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Yields of Radionuclides Created by Photonuclear Reactions on Be, C, Na, Cl, and Ge, Using Bremsstrahlung of 150-MeV Electrons

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Physics Division

Yields of Radionuclides Created by Photonuclear Reactions on Be, C, Na, Cl, and Ge, Using Bremsstrahlung of 150-MeV Electrons

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ABSTRACT

The bremsstrahlung created by 150-MeV electrons impinging on a tantalum radiator was used to study photonuclear reactions on samples containing Be, C, Na, Cl and Ge. For Ge fifteen radioisotopes, ranging in half life between 2.6 min and 271 days, and in mass between'65 and 75, were obtained in sufficient amount to determine their yields quantitatively using'known decay gamma-rays. Special equipment is described which was developed to create the bremsstrahlung using a beam-sharing mode, while minimizing the neutron flux on the sample. Relative production rates were determined. These were analyzed to provide absolute average cross sections for production of three reactions:

$\langle\sigma\rangle$ for $^{35}Cl(\gamma,n)^{34}Cl^{isom}$ = 4.7 mb;

$\langle\sigma\rangle$ for $^{70}Ge(\gamma,n)^{69}Ge$ = 56 mb; and

$\langle\sigma\rangle$ for $^{76}Ge(\gamma,n)^{75}Ge$ = 53 mb,

where the (γ,n) values are averages over the giant resonances of the stable target isotopes.

1.0 INTRODUCTION

As part of a now-terminated program at the Oak Ridge Electron Linear Accelerator (ORELA) facility to measure and report[1] photon-production cross sections for neutron interactions with elements for applications to technology and scientific disciplines, an experiment[2] was initiated to study photonuclear effects also using the ORELA in a beam-sharing'mode. Earlier measurements[3,4] using the ORELA in a dedicated beam mode for photonuclear reactions on Ni (ref. 3) and Ta (ref. 4) verified that non-exclusive (i.e., no chemical separations) gamma-ray-spectra from the irradiated sample 'could be analyzed to obtain yield information for radionuclides created during the photon beam irradiation. The first measurements using the new system (which is discussed in the next section) were for Si, and results, both experimental and calculational, were presented by Wunstorf, et *al.*[2] In this report, results of similar measurements on samples of Be, NaCl, Na_2CO_3, and Ge are presented and discussed.

2.0 EXPERIMENTAL DETAILS

The experiment consisted of irradiating samples in a photon beam created by 150-MeV electrons impinging upon a Ta radiator. After a preset irradiation period the samples were removed to a low-background counting area, and gamma rays emanating from radioisotopes created during the irradiation were detected using a well-calibrated photon detection system based'on a 45-cm^3 intrinsic Ge detector. Spectra were obtained so as not only to identify peaks corresponding to specific transition, but also the effective half-life of the decay of a given transition, thus enhancing the identification of a specific gamma ray with the decay of a given radionuclide.

A schematic of the Photon Activation System is given in Figure 1. Two pulses/second of the normal electron beam are diverted by a pulsed magnet. The diverted beam strikes a Ta foil radiator producing a spectrum of photons from very low energies to more than one hundred MeV; however, the dominant interactions of this photon beam with the sample are in the giant resonance region of about 15 to 30 MeV.[5,6] The water absorbs neutrons made by the bremsstrahlung photon in the radiator foil and surrounding regions. In addition, a magnetic field perpendicular to the beam path removes electrons which pass through the radiator into the water.

Two spectra obtained during this study are shown in Fig. 2. The upper spectrum shows radionuclide decay following irradiation of a sample of Sodium Chloride, and the lower spectrum shows a similar spectrum obtained following irradiation of a sample of Sodium Carbonate.

