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Short-Lived Nuclides Formed by (γ, n) Reactions with 20 MeV Bremsstrahlung

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The most prominent (γ, n) reaction products with half-lives from 10 sec to about 1 min formed by irradiation of some 7 elements with 20 MeV bremsstrahlung have been determined in order to study the applicability of these products to photoactivation analyses. Irradiation was carried out for 1 min with 20 MeV bremsstrahlung radiation with a dose rate of 10^7 R/min. The reactions investigated are $^{24}Mg(\gamma, n)^{23}Mg$, $^{76}Ge(\gamma, n)^{75m}Ge$, $^{78}Se(\gamma, n)^{77m}Se$, $^{92}Mo(\gamma, n)^{91m}Mo$, $^{140}Ce(\gamma, n)^{139m}Ce$, $^{142}Nd(\gamma, n)^{141m}Nd$ and $^{159}Tb(\gamma, n)^{158m}Tb$. The yields of these reactions have also been obtained through the absolute measurement of resultant activities. The isomeric yield ratios have been discussed on the basis of the nuclear properties of the residual nuclides. The observed ratios generally favor the isomer whose spin is closer to that of the target nucleus. For the elements studied, the lower limits of detection averaged about 1 μg under present experimental conditions.

As pointed out in previous reports,[1] the use of high-energy gamma-rays in activation analysis sometimes offers several advantages over the neutron activation method with respect to selectivity and specificity. A number of radioactive species with moderate half-lives (longer than 2 min) arising from photonuclear processes on many elements irradiated with 20 MeV bremsstrahlung have been investigated with respect to yield, and the applicability to activation analysis of those elements.[2]

On the other hand, short-lived nuclides can be obtained selectively with a short-irradiation period, suppressing longer-lived components in the gamma-ray spectra of the activated specimen, and can thereby be used as a rapid, sensitive and non-destructive method of analysis of the relevant element. Until recently, these problems have been investigated exclusively by means of thermal neutron activation, and a number of tabulations or catalogs on gamma-ray spectra of (n, γ) reaction products have been compiled by many workers.[3-8]

The purpose of the present report is to determine gamma-ray spectrometrically most important short-lived (γ, n) reaction products with half-lives from 10 sec to about 1 min, formed in the irradiation of some 7 elements with 20 MeV bremsstrahlung, and to discuss the reaction yields on the basis of the nuclear properties of those nuclides.

Experimental

Samples and Irradiation. Irradiation was carried out with bremsstrahlung radiattion, converted from a 20 MeV electron beam[9] generated from the linear electron accelerator of the Japan Atomic Energy Research Institute. The chemical forms of the sample materials used in these experiments were the elements of magnesium, germanium, selenium and molybdenum, and the oxides of cerium, neodymium and terbium. A definite amount of each sample (5—40 mg) was wrapped in thin aluminum foil and made into a small disk of diameter 5 mm and with a thickness of 1 mm.[10] The bremsstrahlung flux was monitored using a thin sheet of gold (10 μ thick, 5 mm dia.) adjacent to the samples. Each sample with gold monitor was encapsulated in a cylindrical aluminum rabbit[11] together with polyethylene foam to hold the sample tightly. The sample in the rabbit was transferred to the irradiation point immediately behind the photon-producing converter[12] by a pneumatic tube. All irradiations were for one minutes.

The dose rate of the bremsstrahlung photons at the target position was determined by the method of activating a manganese specimen as reported previously.[13] It was estimated at 10^7R/min. Following irradiation, the rabbit was brought back by a reversed air stream,

