R66-51

Health Physics Pergamon Press 1966. Vol. 12, pp. 1581–1587. Printed in Northern Ireland

CALCULATION OF INDUCED RADIOACTIVITY IN ELECTRON STERILIZED FOOD

R. A. MEYER*
U.S. Army Natick Laboratories, Radiation Laboratory, Natick, Massachusetts

and

J. L. BURKHARDT
17 Center Street, Watertown, Massachusetts

(*Received* 30 *August* 1965; *in revised form* 21 *March* 1966)

Abstract—The basis for a tractable equation to predict the amount of radioactivity induced in food from high energy electron sterilization is shown. This equation yields results which are in agreement with previous experimental results. Also, it appears that by using thin plastic packaging the threshold of detectable induced activity may be raised to as high as 17 or 18 MeV.

1. INTRODUCTION

THEORETICALLY, sterilization of food by electron irradiation can lead to detectable induced radioactivity when the electron beam energy is greater than 15 MeV. Most of this radioactivity results from photonuclear reactions produced by the X-rays (bremsstrahlung) accompanying electron absorption. Some additional activity arises from secondary neutron-induced reactions, and a negligible contribution comes from direct electron interactions.

It would be useful to have an equation in simple form which is based on known photonuclear parameters for calculating the expected activity for a given food and irradiation procedure. This paper describes both graphical and analytical methods for making such calculations as well as providing justification for the use of a tractable empirical equation. Only photonuclear reactions (which produce most of the radioactivity) are considered, and a few specific reactions have been computed in detail. The methods are generally applicable, however, and can be used whenever the elemental abundances are known.

2. BREMSSTRAHLUNG SPECTRA

The photon spectrum of thick-target bremsstrahlung is a problem of long standing. Both experiment and theory are very difficult, and little work of either kind has been published. Most of the data available are on high-density, high-Z targets, while in the case of electron sterilization of food, we are concerned with unit density low-Z ($\bar{Z} = 7.1$) targets.

The only bremsstrahlung investigation pertaining directly to food irradiation appears to be the computer calculation performed by NEWKIRK, SMITH and GLASS.[1] Here, we have used extrapolated spectra from those calculated by HANSEN and FULTZ.[2] They give thin-, intermediate- and thick-target bremsstrahlung spectra and energy loss curves for electrons of energies up to 35 MeV in aluminum ($Z = 13$), tin (50), tungsten (74), lead (82) and uranium (92). The bremsstrahlung spectrum for food was obtained from that for aluminum by noting that the radiation cross section is proportional to Z^2 whereas the energy loss per centimeter goes as Z for light elements;[3] thus the spectrum should be proportional to $\overline{Z^2}/\bar{Z}$. For food this ratio is 7.77,[4] while for aluminum it is $169/13 = 13$. Therefore the thick-target bremsstrahlung spectrum for water should be approximately 7.77/13 or 0.6 of that for aluminum. The curves derived on this basis are shown in Fig. 1.

3. INDUCED ACTIVITY

We assume that a mass M of unit density food material is irradiated with N electrons per unit area, distributed uniformly over the face of the food sample and delivered in a time short compared with the half-live T of the radioactive

* Present address: University of California, E. O. Lawrence Radiation Laboratory, Livermore, California. Also of the Department of Chemistry, Research Establishment, Risø, Denmark.

