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A Study on the Yield of (γ, p) Reactions with 20 MeV Bremsstrahlung

Yoshinaga OKA, Toyoaki KATO, Koichi NOMURA, Tatsuya SAITO and Hui-Tuh TSAI

Department of Chemistry, Faculty of Science, Tohoku University, Katahira-cho, Sendai

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The yields of the (γ, p) reactions on 13 elements up to an atomic number of 80, irradiated with 20 MeV bremsstrahlung, have been given through the γ-ray spectrometric measurements of the resultant activities. The yield values, as measured in absolute disintegration rates at the end of irradiation per mole per roentgen, have been plotted against atomic number of the target nuclides. The shape of the yield curve thus obtained is similar to that of the curve for the photoproton yield as a function of atomic number. The yield values are 10^5 order of magnitude in the lower-Z region, and drop rapidly with increasing atomic number up to a Z of 50. For barium and mercury targets, a yield value of 2.2×10^4 was obtained. Neutron excess nuclides were found to be restrictive to the (γ, p) processes. The present results permit a study of general behavior of the (γ, p) reaction yields as well as of the applicabilities of the resultant activities to photoactivation analyses for many elements distributed throughout the periodic table.

A general feature of the photon absorption cross section for a nucleus is characterized by a peak 5—6 MeV wide at an energy located between the photodisintegration threshold and about 30 MeV.[1] For most nuclei, an excitation energy of 20 MeV is sufficient to cause the emission of one neutron, or one proton, and occasionally two particles. When bremsstahlung, with a maximum energy of 20 MeV, is attenuated by various target elements, the resultant nuclides are mainly attributed to those arising from (γ, n), and (γ, p) reactions. In a previous paper,[2] the yields of radioactivities resulting from (γ, n) reactions on many elements from lithium up to bismuth have been determined by utilizing a 20 MeV linear electron accelerator and by applying the residual activity method based on the γ-ray spectrometry. The applicabilities of these photoactivation products to activation analyses have also been discussed from the data thus obtained.[3] Although the proton

1) R. Montalbetti, L. Katz and J. Goldenberg, *Phys. Rev.*, **91**, 659 (1953).

2) Y. Oka, T. Kato, K. Nomura and T. Saito, This Bulletin, **40**, 575 (1967).

3) Y. Oka, T. Kato, K. Nomura and T. Saito, *J. Nucl. Sci. Technol.*, **4**, 346 (1967).

yields from many elements irradiated with bremsstrahlung radiation of moderate energy have been determined by several workers through the direct detection of the emitted protons,[4-9] this only represents the sum of the yields of the (γ, p), (γ, pn), and other proton emitting reactions on the relevant element. These yield data will sometimes be ambiguous when applied to a more general consideration convering photoactivation analyses of many elements using particular residual nuclides generated from each of the (γ, p) reactions.

In the present work, consequently, a study has been carried out to obtain the yields of (γ, p) reactions of some 13 elements irradiated with 20 MeV bremsstrahlung γ-rays through γ-ray spectrometric measurements of the resultant activities. In addition, the yields of some (γ, pn) reactions which occurred in this energy region have been determined separately.

Experimental

Samples and Irradiation. Samples of the target materials with high chemical purity,[10] either in the form of the element or some simple compound, were irradiated for 1 hr with bremsstrahlung γ-rays of 20 MeV maximum energy produced by the linear electron accelerator of the Japan Atomic Energy Research Institute. Each sample had a weight of 100 mg and was contained in a small quartz tube having an internal diameter of 4 mm. It was located in a target holder immediately behind the converter platinum plate. To monitor the bremsstrahlung flux to which the sample was exposed, a gold foil[11] was wrapped around each sample tube, and after irradiation, the 6.18 d ^{196}Au activity produced by the ^{197}Au(γ, n) reaction was measured from its characteristic 0.354 MeV γ-ray photopeak. The dose rate of bremsstrahlung γ-rays to which the sample was exposed has been estimated to be 9.4×10^6 R/min.

The above experimental method is essentially the same as that described in detail in previous papers.[2,3,12]

Chemical Separation. For many elements chosen for this experiment, non-destructive γ-ray spectrometric measurements could be performed after having been cooled for a time. It was possible to let the activities of the desired (γ, p) reaction products be much larger than those from the (γ, n) and other reaction products without any chemical separation. This is not the case, however, for zinc, germanium, strontium, and barium targets. After irradiation, chemical separation was performed for these targets by the following procedures.