1) Y. Oka, T. Kato and M. Sasaki, *Nippon Kagaku Zasshi (J. Chem. Soc. Japan, Pure Chem. Sect.)*, **84**, 588 (1963), and other reports.
2) Y. Oka, T. Kato, K. Nomura and T. Saito, *J. Nucl. Sci. Technol.*, **4**, 364 (1967).
3) M. Okada, *Nucleonics*, **22**, No. 8, 110 (1964).
4) M. Okada, This Bulletin, **39**, 1340 (1966).
5) M. Okada, *Nucleonics*, **19**, No. 9, 79 (1961).
6) M. Okada, *ibid.*, **20**, No. 12, 61 (1962).
7) Y. Kamemoto, K. Shiba, H. Handa and M. Okada, JAERI-4019 (1961).
8) O. U. Anders, NP-14469 (1964).
9) Average current: 40 μA.
10) This was aimed at producing an optimum specific activity in the irradiation in a collimated bremsstrahlung beam.
11) 10 mm i. d., 40 mm long.
12) 2 mm thick platinum plate.
13) Y. Oka, T. Kato and I. Nagai, *J. Nucl. Sci. Technol.*, **4**, 300 (1967).

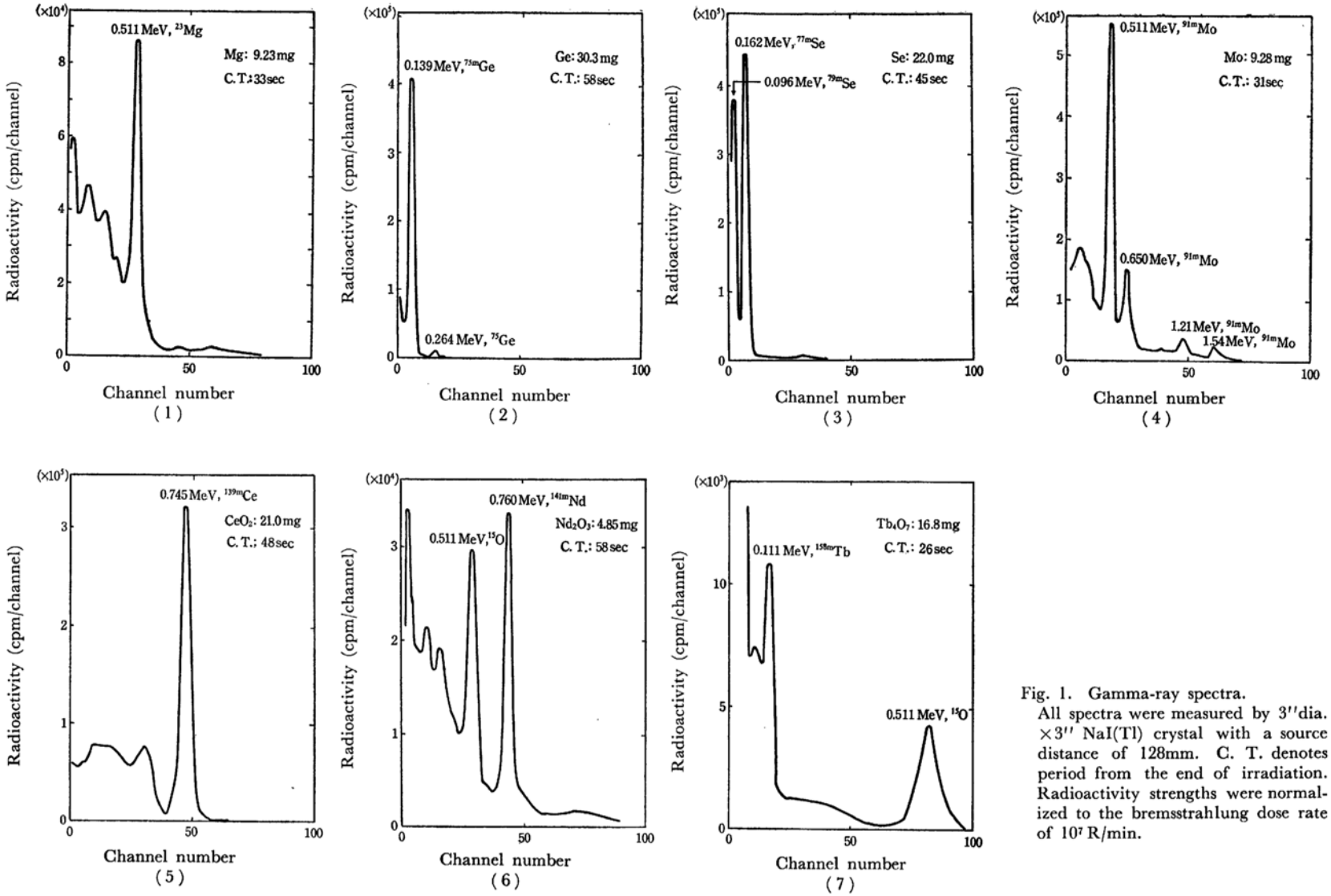

Fig. 1. Gamma-ray spectra.
All spectra were measured by 3″dia. ×3″ NaI(Tl) crystal with a source distance of 128mm. C. T. denotes period from the end of irradiation. Radioactivity strengths were normalized to the bremsstrahlung dose rate of 10^7 R/min.

and the sample disk removed. The travel time of the rabbit was 2 sec.

Radioactivity Measurement. Immediately after the removal of the sample from the rabbit, its gamma-ray spectrum was measured with a 3'' dia. ×3'' NaI(Tl) detector coupled to a TMC 400-channel pulse-height analyzer. The memory of the analyzer was subgrouped into 4×100 channels, and a spectrum collected in each subgroup. This method was most efficient for activities with the half-life range from 10 sec to about 1 min.

Data Treatment. The radioactivity of the desired product was evaluated for the spectrum from the gamma-ray photopeak area by drawing a straight line across the bottom of the photopeak and summing up the counts above this line. These data were corrected both for decay during the time of each count and for the dead time of the analyzer. Further normalizations were applied to the activity data for the decay time, the dose rate, sample weight and counting geometry of the detector. Decay curve analyses were performed to separate out the undesired activities.

Nuclide identification was obtained from a knowledge of the target nuclide, the gamma-ray spectra, decay data, and data in the literature for the nuclides.[14,15]