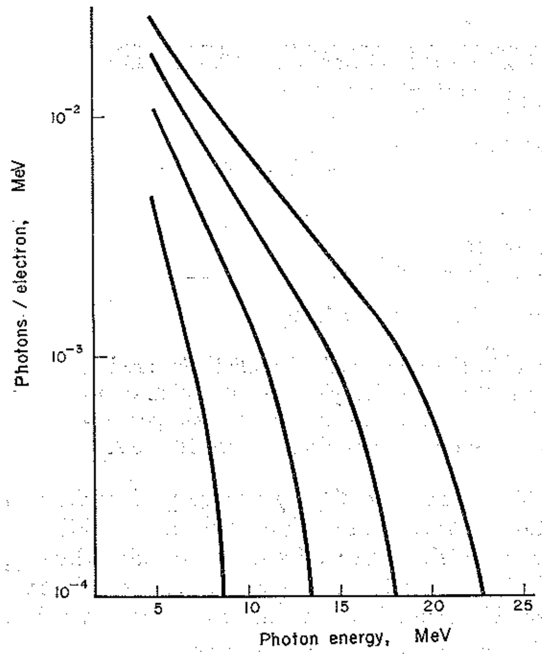

FIG. 1. Thick-target bremsstrahlung in water.

nuclei of interest. The food is unpackaged (or packed in a thin, low-Z material), and it has a surface area S exposed to the beam and a length L in the direction of the beam. The incident electrons have energy E.

Each electron, on the average, produces a photon spectrum like that shown in Fig. 1. We assume that these photons are produced at the entrance surface and that they travel through the food sample without appreciable attenuation. In the case considered here both assumptions are approximately valid for the highest energy photons, which are the most effective in producing photonuclear reactions.

The probability per target atom of producing a particular reaction with a photon of energy k is given by the appropriate cross section σ, a function of k. If there are N electrons per unit area incident on the sample, the probability that a given target nucleus will undergo a reaction is therefore

$$N\int_0^E \sigma \frac{dn}{dk}\, dk,$$

where dn/dk is the bremsstrahlung spectrum. The number of target nuclei per unit mass of sample is fN_0/A, where f is the fractional abundance by weight of the target isotope (atomic weight A) in the food, and N_0 is Avogadro's number. Thus finally, the number of radioactive nuclei present in the food after irradiation, per gram of food, is

$$R = \frac{NfN_0}{A}\int_0^E \sigma \frac{dn}{dk}\, dk. \quad (1)$$

The specific activity is $0.693R/T$ disintegrations/sec per g food or $18.7R/T$ pc/g food.

These expressions are independent of the size or shape of the package since a given number of electrons per unit area incident on the sample produce a certain specific activity, regardless of package size, as long as photon absorption in the sample can be neglected. This situation does not prevail with respect to dose, however. The total amount of energy delivered by the electrons to the sample is $NSeE$, so the average energy per unit mass, or average dose, is $NeE/\rho L$, which varies inversely with package depth. Furthermore, this energy is not deposited uniformly, and none at all is delivered to any portion of the package beyond the maximum range of the electrons. The purpose of electron sterilization is to deliver a certain actual dose to every portion of the food; average doses are meaningless. It is therefore very important to use food packages of the proper depth and to be certain that the electron beam is uniformly distributed over the face of each package.

In view of the above situation, it would be preferable if induced activity data were stated on the basis of a given electron flux on the package. Unfortunately, however, in the past experimental results have been given in either per μA-min of electron beam or per Mrad (or 5 Mrads) of dose. In order to compare the calculations in the remainder of this report with previous work, all activities have been normalized to a 5-Mrad average dose in a sample the size of a #10 can ($6\frac{3}{16}$ in. dia. by 7 in. long). This package is approximately twice as deep as optimum for 24 MeV electrons, and worse for lower energies; however, as mentioned, all experimental data to date have been given in this manner.

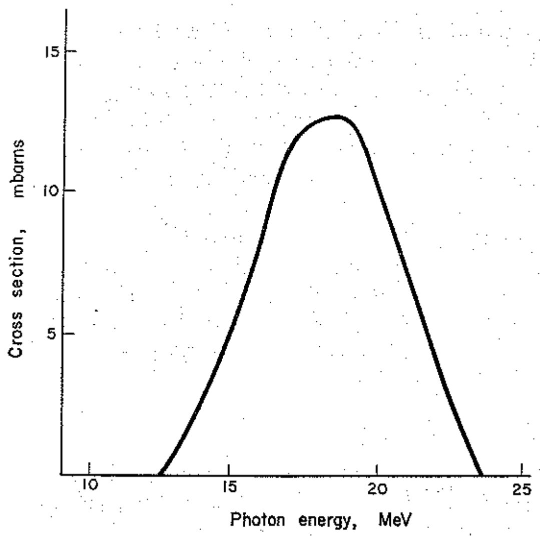

FIG. 2. Photoneutron cross sections ^{23}Na.