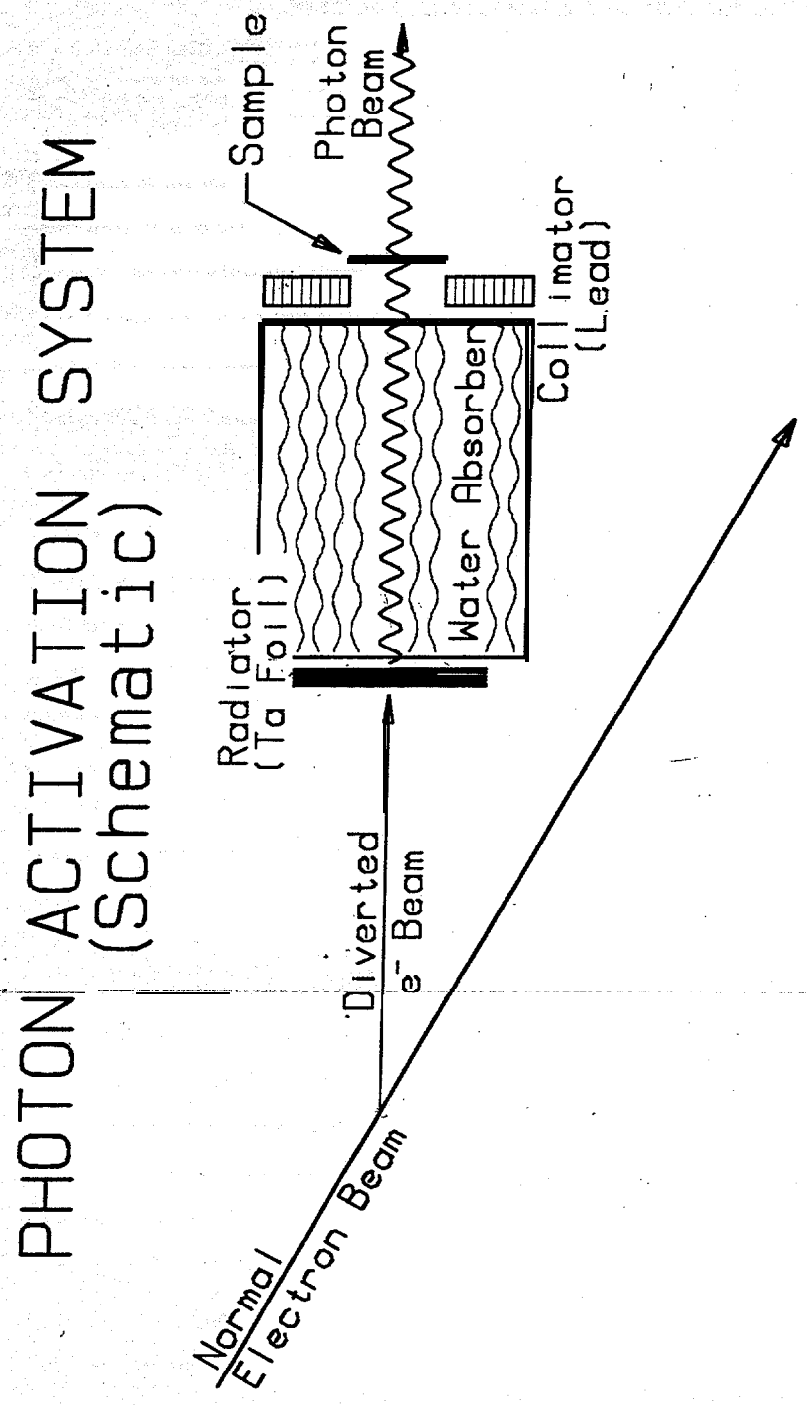
