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The Yields of Radioactivities Induced by (γ, n) Reactions with 20 MeV Bremsstrahlung

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The yields of radioactivities resulting from the (γ, n) reactions on 37 elements covering the periodic table have been determined by means of 20 MeV linear accelerator bremsstrahlung and gamma-ray spectrometry. The yield values, as measured in the number of photodisintegrations per mole per roentgen for each of the (γ, n) reactions, increase in the main regularly with atomic number from 10^3 through 10^7 order of magnitude. Anomalously small yield values are obtained in the irradiation of the nuclides with neutron or proton number of the magic numbers. The present results permit studies of general behavior of the photoneutron reactions and of the application of their products to the photoactivation analyses of many elements distributed throughout the periodic table.

In the previous paper,[1] the gamma-ray spectrometric study has been carried out on the photoactivation products of 75 elements from lithium through bismuth irradiated with 20 MeV bremsstrahlung, and the applicability of these products to the photoactivation analysis has been discussed from these data. On the other hand, the neutron yields from many elements irradiated with bremsstrahlung radiation of moderate energy have been determined by several investigators through the direct detection of the emitted photoneutrons.[2,3] Though the neutron detection method offers several advantages over the residual activity method in observing precise photon-absorption mechanism, it only gives the sum of the yields of the (γ, n), (γ, 2n), (γ, pn) and other neutron emitting reactions, or the total integrated cross section for photon-absorption processes: It can be said that some ambiguities should be present in applying these yield data on the general consideration of photoactivation analysis utilizing particular residual nuclides generated from each of the photonuclear reactions.

In the present work, consequently, a study has been carried out on the yields of the (γ, n) reactions of some 37 elements distributed throughout the periodic table using a 20 MeV accelerator bremsstrahlung and the residual activity method based on the gamma-ray spectrometry.

Experimental

Target Material and Irradiation. Definite amounts of target materials with high chemical purity were irradiated with bremsstrahlung converted from a 20 MeV electron beam (average beam current: 40 μA) generated from the linear electron accelerator of Japan Atomic Energy Research Institute.

The chemical forms of the 37 target materials were as follows: The elements of carbon, chromium, manganese, copper, zinc, germanium, arsenic, selenium, cadmium, tin, tellurium, tungsten, osmium, gold, thallium and lead: the oxides of scandium, iron, cobalt, nickel, yttrium, zirconium, niobium, cerium, thulium, ytterbium and tantalum: the carbonates of sodium and strontium: the nitrate of rubidium: fluorine and chlorine as their lithium salts: iodine as its potassium salt: potassium as sulfate: caesium as chloride: nitrogen as ammonium nitrate: oxygen as boric acid.

The individual target materials were either sealed in a small quartz tube having an internal diameter of 4 mm or holded in a thin aluminum foil and were placed in a water cooled target holder immediately

1) Y. Oka, T. Kato, K. Nomura and T. Saito, "Gamma-ray Spectrometric Study of the Photoactivation Products with 20 MeV Bremsstrahlung," To be published in *J. Nucl. Sci. Technol.*: Presented at the 19th Annual Meeting of the Chemical Society of Japan (April 1966).

2) G. A. Price and D. W. Kerst, *Phys. Rev.*, **77**, 806 (1950).

3) R. Montalbetti, L. Katz and J. Goldenberg, *ibid.*, **91**, 659 (1953).

behind the photon-producing converter.[4] Care was taken to locate the target samples within a constant irradiation condition. The typical sample size had a weight of 50 to 100 mg and a thickness facing the accelerator of 4 mm: This was aimed at producing an optimum specific activity in a standard irradiation in the forward bremsstrahlung beam. Irradiations were generally performed for one hour for each of the samples. In cases where the half-lives of the desired products were relatively short, the irradiations were terminated in shortening periods (1—10 min).

Dose Rate and Monitoring. The dose rate of the bremsstrahlung radiation in this irradiation condition was estimated by the detection of the residual activity of the (γ, n) reaction in manganese, and was determined as 9.4×10^6 roentgens per min. Additional monitor, consisting of a thin sheet of gold, was used to determine the relative intensity of the bremsstrahlung dose to which each of the samples were irradiated: These experimental manners were essentially the same as that described in detail in the previous reports.[1,5]

Radioactivity Measurement. Following irradiation, the samples were cooled for a time, allowing the foreign activities to decay away.[6] The gamma-ray spectra were then measured at a suitable counting geometry by using a $3''\phi\times3''$ NaI (Tl) detector coupled with a TMC 400 channel pulse height analyzer. The counting rate due to the desired gamma-ray was measured in the area under a full energy peak in the spectrum: This was obtained by drawing a straight line across the bottom of the photopeak and by calculating its area above the line. Decay curve analyses of the counting data served to separate the contributions of the various radioactive products. Nuclide identification was obtained from a knowledge of the target nuclides, the gamma-ray spectra, decay data, and literature data for the nuclides.[7-9]

No chemical separations have been performed in this experiment.