Copper from Zinc. After the addition of copper carrier (25 mg), the zinc target was dissolved with 8 N hydrochloric acid, and then subjected to an anion exchange procedure.[13] Copper was eluted from a Dowex 1×8 column with 3.5 N hydrochloric acid, and precipitated from this eluate by the addition of aqueous sodium hydroxide solution at pH 7. After separation by centrifuge, the precipitate was ignited to obtain copper(II) oxide. The chemical yield was 67.3%. The final oxide was packed in a small glass tube having an internal diameter of 4 mm for γ-ray counting.

Gallium from Germanium. An irradiated germanium dioxide target was transferred to a 50 *ml* distilling flask, and was brought into solution by adding 7 N hydrochloric acid along with gallium (269 mg) and zinc carriers. Almost all germanium activities were removed by distillation. From the resultant solution, gallium was extracted with isopropyl ether. The organic layer was evaporated to dryness, and the residue was dissolved in dilute hydrochloric acid. Gallium hydroxide was separated from this solution by the addition of aqueous ammonia, and then ignited to obtain gallium(III) oxide. The chemical yield was 90.9%.

Rubidium from Strontium. A strontium carbonate target was dissolved in dilute hydrochloric acid with the addition of 40 mg of rubidium carrier, and evaporated to dryness. The residue was extracted with water. Sodium tetraphenyl borate was added to precipitate rubidium.

The precipitate was separated by centrifuge, washed with water containing a small amount of acetic acid, and dried at 110°C. The chemical yield was 72.1%.

Cesium from Barium. The chemical procedure for the isolation of cesium activity from a barium carbonate target was similar to that used for rubidium from strontium. Cesium tetraphenyl borate was used to determine the chemical yield, and γ-ray activity. The chemical yield was 91.6%.

Measurement of radioactivity was carried out with a 3″ dia. × 3″ NaI(Tl) detector coupled to a TMC 400-channel pulse-height analyzer.

Yield Determination. The yield value was obtained for each (γ, p) reaction through the absolute measurement of the resultant activity. To obtain the disintegration rate from the counting rate of the characteristic γ-ray photopeak, appropriate corrections were made for the variation of photopeak efficiency with γ-ray energy.[14] The decay schemes used were those listed in the Nuclear Data Sheets.[15] The tables of internal conversion coefficients were also used.[16] Decay curve analyses have been performed to separate the undesired activities. From the data thus obtained, the yield ($Y_{(E)}$) was calculated by the following equation.

4) D. C. Diven and G. M. Almy, *Phys. Rev.*, **80**, 407 (1950).
5) A. K. Mann and J. Halpern, *ibid.*, **82**, 733 (1951).
6) M. E. Toms and W. E. Stephens, *ibid.*, **82**, 709 (1951).
7) M. E. Toms and W. E. Stephens, *ibid.*, **92**, 362 (1953).
8) W. A. Butler and G. M. Almy, *ibid.*, **91**, 58 (1953).
9) E. V. Weinstock and J. Halpern, *ibid.*, **94**, 1651 (1954).
10) 99.99% or over.
11) 10 μ thick, 3 mm × 15 mm.
12) Y. Oka, T. Kato and I. Nagai, *J. Nucl. Sci. Technol.*, **4**, 300 (1967).
13) K. A. Kraus and G. E. Moore, *J. Am. Chem. Soc.*, **75**, 1460 (1953).
14) S. Flügge Ed., "Handbuch der Physik, Bd. XLV. Instrumentelle Hilfsmittel der Kernphysik II," Springer-Verlag, Berlin (1958), p. 113.
15) Nuclear Data Sheets, Natl. Academy of Science-Natl. Research Council, Washington, D. C. (1961).
16) M. E. Rose, "Internal Conversion Coefficient," North-Holland Publ. Co., Amsterdam (1958).

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

where D_0 is the disintegration rate at the end of irradiation; I is the radiation dose rate; M is the amount of target nucleus expressed in moles; λ is the decay constant.