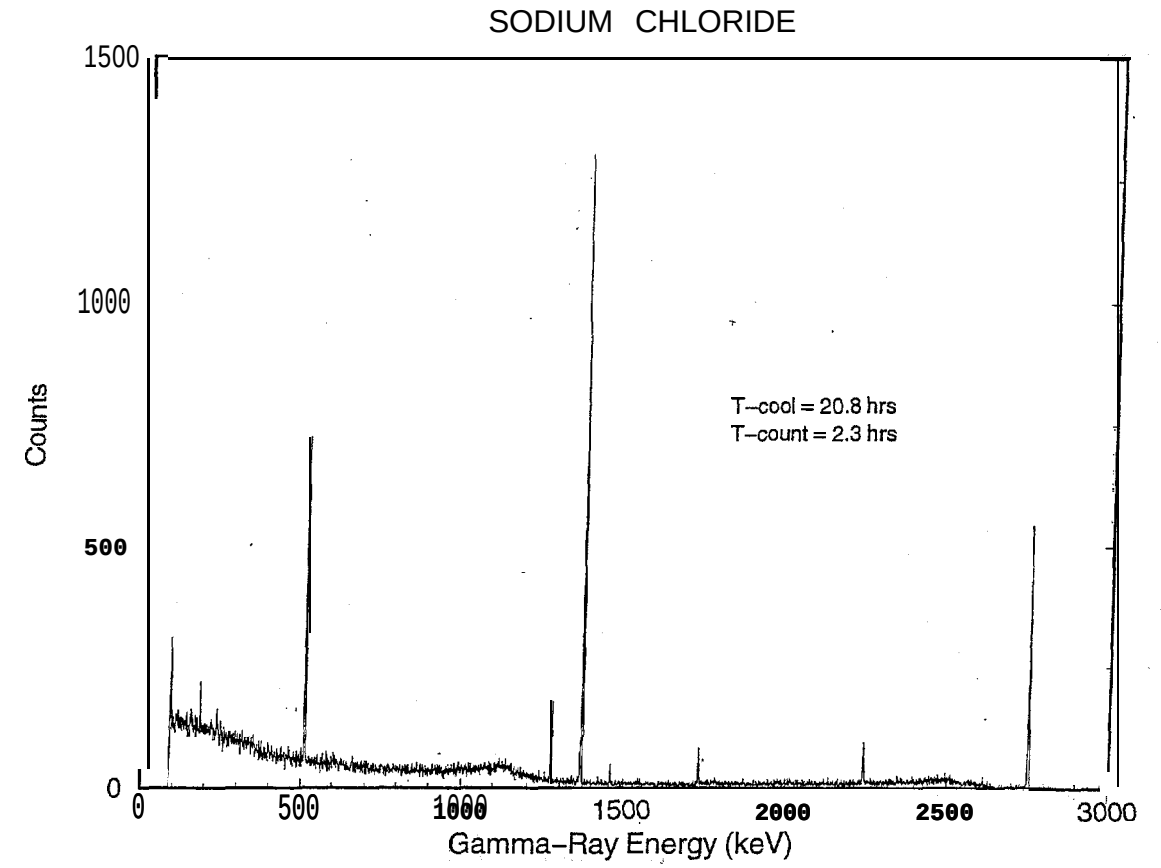