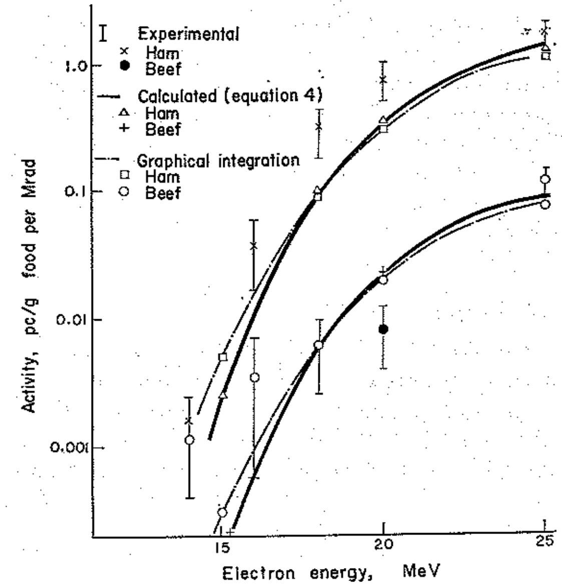

FIG. 3. ^{22}Na activity in meats. (Experimental data from refs.[7][16])

(a) *Sodium-22 activity*

The predominant long-lived activity found in irradiated food is the 2.58-yr ^{22}Na arising from the reaction ^{23}Na(γ, n)^{22}Na. The cross section for photoneutron emission from ^{23}Na has been measured by several investigators.[5,6] We have used the cross sections found by MONTALBETTI *et al.*,[5] shown in Fig. 2. The product of this cross section and the bremsstrahlung spectra were integrated graphically and their results are given in the second column of Table 1.

The specific activity in any type of food from any amount of irradiation at these energies can be found by multiplying the integrated cross sections by appropriate factors. Using equation (1) and the data given by MEYER, we have performed the computation for 5-Mrad doses to beef and ham in #10 cans. The ^{23}Na elemental abundances used in the computation were 5×10^{-4} for beef and 7.4×10^{-3} for ham.[4] The resulting predicted specific activities are given in Table 1 and plotted in Fig. 3.

Also shown in Fig. 3 are some experimental data taken from SMITH[7] and a theoretical curve which is presented in a subsequent section. The experimental data have been modified to take account of two factors:

Table 1. *Computed* ^{22}Na *activities*

Electron energy (MeV)	$\int \sigma \frac{dn}{dk} dk$ (cm^2)	Specific activity (pc/g food/5 Mrad) Beef	Ham
25	1.32×10^{-28}	0.085	1.3
20	2.9×10^{-29}	0.024	0.36
15	3.6×10^{-31}	0.0004	0.006

(i) These irradiations were performed in "tin" (iron) cans. Since the increased bremsstrahlung from the electrons passing through the can lids raises the observed activity over that which would have been obtained with no package by factors of 1.3, 1.6 and 3.3 at 25, 20 and 15 MeV respectively, the experimental points have been reduced to take account of this effect.

(ii) GLASS and SMITH[8] divided their raw experimental activities by factors designed to correct for the "non-ideal" size of the #10 cans at different energies. The magnitudes of these corrections are in the inverse ratio of the electron energies, that is 1, 1.25 and 1.67 for 25, 20 and 15 MeV; consequently, we have multiplied the experimental points by $25/E$ as well as the SRI computer results[1] which appear to have been performed for unpackaged food, with the "non-ideal package size" factor again included.