Absolute Determination. The disintegration rate was determined by absolute calculation from the observed counting rate of the area under a full energy peak in the spectrum with the aid of the decay scheme parameter for a particular nuclide and the efficiency factor of the counting arrangement by the following equation.

$$D=\frac{C}{R_{(E)}\Omega\varepsilon_{(E)}f_bf_c}$$

where D: disintegration rate (dpm); C: counting rate (cpm); $R_{(E)}$: the ratio of the area of the full energy photopeak to the area of the total spectrum of a photon energy (E)[10]; Ω: geometry; $\varepsilon_{(E)}$: total intrinsic efficiency of the detector with a photon energy (E)[11]; f_b: branching ratio factor[8,9]; and f_c: total conversion ratio factor, $1/(1+\alpha_K+\alpha_L+\cdots)$.[12]

The disintegration rates determined were generally corrected to that at a time of the end of the irradiation.

Yield Calculation. Yields were then given for each of the reactions as calculated from the data of absolute determinations by the following equation.

$$Y_{(E)}=\frac{D_0}{IM(1-e^{-\lambda t})}$$

where $Y_{(E)}$ is the yield; D_0 is the disintegration rate at the end of the irradiation; I is the radiation dose rate; M is the amount of the target nuclide expressed in the number of moles; λ is the decay constant; and t is the irradiation period. The value $(1-e^{-\lambda t})$ can be replaced by λ when the half-life of the product is enough long to the irradiation period. (In this case, I is the total radiation dose.)

Results and Discussion

The (γ, n) Reaction Yield. Of the nuclides identified as the products of the (γ, n) reactions on 37 elements studied, 43 were described for their nuclear characteristics and the measured yields for the particular (γ, n) reactions: These are shown in Table 1.

Columns one and two give the parent and the residual nuclides, and column three shows the neutron separation energy (S_n), the energy required to separate one neutron from a parent nucleus: This is a value which has been estimated by Yamada and Matsumoto[13] for most of the nuclei by the use of mass formulas and nuclear systematics. Columns four, five and six describe the properties of gamma-ray used to absolute determination, the energy, the branching ratio and the type of multipole transition,[8,9] respectively. The last two columns show the results of the yield determinations. A specific activity was measured as microcuries per milligram of the element and was corrected to the following conditions; the dose rate was 9.4×10^6 roentgens per min and the irradiation period was 1 hr. The yield value was expressed as the disintegration rate at the end of the irradiation per mole of target nuclide per roentgen. This can be quoted as the number of photodisintegrations occurred during the irradiation.

4) 2 mm thick platinum plate.

5) Y. Oka, T. Kato and I. Nagai, "Photoactivation of Rare Earth Elements with 20 MeV Bremsstrahlung," To be published *J. Nucl. Sci. Technol.*: Presented at the 9th Symposium on Radiochemistry in Japan (Oct. 1965).

6) ^{15}O(124 sec), ^{28}Al(2.27 min) and ^{29}Al(6.56 min) were produced from the elements in a quartz tube by O(γ, n) and Si(γ, p) processes. When a desired product was shorter-lived, a quartz tube was destroyed and its content was transferred to a "cold" glass tube for gamma-counting.

7) D. Strominger, J. M. Hollander and G. T. Seaborg, *Rev. Mod. Phys.*, **30**, No 2. Part 2 (1958).

8) Nuclear Data Sheets, Natl. Academy of Science-Natl. Research Council, Washington (1961).

9) B. S. Dzhelepov and L. K. Peker, "Decay Schemes of Radioactive Nuclei," Pergamon Press., New York (1961).

10) The relation between $R_{(E)}$ and E have been determined with the aid of gamma-rays from ^{57}Co, ^{51}Cr, ^{22}Na, ^{137}Cs, ^{54}Mn, ^{65}Zn, ^{88}Y and other nuclides.

11) S. Flügge Ed., "Handbuch der Physik, Bd. XLV. Instrumentelle Hilfsmittel der Kernphysik II," Springer-Verlag, Berlin (1958), p. 113.

12) M. E. Rose, "Internal Conversion Coefficient," North-Holland Publ. Co., Amsterdam (1958).