Effect of Photoneutrons. During irradiation in this experiment, there was a considerable neutron flux at the site of the target sample which was due to photoneutrons ejected from the platinum converter. These photoneutrons induce extraneous neutron reactions such as (n, p), (n, α), and (n, 2n) reactions. For a number of elements the resultant nuclide formed by the (n, p) reaction is the same as that from the (γ, p) reaction on the corresponding element. In such cases, it was necessary to correct the observed activity for the contribution of the (n, p) reaction. To investigate such contributions, the neutron flux to which the sample was exposed has been determined. For this purpose, $^{27}Al(n, \alpha)^{24}Na$ reaction was chosen as a monitor reaction.[17] The energy distribution curve of photoneutrons emitted from a bismuth target irradiated with 20 MeV bremsstrahlung which had been given by Mutsuro *et al.*[19] was also used.[20]

When a piece of aluminum metal of 4 mm×4 mm (126 mg) was irradiated for 1 hr with 20 MeV bremsstrahlung, as pecific activity of 3.2×10^{-3} μCi ^{24}Na/mg Al was found at the end of irradiation. By using this value, the intensity of the energy distribution curve was normalized to the neutron flux at the various neutron energies. The resultant curve is shown in Fig. 1-(1).[21]

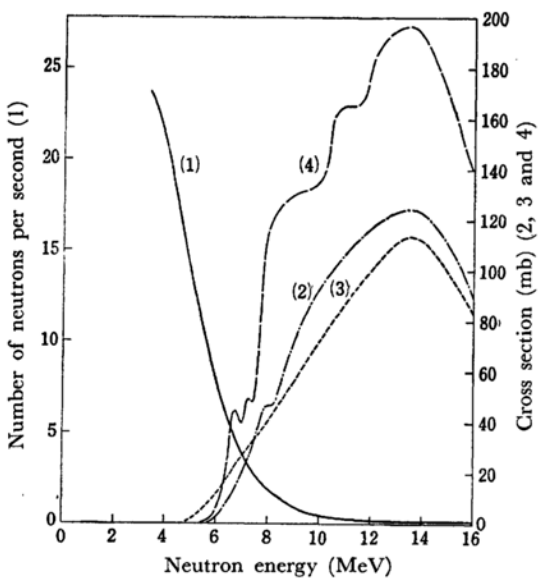

Fig. 1. Neutron flux at the target position as a function of neutron energy (1); The excitation curves for (2) the $^{27}Al(n, \alpha)^{24}Na$[18], (3) the $^{56}Fe(n, p)^{56}Mn$[23] and (4) the $^{24}Mg(n, p)^{24}Na$[18] reaction.

By using this curve and the excitation function for the particular (n, p) reaction, the reaction rate can be calculated in the form of $NS\int_{E_{th}}^{\infty}\sigma(E)\phi(E)dE$, where N is the number of target nuclides; S is the saturation factor; $\sigma(E)$ is the cross section at a neutron energy (E); $\phi(E)$ is the neutron flux at energy (E); and E_{th} is the threshold energy of the reaction.

In the irradiation of an iron target, ^{56}Mn activity is produced both by the $^{57}Fe(\gamma, p)^{56}Mn$, and by the $^{56}Fe(n, p)^{56}Mn$ reactions. The production rate of the latter was estimated by the proposed method[22] to be 2.3×10^{-3} μCi ^{56}Mn/mg Fe. This corresponds to 2.8% of the total ^{56}Mn activity. Next, ^{24}Na activity from a magnesium target formed by the $^{24}Mg(n, p)^{24}Na$ reaction[24] was calculated to be 5% of the total ^{24}Na activity. Mostly, the excitation curve for the (n, p) reaction exhibits a threshold energy of 2—5 MeV, and is characterized by a maximum value of ~100 mb.[23] Because of this, it can be concluded that the (n, p) reactions contribute only a small fraction in the total resultant activity, which can thereby be attributed, in major part, to the decay of the (γ, p) reaction products.

Results and Discussion

The (γ, p) Reaction Yield. Table 1 lists the particulars and the data obtained on 16 γ-ray emitting products of the (γ, p) reactions from the elements studied. Column three in Table 1 gives the proton separation energy (S_p), the energy required to separate one proton from a parent nucleus, presented by Yamada and Matsumoto,[25] for most of the nuclei by the use of mass formulas and nuclear systematics. The last two columns show the experimental results. The yield value was expressed as the disintegration rate at the end of irradiation per mole of target nuclide per roentgen. This can be taken as the number of the (γ, p) processes which occurred during irradiation. The yield values were also plotted against atomic number of the parent nuclides in Fig. 2 in which the symbol noted near each plot indicates the parent nuclide. The shape of the yield curve is similar to that of the curve for the photoproton yield as a function of atomic number presented by Weinstock and Halpern.[9]

17) The cross section values for this reaction were taken from the data of Butler *et al.*,[18] which is shown in Fig. 1-(2).

18) J. P. Butler and D. C. Santry, *Can. J. Phys.*, **41**, 372 (1963).