Figure 1. Schematic of the Photon Activation System, Not shown is a permanent magnet providing a magnetic field perpendicular to the photon beam.



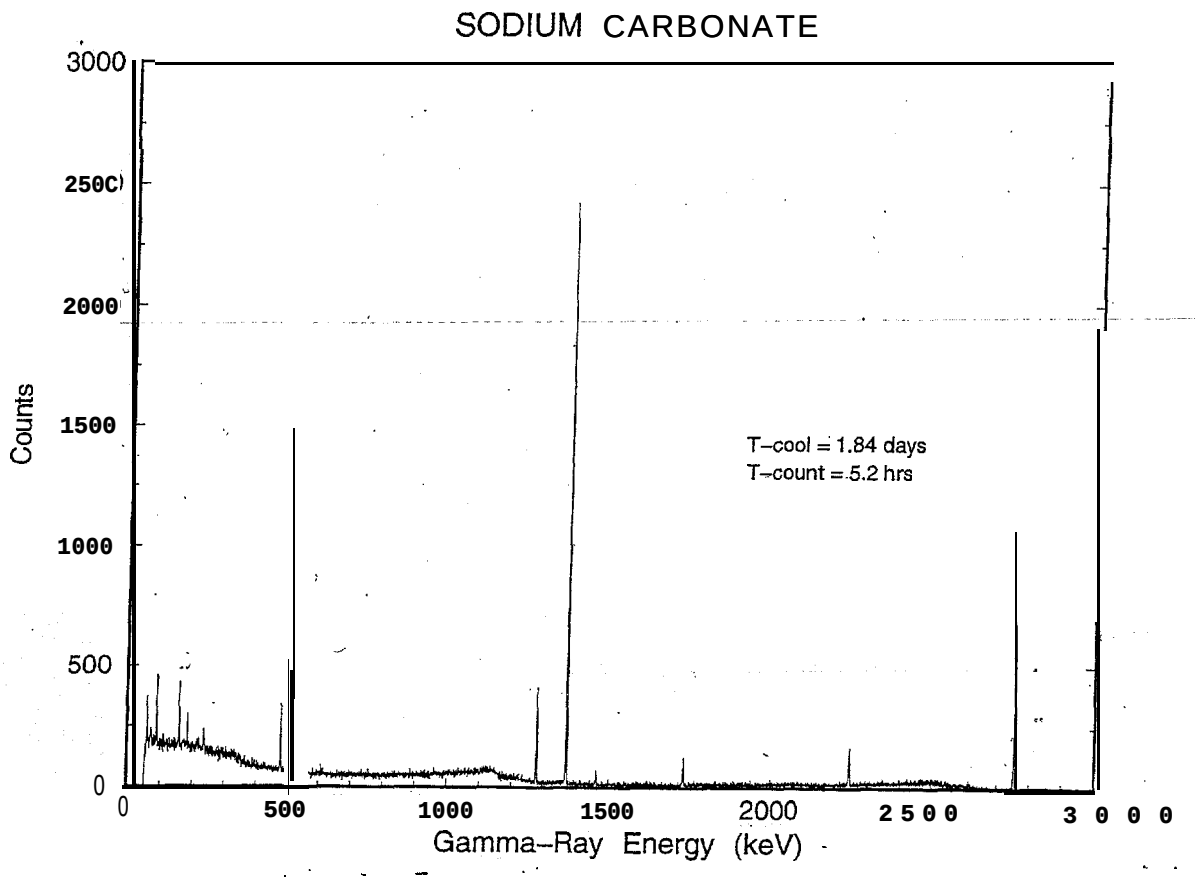


Figure 2. Spectra obtained for samples of (upper) Sodium Chloride and (lower) Sodium Carbonate.

The spectral data were reduced using TPASS,[7] a gamma-ray spectrum analysis and isotope identification computer code. The photon-peak yields, after a table look-up process to identify the responsible radioisotopes, were analyzed to deduce the overall yields of the radioisotopes. These, then, were modified by the irradiation time *vis-a-vis* the radioisotope half life to provide the production rate, R, for each radioisotope created by the specific irradiation. As there **was** no beam monitor in place for these experiments, the tabulated R values are relative. However, one can deduce interesting results from these relative values.

2.A. Sodium 'Chloride

A sample of -1.5 gm of NaCl (made up of three small packages of ordinary salt) was irradiated for 30 minutes. Initial spectra were dominated by the decay of $^{34}Cl^{isom}$; a spectrum for a cooling time of 900 sec is presented in the earlier report of Wunstorf, **et** *al.*[2] The spectrum obtained after 20.8 hours of cooling time is given in the upper part of Figure 1. Deduced isotope production rates are presented in Table 1..

Table 1. Relative Isotope Production Rates for Photonuclear Reactions with NaCl

Radio-Isotooe	Half-Life	Production Rate (atoms/sec)
$^{38}Cl^{a}$	37.2 min	1.05E3[b]
$^{34}Cl^{isom}$	32.0 min	1.19E5
^{29}Al	6.6 min	9.532
$^{24}Na^{a}$	15.0 hrs	4.2733
^{22}Na	2.6 yrs	2.56E5
^{18}F	1.8 hrs	1.3E4

[a] This isotope is due to thermal-neutron capture.
[b] Read as 1.05 x 10^3 (i.e., 1030)

2.B. Sodium Carbonate

A sample of -6 gm of Na_2CO_3 powder contained in a plastic bottle was irradiated for 30 minutes. The spectrum obtained after about 44 hours of cooling time is given in the lower part of Figure 1. In addition to peaks associated with decay of ^{22}Na and ^{24}Na, and several background peaks, there is a peak at 478 keV due to decay of ^{7}Be. In addition, for short cooling times, the annihilation radiation peak at 511 keV is very large, much larger than the same peak for equivalent cooling times in the 'spectra from the NaCl sample; analysis of its decay verifies a component having a half life of about 20 minutes. This component is assigned to the decay of ^{11}C. Deduced isotope production rates are presented in Table 2; one caveat is that the amount of carbon in the plastic bottle was not determined.