It is clear from Fig. 3 that the graphical integration agrees very well with the experimental data. None of the meat samples used in the experiments were analyzed for sodium content; consequently, some vertical displacement of the curves is to be expected. However, this variation is less than a factor of 2.[7,8] On the other hand, SMITH and GLASS calculated curve appears to be seriously in error, particularly at lower electron energies. The difference between our results and their curve is doubtless due to the difference in the shapes of the bremsstrahlung spectra computed by SMITH and GLASS[8] and BRAMBLETT *et al.*[9]

(b) *Short-lived activities*

Considerable amounts of ^{11}C, ^{13}N and ^{15}O activity are present in irradiated foods immediately after processing. These isotopes have half-lives of 20.5, 10.0 and 2.07 min, respectively, so they are of interest principally with regard to handling the food immediately after irradiation.

Measured cross sections for photoneutron reactions in ^{12}C, ^{14}N and ^{16}O are shown in Fig. 4.[5,10–12] The cross sections chosen for graphical integration are indicated in the figure, and the results of the integration for 25-MeV bremsstrahlung are given in Table 2. Also listed in the table are the corresponding specific activities used, computed with the aid of equation (1), for 5-Mrad doses to unpackaged beef samples the size of a #10 can. The elemental abundances used in the calculation are 0.18, 0.023 and 0.70 for ^{12}C, ^{14}N and ^{16}O, respectively.[4] Radioactivity levels calculated by SRI and several experimental results are also shown in the table.

(c) *Sodium-24 activity*

The only other activity regularly observed in irradiated food is 15-hr ^{24}Na. This isotope is produced by photoproton emission from ^{25}Mg and by neutron-capture in ^{23}Na. The photo-

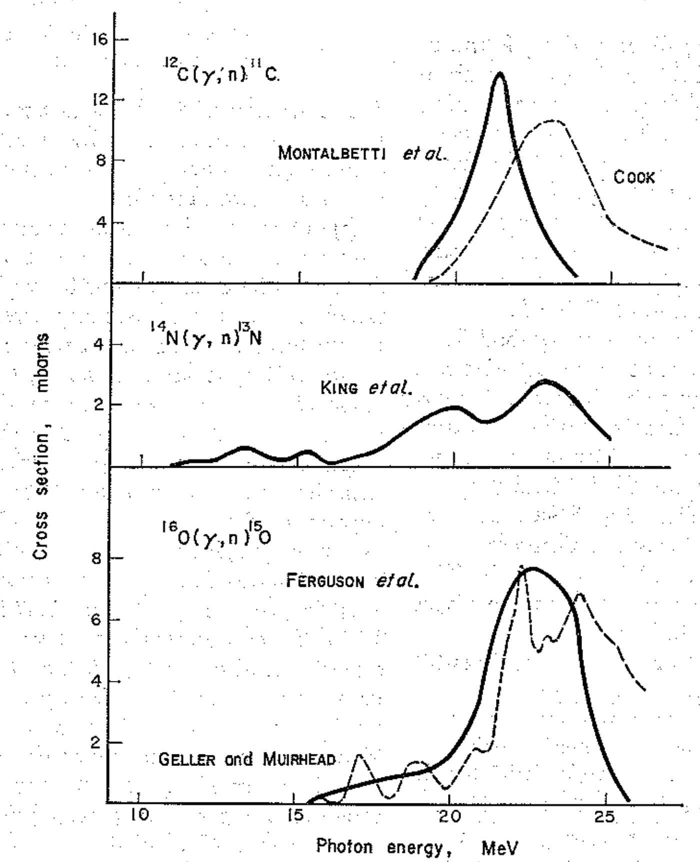

FIG. 4. Photoneutron cross sections. Solid lines denote cross sections used for graphical integration.