The yield was measured for each (γ, n) reaction through the absolute determination of the resultant radioactivity. To obtain the disintegration rate from the counting rate of the relevant gamma-ray photopeak, appropriate corrections were made for the variation of photopeak efficiency with gamma-ray energy. The decay schemes used were those listed in the Nuclear Data Sheets.[14] The table of internal conversion coefficients[16] was also used.

Results

Induced Radioactivity. As reported earlier,[2] the prominent photoactivation products found in the energy region of 20 MeV bremsstrahlung are those from (γ, n) and (γ, p) reactions. A preliminary experimental investigation of the short-lived photoactivation properties of some 18 elements have indicated the usefulness of the present method for the determination of magnesium, germanium, selenium, molybdenum, cerium, neodymium and terbium.

With these elements, it was possible to let the activities of short-lived products be much larger than those of longer ones produced from irradiation for one minute.[17] The gamma-ray spectra obtained from 7 elements are given in Fig. 1 (1)—(7). Seven nuclides including ^{23}Mg, ^{75m}Ge, ^{77m}Se, ^{91m}Mo, ^{139m}Ce, ^{141m}Nd and ^{158m}Tb were found to be the most prominent species.

Table 1 lists particulars on the products and the data obtained in this experiment. Of the present results, the half-life of ^{23}Mg was determined as 9.9 sec, which is shorter than that listed in the Nuclear Data Sheets (12 sec).[14]

Limit of Detection. The limit of detection was calculated on the basis of the minimum detectable photopeak area. All activity strengths have been expressed in counts per milligram of the element at the end of one-minute irradiation with a counting geometry of 20% using a 3''dia.×3'' NaI(Tl) detector, and have been normalized to a bremsstrahlung dose rate of 10^7 R/min. Because of the short-lives of the products to be determined, the limits of detection were assumed to be the quantities needed to give 1000 cpm under the above condition. These limits of detection are given in the seventh column of Table 1. It is seen that, on the average, these elements can be detected easily down to about 1 μg per sample by the proposed method.