13) M. Yamada and Z. Matsumoto, *J. Phys. Soc. Japan*, **16**, 1497 (1961).

TABLE 1. SUMMARY OF DATA ON (γ, n) REACTIONS WITH 20 MeV BREMSSTRAHLUNG

Nuclide		S_n MeV	Observed gamma-ray			Yield determined	
Parent (Natural abundance, %)	Residual (Half-life)		Energy MeV	Branching ratio %	Type of multipole transition	μC_i/mg[a]	Yield/mol·R
^{12}C(98.892)	^{11}C(20.4 min)	18.72	0.511	200	(Annihilation)	1.0	3.4×10^{3}
^{14}N(99.635)	^{13}N(10.05 min)	10.55	0.511	200	(Annihilation)	2.5	8.5×10^{3}
^{16}O(99.759)	^{15}O(2.02 min)	15.67	0.511	200	(Annihilation)	2.6	9.7×10^{3}
^{19}F(100)	^{18}F(110 min)	10.44	0.511	194	(Annihilation)	13	1.7×10^{5}
^{23}Na(100)	^{22}Na(2.6 yr)	12.41	1.27	100	$E2$	3.7×10^{-4}	5.9×10^{4}
^{35}Cl(75.53)	^{34m}Cl(32.4 min)	12.72	3.3	16.6	$E2$	6.7	9.1×10^{4}
^{39}K(93.08)	^{38}K(7.7 min)	13.08	2.16	100	$E2$	8.4	7.7×10^{4}
^{45}Sc(100)	^{44m}Sc(2.44 d)	11.59	0.271	99.2	$E4$	0.14	1.2×10^{5}
^{45}Sc(100)	^{44}Sc(3.92 hr)	11.32	1.16	99.6	$E2$	21	1.3×10^{6}
^{52}Cr(83.76)	^{51}Cr(28 d)	12.05	0.325	9	$M1+E2$	0.016	2.2×10^{5}
^{55}Mn(100)	^{54}Mn(314 d)	10.22	0.840	100	$E2$	0.015	2.0×10^{6}[b]
^{54}Fe(5.84)	^{53}Fe(8.9 min)	13.62	0.383	44.4	$M1+E2$	1.7	3.8×10^{5}
^{59}Co(100)	^{58}Co(71 d)	10.46	0.805	99.5	$E2$	0.063	2.2×10^{6}
^{58}Ni(67.76)	^{57}Ni(37 hr)	12.19	1.37	86	$E2$	1.3	1.4×10^{6}
^{65}Cu(30.9)	^{64}Cu(13 hr)	9.91	1.34	0.6	$E2$	5.1	4.8×10^{6}
^{66}Zn(27.81)	^{65}Zn(245 d)	11.04	1.12	49	$E2$	8.7×10^{-4}	4.2×10^{6}
^{76}Ge(7.67)	^{75}Ge(82 min)	9.59	0.265	11.06	$M1+E2$	3.1	1.8×10^{6}
^{75}As(100)	^{74}As(18 d)	10.25	0.596	61.1	$E2$	0.25	2.8×10^{6}
^{76}Se(9.02)	^{75}Se(120 d)	11.14	0.265	56	$M1+E2$	7.3×10^{-3}	5.4×10^{6}
^{82}Se(9.19)	^{81m}Se(52 min)	9.44	0.103	100	$E3$	2.3	7.9×10^{5}
^{85}Rb(72.15)	^{84m}Rb(20 min)	11.00	0.465	35	$M4$	57	1.8×10^{6}
^{85}Rb(72.15)	^{84}Rb(33 d)	10.50	0.880	73.4	$E2$	0.19	5.9×10^{6}
^{86}Sr(9.86)	^{85}Sr(64 d)	11.51	0.514	100	$M2$	0.020	8.9×10^{6}
^{89}Y(100)	^{88}Y(105 d)	11.43	1.85	99	$E2$	0.046	3.6×10^{6}
^{90}Zr(51.46)	^{89m}Zr(4.3 min)	12.53	0.588	93	$M4$	155	6.4×10^{6}
^{90}Zr(51.46)	^{89}Zr(79 hr)	11.94	0.915	100	$M4$	2.9	1.3×10^{7}
^{93}Nb(100)	^{92}Nb(10 d)	8.86	0.93	98	$E2$	0.36	2.8×10^{6}
^{116}Cd(7.58)	^{115}Cd(2.3 d)	8.75	0.335	94.5	$M4$	0.27	7.7×10^{6}
^{114}Sn(0.65)	^{113}Sn(119 d)	10.30	0.393	100	$M4$	3.1×10^{-4}	5.3×10^{6}
^{124}Sn(5.98)	^{123}Sn(41 min)	8.45	0.160	100	$M1+E2$	3.4	2.6×10^{6}
^{122}Te(2.46)	^{121}Te(17 d)	9.76	0.575	81.7	$E2$	6.6×10^{-3}	4.1×10^{6}
^{127}I(100)	^{126}I(13 d)	9.14	0.386	34	$E2$	0.87	1.2×10^{7}
^{133}Cs(100)	^{132}Cs(6.5 d)	9.02	0.673	99	$E2$	1.4	9.8×10^{6}
^{140}Ce(88.48)	^{139m}Ce(58 sec)	9.98	0.745	100	$M4$	35	1.3×10^{6}
^{140}Ce(88.48)	^{139}Ce(140 d)	9.23	0.166	100	$M1$	0.72	1.3×10^{7}
^{148}Nd(5.72)	^{147}Nd(11 d)	7.36	0.532	20	$M1+E2$	0.050	1.2×10^{7}
^{169}Tm(100)	^{168}Tm(85 d)	8.06	0.198	58	$E1$	0.12	1.4×10^{7}
^{168}Yb(0.140)	^{167}Yb(18 min)	9.11	0.176	14	$E1$	0.38	1.2×10^{7}
^{180}W(0.135)	^{179m}W(5.0 min)	8.67	0.222	100	$M3$	0.49	1.5×10^{7}
^{186}Os(1.59)	^{185}Os(94 d)	8.31	0.646	80	$E2$	2.9×10^{-3}	1.5×10^{7}
^{197}Au(100)	^{196}Au(6.18 d)	8.07	0.426	6	$E2$	2.8	2.9×10^{7}
^{204}Hg(6.85)	^{203}Hg(46.9 d)	7.52	0.279	100	$M1+E2$	0.034	3.9×10^{7}
^{203}Tl(29.50)	^{202}Tl(12 d)	7.87	0.439	37	$E2$	0.68	4.1×10^{7}