Table 2. Short-lived activities at 25 MeV in beef

	$\int \sigma \frac{dn}{dk} dk$	Specific activity (μg beef — 5 Mrad)				
		Calculated		Experimental		
Isotope	(cm²)	equation (1)	SRI	A	B	C
^{11}C	1.3×10^{-29}	0.40	1.0	0.36	0.31	0.36
^{13}N	1.7×10^{-29}	0.12	—	0.16	0.075	—
^{15}O	9.1×10^{-30}	7.9	35	0.44	10	7*

Notes: Experiment A: 24 MeV, beef: Ref. (11), p. 17.
Experiment B: 24 MeV, Enriched beef: Ref. (11), p. 18.
Experiment C: 24 MeV, beef: Ref. (8), p. 59.
* Measured in green beans; value computed for beef.

proton contribution is calculated below; however, contribution by capture process is too complicated for this type of analysis.

The cross section for the $^{25}Mg(\gamma, p)^{24}Na$ reaction is shown in Fig. 5.[13–15] Table 3 gives the results of the graphical integration for bremsstrahlung of three energies and the predicted ^{24}Na activities in beef from this reaction. The fractional abundance of ^{25}Mg in beef was taken[4] as 2.7×10^{-5}. The experimental results shown in the table have been corrected for the effect of bremsstrahlung from the tin can and for the "ideal package size" factor, as explained earlier. As previously shown by MEYER,[4] the measured activities are substantially higher than the calculations, almost certainly because of neutron capture in ^{23}Na, especially at low electron energies near the 12.1 MeV (γ, p) threshold in ^{25}Mg.

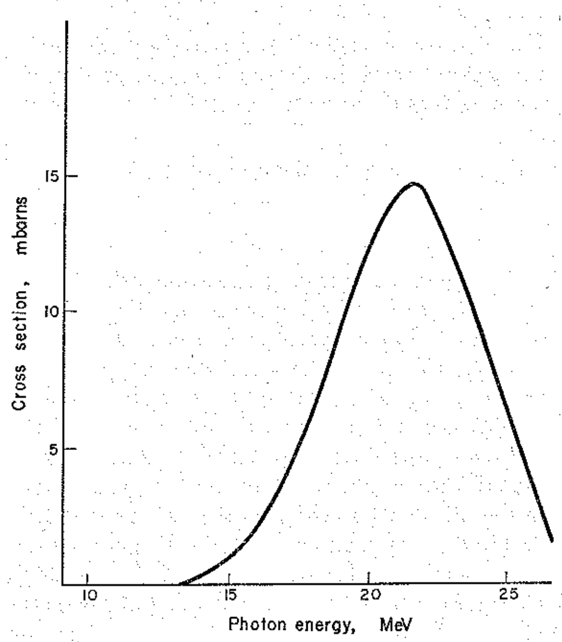

FIG. 5. Photoproton cross section ^{25}Mg.

4. ANALYTIC SOLUTION

The calculations in the previous section require graphical integration of the product of the reaction cross section and the bremsstrahlung spectrum. While this process is not difficult, it would be desirable to have a simple formula for the induced activity from a given reaction as a function of electron energy. Such an equation is MEYER's equation:[4,16]

$$R = KAnDT^{-1}(E - E_0)^3 \quad (3)$$

where:

R = activity in pc/g 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 on product activity in years,
E = initial electron energy in MeV,
E_0 = threshold energy for the reaction producing the product activity.

Here we investigate the basis for such an equation. The bremsstrahlung spectra of Fig. 1 are replotted in Fig. 6 as a function of $E - k$, electron beam energy minus the photon energy. The bremsstrahlung spectra can be represented, with an error which never exceeds 5 per cent, by the single expression.

$$\frac{dn}{dk} = \frac{6.18 \times 10^{-3}}{E^{1.7}} (E - k)^{2.2}.$$

Table 3. Activities of ^{24}Na *in beef*

Electron energy (MeV)	$\int \sigma \frac{dn}{dk} dk$ (cm^2)	Specific activity (pc/g beef — 5 Mrad) equation (1)	Experimental[16]
25	6.1×10^{-29}	3.1	8.0 ± 0.9
20	6.0×10^{-30}	0.38	2.9 ± 0.4
15	6.0×10^{-33}	0.0005	0.25 ± 0.08