TABLE 1. THE PARTICULARS OF THE (γ,n) REACTION PRODUCTS AND THE DATA OBTAINED WITH 20 MeV BREMSSTRAHLUNG

Nuclide		Half-life of product (sec)	Gamma-ray determined			Limit of detection (μg)	Yield ($mol^{-1}\cdot R^{-1}$)
Parent (Natural abundance, %)	Residual		Energy (MeV)	Branching ratio (%)	Photopeak activity (cpm/mg)[a]		
^{24}Mg(78.60)	^{23}Mg	9.9	0.511	200	2.04×10^6	0.49	8.1×10^4
^{76}Ge(7.67)	^{75m}Ge	48	0.139	100	6.37×10^5	1.6	1.1×10^4
^{78}Se(23.52)	^{77m}Se	17	0.162	100	1.82×10^6	0.55	1.2×10^6
^{92}Mo(15.86)	^{91m}Mo	65	0.650	57	2.22×10^5	4.5	2.7×10^6
^{140}Ce(88.48)	^{139m}Ce	58	0.745	100	1.06×10^6	0.95	1.3×10^6
^{142}Nd(27.13)	^{141m}Nd	64	0.760	100	3.19×10^5	3.1	1.4×10^6
^{159}Tb(100)	^{158m}Tb	11	0.111	100	2.56×10^5	3.8	2.2×10^6

a) The value corrected at the end of one-minute irradiation with the dose rate of 10^7 R/min; Counting geometry is 20% with a 3''dia.×3''NaI(Tl) detector.

14) Nuclear Data Sheets, Natl. Academy of Science-Natl. Research Council, Washington, D. C. (1961).
15) B. S. Dzhelepov and L. K. Peker, "Decay Schemes of Radioactive Nuclei," Pergamon Press., New York (1961).
16) M. E. Rose, "Internal Conversion Coefficient," North-Holland Publ. Co., Amsterdam (1958).
17) The extraneous activities were due to the reactions, ^{27}Al$(\gamma, n)^{26m}$Al (6.6 sec, β^+ emitter) and ^{16}O$(\gamma, n)^{15}$O (124 sec, β^+ emitter).

Reaction Yield. The yield ($Y_{(E)}$) was obtained by the absolute measurement of the resultant activity through the following equation.

$$Y_{(E)} = \frac{D_o}{IM(1-e^{-\lambda t})}$$

where D_o is the disintegration rate at the end of irradiation, I the radiation dose rate, M the amount of target nucleus expressed in moles, and λ the decay constant. The data obtained are shown in the last column of Table 1. These values can be taken as the numbers of (γ, n) processes which occurred during the irradiation.

Discussion

It has been pointed out that the photon absorption cross section for a nucleus is an increasing function of Z.[18] In our measurements, through the residual activity method,[19] the yield of radioactive nuclides increases regularly with atomic number from 10^3 to 10^7 orders of magnitude. Of the yields given in Table 1, that for the reaction $^{24}Mg(\gamma, n)^{23}Mg$ is in excellent agreement in absolute magnitude with those expected from the yield curve above. However, the yields for other 6 nuclides, of metastable states, were obtained as $(1—3)\times 10^6$/mol·R, and do not vary significantly with Z values. This implies that the relative yield for the formation of a residual nuclide in its metastable state to that in the ground state should fall for heavier nuclei.

The relative probability of forming each member of a pair of nuclear isomeric states as a result of photonuclear process in the energy region above a giant resonance has been studied by several workers in order to obtain quantitative information about the spin distribution of the excited states.[20-28]

Table 2 shows the results of the isomeric yield ratios obtained for 5 isomeric pairs together with the spins of both the parent nuclei and the residual isomeric states.[29] The isomeric yield ratio has been expressed as a ratio of the yield for the metastable state to the sum of those for both states. Literature values are also included in Table 2.[30]