Table 2. Relative Isotope Production Rates for Photonuclear Reactions with Sodium Carbonate

Radio-Isotope	Half-Life	Production Rate (atoms/sec)
^{22}Na	2.6 yrs	5.9E5
^{11}C	20.4 min	1.4436
^{7}Be	53.3 days	8.634

2.c. Beryllium

The Beryllium sample was a machined dis-k, 0.6 mm thick by about 7.8 cm diam. However, the collimation shown in Figure 1, permitted only the central portion of the disk to be illuminated by the photon beam. The amount illuminated was -2.8 gm. The irradiation period was 15 minutes. The sample initially registered 50 mR; however, the radiation was due to impurities in the Be sample. No determination was made of the amount of impurities; however, Ahrens, et *al.*[6] reported 0.448% impurities in their Be sample, an amount probably similar to the amount in the present sample., The final gamma-ray spectrum had a cooling time of 3.08 days and a counting period of 0.69 days and is shown in the report of Wunstorf, et *al.*[2] The deduced ^{7}Be production rate was 1.3×10^{5} atoms/sec.

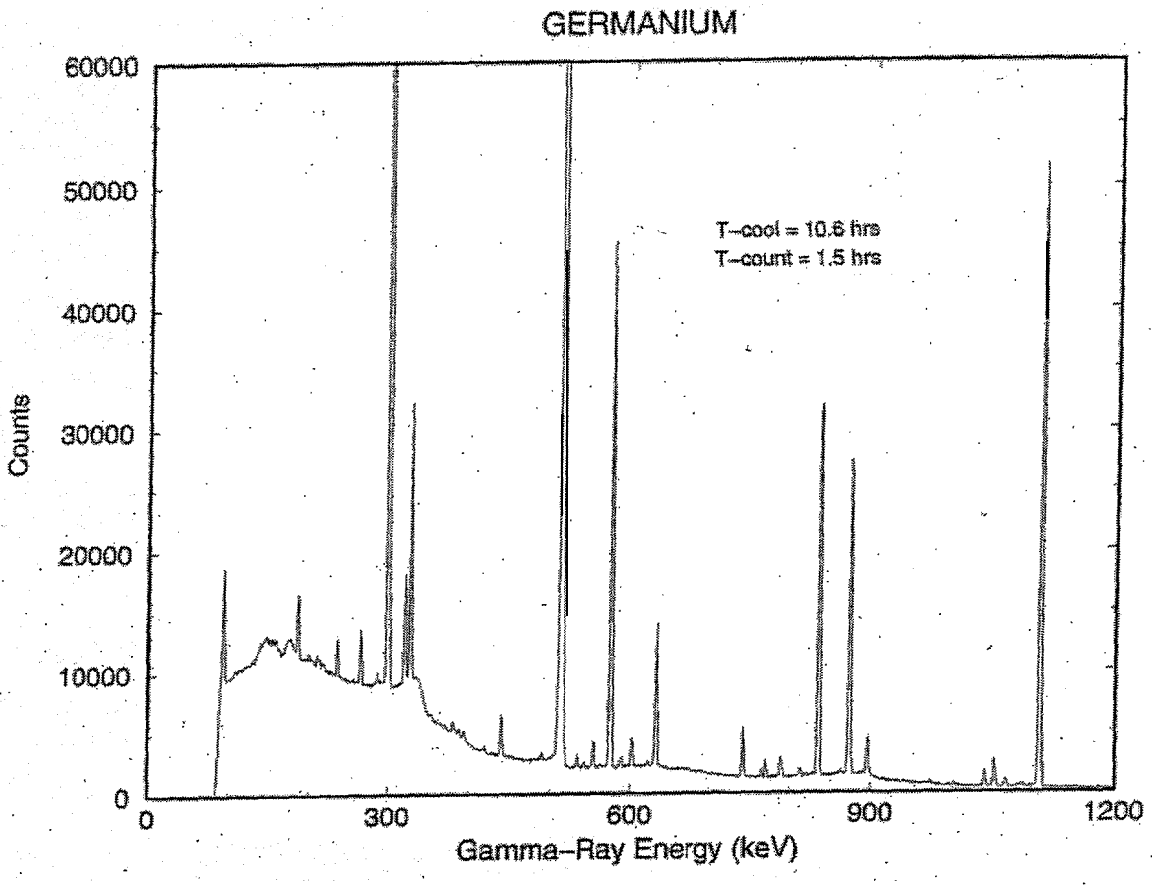
2.D. Germanium

The 11.2-gm high-purity Germanium sample was irradiated twice, the first time for 16 minutes, and the second time for five days. Most of the'isotope production rates were obtained from analysis of the first set of spectra. Figure 3 shows two portions of one spectrum; the upper part exhibits the low-energy portion and the lower part exhibits the high-energy portion. The second irradiation was analyzed for the longer-lived isotopes, primarily ^{68}Ge and ^{65}Zn. A spectrum taken following the second irradiation is shown in the report of Wunstorf, **et *al*.**[2] The deduced isotope production rates are collected in Table 3.