19) N. Mutsuro *et al.*, To be published.

20) It can be said that this energy distribution curve is practically the same as that for photoneutrons from the converter platinum taking account of the characteristics of the photonuclear process.

21) From the result, the neutron flux necessary to produce the ^{24}Na activity was estimated to be 5.3×10^5 n/sec at the target position.

22) The excitation function for the $^{56}Fe(n, p)^{56}Mn$ reaction, shown in Fig. 1-(3), was taken from the data compiled by Hughes *et al.*[23]

23) D. N. Hughes and J. A. Harvey, "Neutron Cross Sections," BNL-325 (1955).

24) The excitation function presented by Butler *et al.*,[18] shown in Fig. 1-(4), was used.

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

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

Nuclide: Parent (Natural abundance, %)	Nuclide: Residual (Half-life)	S_p (MeV)	Observed γ-ray: Energy (MeV)	Observed γ-ray: Branching ratio (%)	Observed γ-ray: Type of multipole transition	Yield determined: μCi/mg[a)]	Yield determined: Yield/mol·R
^{25}Mg (10.11)	^{24}Na (15 hr)	12.06	1.37	100	$E2$	1.48×10^{-1}	1.7×10^{5}
^{29}Si (4.71)	^{28}Al (2.27 min)	12.33	1.78	100	$E2$	1.91	2.8×10^{5}
^{30}Si (3.12)	^{29}Al (6.56 min)	13.59	1.28	93.8	$E2+M1$	6.51×10^{-1}	1.5×10^{5}
^{44}Ca (2.06)	^{43}K (22.4 hr)	12.17	0.374	85	$E2+M1$	7.86×10^{-3}	1.3×10^{5}
^{47}Ti (7.32)	^{46}Sc (84.1 d)	10.47	0.887	100	$E2$	7.11×10^{-4}	3.1×10^{5}
^{48}Ti (73.99)	^{47}Sc (3.4 d)	11.44	0.160	100	$E2+M1$	6.83×10^{-2}	1.2×10^{5}
^{49}Ti (5.46)	^{48}Sc (1.8 d)	11.35	1.31	100	$E2$	4.40×10^{-3}	5.8×10^{4}
^{53}Cr (9.55)	^{52}V (3.8 min)	11.15	1.43	100	$E2$	5.01×10^{-1}	6.6×10^{4}
^{57}Fe (2.17)	^{56}Mn (2.58 hr)	10.57	1.81	23.5	$E2+M1$	8.10×10^{-2}	2.1×10^{5}
^{74}Ge (36.74)	^{73}Ga (4.8 hr)	10.92	0.295	97	($E2$)	3.70×10^{-1}	1.3×10^{5}
^{77}Se (7.58)	^{76}As (26.5 hr)	9.61	0.559	41	$E2$	1.48×10^{-2}	1.3×10^{5}
^{87}Sr (7.02)	^{86}Rb (19 d)	9.41	1.08	9	$E2$	5.15×10^{-4}	9.9×10^{4}
^{113}Cd (12.26)	^{112}Ag (3.2 hr)	9.74	1.39	35	$E2$	1.91×10^{-2}	2.1×10^{4}
^{117}Sn (7.57)	^{116m}In (54 min)	9.58	1.27	84	$E2$	9.80×10^{-3}	6.9×10^{3}
^{137}Ba (11.32)	^{136}Cs (13 d)	8.67	0.830	100	$E2$	1.68×10^{-4}	2.2×10^{4}
^{199}Hg (16.84)	^{198}Au (2.7 d)	7.27	0.412	100	$E2$	8.43×10^{-4}	2.2×10^{4}

a) The value corrected at the end of 1 hr irradiation (9.4×10^{6} R/min).

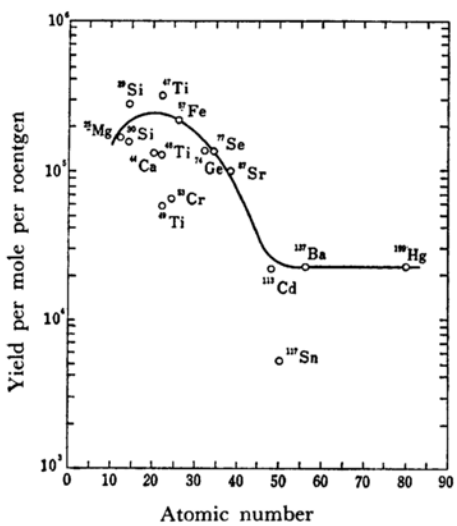

Fig. 2. The yield curve for the (γ, p) reaction with 20 MeV bremsstrahlung.

