (1944).
4. R. Montalbetti, L. Katz and J. Goldenberg, *Photoneutron Cross Sections*, Phys. Rev., 91, 659-73 (1953).
5. G. A. Ferguson, J. Halpern, R. Nathans and P. F. Yergin, *Photoneutron Cross Sections in He, N, O, F, Ne and A*, Phys. Rev., 95, 776-80 (1954).
6. L. S. Taylor, *Current Situation with Regard to Permissible Radiation Exposure Level*, Radiology, 69, 6-11 (1957).
7. International Commission on Radiological Protection, *Recommendations of the I.C.R.P.*, British Journal of Radiology, Supplement No. 6 (1955).
8. H. A. Wooster Jr. and F. C. Blanck, *Nutritional Data*, H. J. Heinz Co., P.O. Box 57, Pittsburgh (1949).