Table 3. Relative Isotope Production Rates for Photonuclear Reactions with Ge

Radio-Isotope	Half-Life	Production Rate (atoms/sec)
^{77}Ge[a]	11.3 hr	2.9E2
^{75}Ge	1.38 hr	6.4E5
^{69}Ge	1.63 d	1.7536
^{68}Ge	270.8 d	2.435
^{67}Ge	19.0 min	5.633
^{66}Ge	2.26 hr	2.0E3
^{74}Ga	8.1 min	6.45E3
^{73}Ga	4.87 hr	4.4E5
^{72}Ga	14.1 hr	1.2435
^{70}Ga	21.1 min	2.0E5
^{68}Ga	1.13 hr	1.4E5
^{67}Ga	3.26 d	7.1E4
^{66}Ga	9.5 hr	1.3E4
^{65}Ga	X5.2 min	1.06E3
^{64}Ga	2.63 min	3.0E2
^{72}Zn	46.5 hr	8.832
$^{71}Zn^{isom}$	3.97 hr	6.0E2
$^{69}Zn^{isom}$	13.8 hr	6.633
^{65}Zn	243.8 d	4.2534

[a] This isotope is due to thermal-neutron capture.



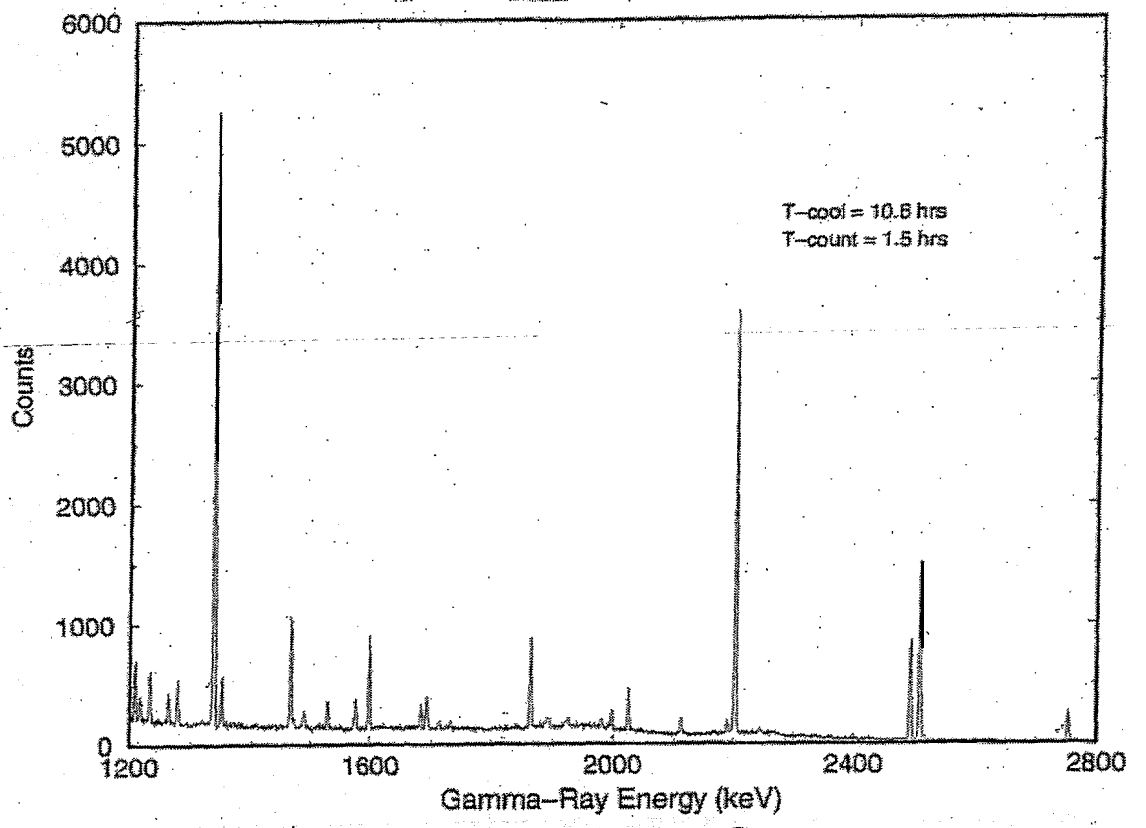


Figure 3. Spectrum obtained for Ge sample: (upper) low-energy portion and (lower) high-energy portion.