a) The value corrected at the end of one-hour irradiation (9.4×10^{6} R/min).
b) The value which had been estimated by Montalbetti *et al.*[3]

The yield values were also plotted, being marked with the residual nuclides, against atomic number of the nuclides in a graphic form in Fig. 1.

The values increase in the main regularly with atomic number from 10^3 through 10^7 order of magnitude. This behavior is analogous to that of the yields of photoneutrons which had been obtained as a function of Z values by Price *et al.*[2] and by Montalbetti *et al.*[3] This can also be well appreciated from Levinger-Bethe's theory,[14] one of the fundamental predictions of the theories arised from

14) J. S. Levinger and H. A. Bethe, *Phys. Rev.*, **78**, 115 (1950).

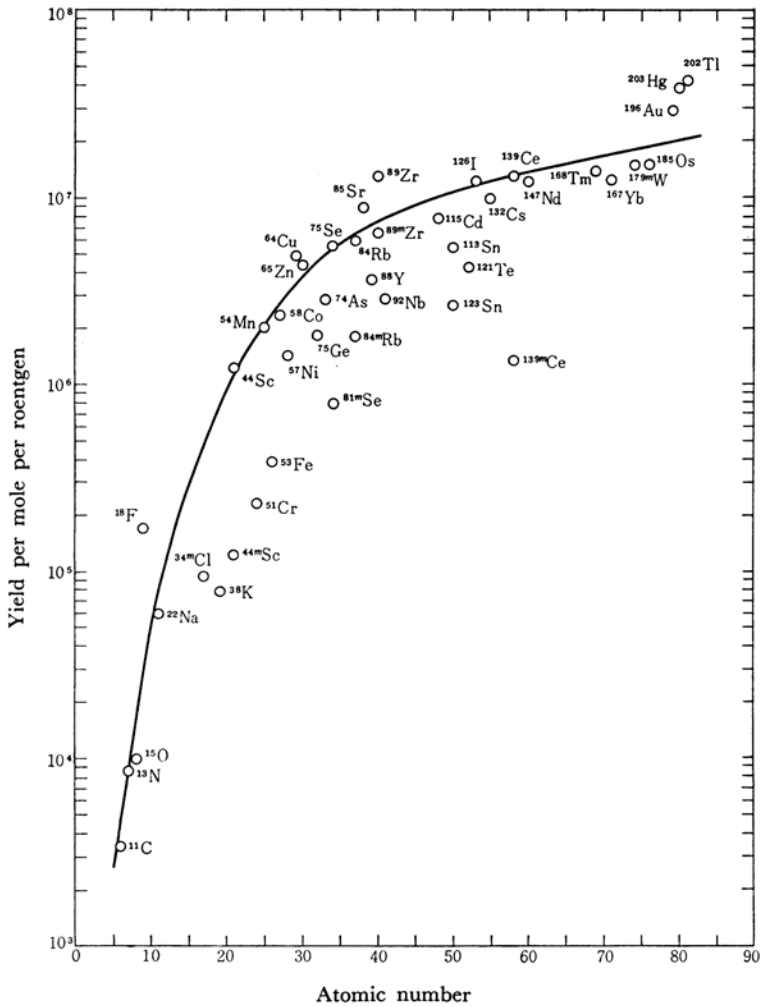

Fig. 1. The yields of radioactivities from (γ, n) reactions as a function of atomic number with 20 MeV bremsstrahlung.

the application and extension of the same rules for dipole photon-absorption to absorption by nuclei.

In some nuclei, especially light ones, the bremsstrahlung energy was not enough high to cause the high yields because of the relatively high values of the (γ, n) reaction thresholds. Thus the yields of ^{11}C from ^{12}C (S_n: 18.72 MeV) and ^{15}O from ^{16}O (S_n: 15.67 MeV) were considerably low.

Anomalously small values were obtained for the yields of radioactivities from the irradiations of the parent nuclides with neutron or proton number of the magic numbers. The yields of the (γ, n) products from ^{39}K (N=20), ^{54}Fe (N=28), ^{52}Cr (N=28), ^{89}Y (N=50), ^{58}Ni (Z=28), ^{114}Sn (Z=50) and ^{124}Sn (Z=50) were fairly low compared with those of their neighboring nuclides. It can also be seen from Fig. 1 that the yields of several isomeric pairs, including ^{44}Sc-^{44m}Sc, ^{84}Rb-^{84m}Rb, ^{89}Zr-^{89m}Zr, ^{139}Ce-^{139m}Ce, show another anomaly. The yields of ^{44m}Sc and ^{139m}Ce were obtained as about 10 per cent or less of those of the ground state isomers. In these and other cases, including ^{34m}Cl, ^{81m}Se, and other nuclides, where the isomers in upper excited level possess very high spin, the producibilities were found to be considerably restricted.