It has been pointed out that the photon absorption cross section for a nucleus is an increasing function of Z, and this fact has been well explained in a study on the relation between the (γ, n) reaction yield and the atomic number of the target nuclide.[1,2] The yield of radioactive nuclide increases regularly with atomic number from 10^3 to 10^7 orders of magnitude.[2] On the other hand, photoproton emission from a nucleus is a decreasing function of Z because of the restrictive effect of the Coulomb barrier. Accordingly, the yield curve for the photoproton emission will exhibit a peak.

According to the results of Weinstock and Halpern,[9] this peak occurs at nickel (Z=28). They have also shown that between a Z of 28 and 50 the experimental photoproton yields are in excellent agreement in absolute magnitude with those calculated from the evaporation model.

Although the initial rise of the yield curve, shown in Fig. 2, is not markedly observed in the lower-Z region, a typical decrease in the yield value with Z can be seen from iron (Z=26) up to tin (Z=50). On the barium and mercury targets, however, the yield values are much higher than those expected from a rapid drop until a Z of 50. These discrepancies can be attributed to the direct photoelectric emission mechanism. It has often been pointed out that the evaporation model as usually formulated is inadequate to explain the photoproton yields for elements of high atomic number.[9,26] In such cases, a considerable portion of the photoproton yield can be accounted for by the direct photoelectric emission of a proton without the formation of a compound nucleus.

As for titanium isotopes, including ^{47}Ti, ^{48}Ti, and ^{49}Ti, the yield values decrease in order. This fact shows that the cross section for photoproton emission from a neutron excess nuclide is expected to be small. Similarly, the yield for the ^{29}Si(γ, p)^{28}Al reaction is higher than that for the ^{30}Si(γ, p)^{29}Al reaction.

Of the target nuclides under study, ^{44}Ca (Z=20) and ^{117}Sn (Z=50) are proton magic. The yield values for these nuclides are fairly low as compared with those for the neighboring nuclides in the yield curve.

26) V. G. Shevchenko and B. A. Yuryev, *Nucl. Phys.*, **37**, 495 (1962).

In the comparison of the results in Table 1 with the yield values of the (γ, n) reactions, the (γ, p) reactions can be produced by same order of magnitude as the latter in elements with lower-Z. With higher-Z elements, however, they contribute only a small fraction to the observed activity. The yield values are less than 0.1% of those for the (γ, n) reaction in elements with Z greater than 50.

It would appear that the use of the resultant activities in Table 1 for photoactivation analysis of lighter elements should enhance the efficacy. Silicon, calcium, and titanium can be specifically detected by γ-ray counting. They are elements from which either very short-lived nuclides, or only low γ-activities, are produced by activation with thermal neutrons.

In conclusion, the authors believe that the present results permit a study of the general considerations covering photoactivation analyses of many elements utilizing (γ, p) reaction products.

The (γ, pn) Reaction. It is expected that the (γ, pn) reactions take place to an appreciable extent, apart from the (γ, n) and (γ, p) reactions, in various elements in the energy region covered in this experiments. The resultant activities from (γ, pn) reaction in iron, zinc,[27] and palladium targets could be measured. The yield values obtained are given in Table 2. This is a preliminary survey to indicate the yield values from the (γ, pn) reaction in various elements. It is obvious that more points at higher atomic number are required. It is planned to investigate these problems in the near future.

Table 2. The yields of some (γ, pn) reactions with 20 MeV bremsstrahlung

Reaction	Half-life of product	Specific activity[a] (μCi/mg)	Yield ($mol^{-1} \cdot R^{-1}$)
$^{56}Fe(\gamma, pn)^{54}Mn$	314 d	2.5×10^{-6}	3.6×10^{2}
$^{66}Zn(\gamma, pn)^{64}Cu$	13 hr	7.2×10^{-3}	7.5×10^{3}
$^{104}Pd(\gamma, pn)^{102}Rh$	210 d	1.1×10^{-6}	1.7×10^{3}

a) The value corrected at the end of 1 hr irradiation (9.4×10^{6} R/min).

27) Apart from ^{64}Cu, ^{67}Cu from ^{68}Zn by the (γ, p) process was measured by its characteristic 0.093, 0.184, 0.296, and 0.388 MeV γ-rays. The yield calculation for this reaction was excluded in this experiment since the decay scheme for ^{67}Cu has not been well-established.