A typical photonuclear reaction cross section has a resonance shape which might be approximated for calculational purposes by a triangle, an inverted parabola, a Gaussian curve or some more complicated expression. However, for our purposes, the simplest approximation is a rectangle, where we require:

$$\sigma = \begin{cases} 0 & k < E_1 \\ \sigma_m & E_1 < k < E_2 \\ 0 & k > E_2 \end{cases}$$

The values of σ_m, E_1 and E_2 are selected to give a proper representation of the shape of the cross section and to agree with the proper area of $\int \sigma dk$.

Using the two expressions given above, the required integral of the cross section and bremsstrahlung spectrum can be computed.

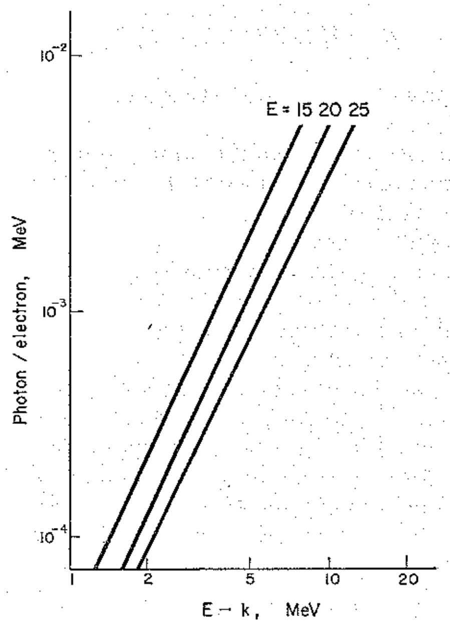

Fig. 6. Thick target bremsstrahlung in water.

For $E < E$:

$$\int_0^E \sigma \frac{dn}{dk} dk = 0.$$

For $E_1 < E < E_2$:

$$\int_0^E \sigma \frac{dn}{dk} dk = \frac{6.2 \times 10^{-3} \sigma_m}{E^{1.7}} \int_{E_1}^E (E - k)^{2.2} dk$$

$$= \frac{1.9 \times 10^{-3} \sigma_m (E - E_1)^{3.2}}{E^{1.7}}.$$

For $E > E_2$:

$$\int_0^E \sigma \frac{dn}{dk} dk = \frac{6.2 \times 10^{-3} \sigma_m}{E^{1.7}} \int_{E_1}^{E_2} (E - k)^{2.2} dk$$

$$= \frac{1.9 \times 10^{-3} \sigma_m}{E^{1.7}} \{(E - E_1)^{3.2} - (E - E_2)^{3.2}\}.$$

For electron energies less than about 30 MeV, the second term in the last expression can be neglected, and the three results can be combined as:

$$\int \sigma \frac{dn}{dk} dk \simeq \frac{1.9 \times 10^{-3} \sigma_m (E - E_1)^{3.2}}{E^{1.7}} \quad E > E_1 \tag{4}$$

To give an example of the application of this formula, we have computed the ^{22}Na activity in beef and ham at various energies. The values of E_1 and E_2 are 14 and 22 MeV, respectively, and σ_m was taken as 9.75 mbarns. The calculated results are compared with the results of the graphical integration in Table 4, and the predicted ^{22}Na activities are plotted as dashed lines in Fig. 3. Besides justifying the use of the simple rectangular approximation to σ the good agreement confirms Meyer's equation[4,16] in which the energy dependence is contained in the factor $(E - E_0)^3$, where E_0 is the threshold for the reaction.