Other Photonuclear Reaction. The yields of some (γ, 2n) reactions obtained in this experiment are described in Table 2.[15] The threshold energies of the (γ, 2n) reactions of most of the light nuclei are high and hence the productions of the radioactivities due to the (γ, 2n) reactions were energetically forbidden in this bremsstrahlung

15) The chemical forms of the target materials used in this experiment were as follows: the element o bismuth: the oxides of cobalt, lanthanum, praceodymium and lutetium: bromine as its lithium salt.

TABLE 2. THE YIELDS OF SOME (γ, 2n) REACTIONS WITH 20 MeV BREMSSTRAHLUNG

Reaction	Half-life of product	Yield obtained	
		μC_i/mg*	Yield/mol·R
$^{59}Co(\gamma, 2n)^{57}Co$	270 d	5.3×10^{-5}	7.0×10^{3}
$^{79}Br(\gamma, 2n)^{77}Br$	58 hr	3.6×10^{-3}	9.8×10^{3}
$^{138}La(\gamma, 2n)^{136}La$	9.5 min	3.1×10^{-2}	1.1×10^{6}
$^{141}Pr(\gamma, 2n)^{139}Pr$	4.5 hr	3.1	7.1×10^{5}
$^{175}Lu(\gamma, 2n)^{173}Lu$	625 d	1.1×10^{-3}	1.0×10^{6}
$^{209}Bi(\gamma, 2n)^{207}Bi$	8.0 yr	6.7×10^{-5}	3.0×10^{5}

* The value corrected at the end of one-hour irradiation.

energy region. The yield values, however, became an appreciable extent with medium weight nuclei and reached about 10 per cent of those of the (γ, n) reactions with high Z nuclei. In the main the product nuclei from the (γ, 2n) reactions only emit soft gamma-radiations which are difficult to detect, and hence would not disturb the gamma-ray spectrometric measurements in the photoactivation analyses.

The productions of the (γ, p) reactions were appreciable and about same order of the yield values have been measured with light nuclei, but with heavier nuclei they show only small contributions to the total residual activities because of Coulomb barrier restrictions. The yields of radioactivities from the (γ, p) reactions on many elements, as well as those from another charged particle emitting reactions, have been determined and it is planned to publish these results elsewhere.

It is believed that the present result permit a study of the general consideration on photoactivation analyses of many elements utilizing the (γ, n) reaction products.