It has been noted that the formation of both isomeric states follows the steps: formation of the compound state upon electric dipole photon absorption; neutron decay of the compound states to form excited states of the residual nucleus; and gamma-ray cascades from these excited states to the two isomeric levels. In many cases in which an excited state of the residual nucleus can decay to either of the isomers, through intermediate states in gamma-cascades, the gamma-transitions of low multipolarity are accorded preference over those of high multipolarity. Thus the spins of each state of the isomers, as well as the spin differences between states, are the most important factors determining the isomeric yield ratio. Since the absorption of 20 MeV bremsstrahlung by a nucleus can be regarded as $E1$ absorption, the spins of the excited states may not differ remarkably from that of the parent nucleus. The observed ratios generally favor the isomer whose spin is closer to that of

Table 2. The yield ratio of some isomeric pairs

Parent		Residual				Type of multipole transition	Isomeric yield ratio, $Y_m/(Y_m+Y_g)$	
		Ground state		Metastable state				
Target nucleus	Spin	Spin	Half-life	Spin	Half-life		This work	Literature value
^{45}Sc	$7/2^-$	^{44}Sc 2^+	3.9 hr	6^+	2.4 d	$E4$	0.084	0.16[28] a)
^{76}Ge	0^+	^{75}Ge $1/2^-$	82 min	$7/2^+$	48 sec	$E3$	0.38	0.48[26] b)
^{85}Rb	$5/2^-$	^{84}Rb 2^-	33 d	6^+	20 min	$M4$	0.24	—
^{90}Zr	0^+	^{89}Zr $9/2^+$	79 hr	$1/2^-$	4.3 min	$M4$	0.33	0.49[26] b)
^{140}Ce	0^+	^{139}Ce $3/2^+$	140 d	$11/2^-$	58 sec	$M4$	0.091	0.087[26] b)

a) The value obtained with 24 MeV bremsstrahlung.
b) The value obtained with 30 MeV bremsstrahlung.

18) R. Montalbetti, L. Katz and J. Goldenberg, *Phys. Rev.*, **91**, 659 (1953).
19) Y. Oka, T. Kato, K. Nomura and T. Saito, This Bulletin, **40**, 575 (1967).
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21) L. Katz, L. Pease and H. Moody, *Can. J. Phys.*, **30**, 476 (1952).
22) L. Katz, R. G. Baker and R. Montalbetti, *ibid.*, **31**, 250 (1953).
23) J. Goldenberg and L. Katz, *ibid.*, **90**, 308 (1953).
24) P. Axel and J. D. Fox, *ibid.*, **102**, 400 (1956).
25) J. H. Caver, G. E. Coote and T. R. Sherwood, *Nucl. Phys.*, **37**, 449 (1962).
26) S. Costa, F. Ferrero, S. Ferroni and L. Pasqualini, *ibid.*, **72**, 158 (1965).
27) J. R. Tatarczuk and H. A. Medicus, *Phys. Rev.*, **143**, 818 (1966).
28) W. B. Walters and J. P. Hummel, *ibid.*, **150**, 867 (1966).
29) The yields for the $^{45}Sc(\gamma, n)$, $^{86}Rb(\gamma, n)$ and $^{90}Zr(\gamma, n)$ reactions are taken from data reported previously.[19]
30) Although experiments were carried out with different excitation energies, there are no significant variations in the observed isomeric yield ratios with change in the bremsstrahlung maximum energy above a giant resonance region.[26]

the target nucleus. The yield of isomers with very high spins, including ^{44m}Sc, ^{75m}Ge, ^{84m}Rb and ^{139m}Ce, as well as ^{77m}Se, ^{141m}Nd and ^{158m}Tb, are found to be low. The result for ^{89}Zr isomers is unusual, and it is planned to investigate the reasons responsible for this behavior in the near future.

For analytical purposes, the lower limit of detection under the present experimental conditions is 1 μg on the average. As discussed above, the yield of short-lived metastable isomers is not as high as expected from the general behavior of the (γ, n) reaction yield curve. In addition, high multipolarity gamma-transitions with large internal conversion coefficients are inherent in the decay of these isomers. Despite these unfavorable properties, the use of short-lived activities in photoactivation analysis should enhance the efficacy of the method because of rapidity, selectivity and specificity.