Table 4. ^{23}Na (γ, n) ^{22}Na *reaction*

Electron energy (MeV)	$\int \sigma \frac{dn}{dk} dk$ (cm^2)	
	Equation (4)	Graphical integration
25	1.51×10^{-28}	1.3×10^{-28}
20	3.24×10^{-29}	2.9×10^{-29}
15	1.80×10^{-31}	3.6×10^{-31}

Equation (4) can be used to find the activity resulting from any reaction for which the cross section is known. If the cross section has not been measured, a rough estimate can be made from a knowledge of the systematics of the dipole resonance.[17] In heavy elements ($Z > 50$) the energy of the peak of the resonance is found to follow roughly the formula $82A^{-1/3}$ MeV, and the area under the photon-absorption cross section is approximately $75Z(A - Z)/A$ MeV-mbarn. A very large fraction of this cross section goes into neutron emission. The photonuclear threshold can usually be found from nuclear mass data, and the shape of the cross section can then be estimated. In lighter elements the photoneutron and photoproton cross sections become more nearly equal, and the total cross section drops to the order of $35Z(A - Z)/A$ MeV-mbarn. The energy of the maximum is generally within the range 20 ± 2 MeV. Thresholds can again be obtained from mass data. Estimates of induced activity made with these approximate cross section parameters can only be trusted as to order of magnitude, but they give a better approximation to induced activity in food than those obtained with formulas of the type developed by HERSCHMAN[18] and SKAGGS.[19]

In general, we can state that there is valid basis for an equation in simple form such as MEYER's equation. Also, from the results of calculating "tin" (iron) can bremsstrahlung as well as thin and thick target bremsstrahlung, it appears that by the use of thin packages in the electron sterilization of food in low Z materials (such as plastic packaging) will yield no detectable activity using electrons of energy as high as possibly 17 or 18 MeV. This in itself is a very important point which demands further experimental investigation.

REFERENCES

1. W. H. NEWKIRK, H. D. SMITH and R. A. GLASS, *Stanford Research Institute Project SU-2424 Report No.* 11 (1960).
2. N. E. HANSEN and S. C. FULTZ, *University of California, Lawrence Radiation Laboratory Report UCRL*-6099 (1960).
3. W. HEITLER, *Quantum Theory of Radiation* (3rd Ed.). Clarendon Press, Oxford (1954).
4. R. A. MEYER, *U.S. Army Natick Lab. Tech. Rep., FD*-6 (1964).
5. R. MONTALBETTI, L. KATZ and J. GOLDEMBERG, *Phys. Rev.* **91,** 659 (1953).
6. K. SATO, *J. Phys. Soc. Japan* **18,** 1353 (1963).
7. H. D. SMITH, *Stanford Research Institute Project PHU*-2424 *Report No.* 17, p. 26 (1962).
8. R. A. GLASS and H. D. SMITH, *Stanford Research Institute Project SU*-2424 *Report No.* 10 (1960).
9. R. BRAMBLETT, J. CALDWELL, R. HARVEY and S. FULTZ, *Phys. Rev.* **133,** B869 (1964).
10. B. C. COOK, *Phys. Rev.* **106,** 300 (1957).
11. J. KING, R. HASLAM and R. PARSONS, *Can. J. Phys.* **38,** 231 (1960).
12. G. FERGUSON, J. HALPERN, R. NATHANS and P. YERGIN, *Phys. Rev.* **95,** 776 (1954).
13. K. N. GELLER and E. G. MUIRHEAD, *Phys. Rev. Lett.* **11,** 371 (1963), as corrected in footnote 14 of *Phys. Rev.* **136,** B418 (1964).
14. L. KATZ and A. G. W. CAMERON, *Phys. Rev.* **84,** 1115 (1951).
15. M. E. TOMS and W. E. STEPHENS, *Phys. Rev.* **82,** 709 (1951).
16. R. A. MEYER, *Health Phys.* **12,** 1027 (1966).
17. E. HAYWARD, *Rev. Mod. Phys.* **35,** 324 (1963).
18. A. HERSCHMAN, *U.S. Army Quartermaster Food and Container Institute Project* 7-84-01-001 *Report No.* 1 (1956).
19. L. S. SKAGGS, *Argonne Cancer Research Hospital Report No.* 541/5 (1956).