3.0 WHAT CAN BE GLEANED FROM THE DATA?

From Tables 1 and 3 evidently some thermal neutrons are not absorbed by the water, but instead interact with the sample creating radionuclides that can be produced only by neutron capture, Of interest, then, is the ratio of n(y), the number of "high-energy" photons (having energies > approximately 10 MeV), to n(n), the number of thermal neutrons impinging on the sample. From the results in Table 1, we can infer as follows:

$$\frac{256E3}{4.27E3} = \frac{^{22}Na}{^{24}Na} = \frac{\sigma(\gamma)}{\sigma(n)}\,\frac{n(\gamma)}{n(n)}$$

Using an average value of 9.5 mb for a(y) from Berman's compilation* and 530 mb for a(n) from the *Table of Isotopes*[8] results in deducing the desired ratio:

$$n(\gamma) / n(n) \approx 3350.$$

From the results for the two Cl isotopes, the average photoneutron cross section for $^{35}Cl(\gamma,n)^{34}Cl^{isom}$ can be deduced:

$$\frac{1.19E5}{1.05E3} = \frac{^{34}Cl^{isom}}{^{38}Cl} = \frac{\sigma(\gamma,n)}{\sigma(n,\gamma)}\,\frac{n(\gamma)}{n(n)}\,\frac{\%(35)}{\%(37)}$$

$$\frac{\sigma(\gamma,n)}{433\ mb}\,\frac{3350}{1}\,\frac{75.8}{24.2}$$

where the neutron capture cross section for ^{37}Cl and isotopic abundances for the stable Cl isotopes were taken from Table *of Isotopes.?* Finally, one deduces:

$$\sigma(\gamma,n) \text{ populating the } ^{34}Cl^{isom} = 4.7 \text{ mb}.$$

The data presented in Table 3 for Germanium may be manipulated in similar fashion to obtain several "average" photoneutron'cross sections.[10] The results are:

$$\sigma[^{76}Ge(\gamma,n)^{75}Ge] \approx 53 \text{ mb},$$

a value comparable to an estimated average photoneutron cross section for ^{75}As of ≈ 50 mb from Berman's compilation.[5] Also:

$$\sigma[^{70}Ge(\gamma,n)^{69}Ge] \approx 56 \text{ mb};$$

almost the same as the ^{76}Ge determination, although the ^{69}Ge yield may include a small amount from the $^{72}Ge(\gamma,3n)$ reaction..

4.0 CONCLUSIONS AND RECOMMENDATIONS

Photonuclear reactions have the potential to create proton-rich radioisotopes not easily available using other reactions. The present results, along with the earlier reports[3,4] mentioned in the Introduction, indicate the breadth of radioisotopes which can be created. The present system uses only a small fraction of the total 150-MeV electron beam energy available, limited, at least in part, by the heating of the air-cooled tantalum radiator. The present system also lacks a photon beam monitor, so the measurements are relative. Nevertheless, absolute cross-section values were determined by comparison with well-known $\sigma(n,\gamma)$ cross sections when a capture cross section was unequivocally determined and could be used as the monitor.

Other possible avenues of research using the photonuclear effect are available and should be explored. The present measurements may be useful as a guide for further studies.

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9. "Table of Isotopes," ***op cit.,*** pages'75 and 80; S. F. Mughabghab, ***et al., op cit.,*** p. 17-4.

10. The effective reference neutron-capture cross section for these two Ge results was $\sigma_c(^{76}Ge(n,\gamma)^{77}Ge)$ = (81 ± 11) mb, made up of 0.21*σ_c(isomer) + σ_c(ground-state), the former = (100 ± 10) mb and the latter = (60 ± 10) mb, taken from S. F. Mughabghab, ***et al., op cit.,*** p. 32-6. Gamma-ray branching ratios and isotopic compositions were obtained from "Table of Isotopes," ***op cit.,*** pages 342 and 395.

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