

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

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The yields of the (γ, α) reactions on 7 elements up to an atomic number of 49, irradiated with 20 MeV bremsstrahlung, have been obtained through gamma-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 yield curve shows a maximum in the region of copper and zinc, and drops rapidly with increasing atomic number. The shape of this yield curve is similar to that for the photoalpha yield as a function of atomic number. It can be seen that the yield levels off in the region of silver and indium. A mechanism such as the direct photoelectric effect is thought to be responsible for this behavior. The yield of the (γ, α) reaction of an element amounts to only a small fraction, at most a few tenths of a per cent, of the total photonuclear yield.

When bremsstrahlung photons with energies in the giant resonance region are attenuated by various target elements, the main nuclear events are the emission of one neutron or one proton. In previous work,¹⁻⁴⁾ the yield of the (γ, n) and (γ, p) reactions have been determined with 20 MeV bremsstrahlung as a function of target nuclear charge, and the applicabilities of these reaction products to activation analyses have also been discussed from the data thus obtained. Since the binding energy of alpha-particles is rather low, it is expected that the (γ, α) processes, apart from (γ, n) and (γ, p) processes, can occur in the energy region of 20 MeV bremsstrahlung. Especially for medium weight nuclei, the resultant activities due to (γ, α) processes are expected to be an appreciable fraction of the total resultant activity, because of the relatively low Coulomb barriers. Greenberg *et al.*⁵⁾ reported

on the yields of alpha-particles emitted from 9 elements up to an atomic number of 58 in the energy region of 23 MeV bremsstrahlung. Some additional details have been clarified by several workers,⁶⁻⁹⁾ but not studied sufficiently.

In this report, the yields of (γ, α) reactions produced by irradiation of several medium weight nuclei with 20 MeV bremsstrahlung have been given through gamma-ray spectrometric measurements of the resultant activities.

Experimental

Samples and Irradiation. Samples of the target materials with high chemical purity, either in the form of the element or some simple compound, were irradiated for 1 hr with bremsstrahlung gamma-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

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

2) Y. Oka, T. Kato, K. Nomura, T. Saito and H-T. Tsai, *ibid.*, **41**, 380 (1968).

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

4) Y. Oka, T. Kato and I. Nagai, *ibid.*, **4**, 300 (1967).

5) L. H. Greenberg, J. G. V. Taylor and R. N. H. Haslam, *Phys. Rev.*, **95**, 1540 (1954).

6) P. Erdős, P. Scherrer and P. Stoll, *Helv. Phys. Acta*, **30**, 639 (1957).

7) F. Heinrich, H. Wäffler and M. Walter, *ibid.*, **29**, 3 (1956).

8) J. H. Carver, *Proc. Phys. Soc.*, **77**, 417 (1961).

9) L. Meneghetti and S. Vitale, *Nucl. Phys.*, **61**, 316 (1965).

4 mm. The experimental method involving bremsstrahlung flux monitoring and details on irradiation are essentially described in detail in previous papers.¹⁻⁴⁾

Chemical Separation. For the vanadium target, gamma-ray spectrometric measurements of the scandium activities were performed non-destructively after cooling. The other seven targets were processed radiochemically by the following procedures. In each case, the final precipitate was packed in a small glass tube having an internal diameter of 4 mm to determine the chemical yield and for gamma-ray counting.

Cobalt from Copper. After the addition of cobalt carrier (20.0 mg), the copper target was dissolved in nitric acid and evaporated nearly to dryness. The residue was dissolved in 6 N hydrochloric acid and subjected to anion exchange separation. Cobalt was eluted from a Dowexl X8 column with 6 N hydrochloric acid, and precipitated from this eluate by the addition of an aqueous solution of α -nitroso- β -naphthol. The precipitate was centrifuged and ignited to obtain cobalt(II) oxide. Almost complete recovery was attained.

Copper from Gallium. A gallium(II) oxide target was brought into solution by adding nitric acid along with copper carrier (67.3 mg). After evaporating nearly to dryness, the residue was made 3.5 N in hydrochloric acid, and precipitated from this solution by the addition of aqueous ammonium sulfide. After separation with a centrifuge, the sulfide was ignited to obtain copper(II) oxide. The chemical yield was 88.8%.

Zinc from Germanium. An irradiated germanium dioxide target was transferred to a 50 ml distilling flask, and brought into solution by adding 7 N hydrochloric acid along with gallium (269 mg) and Zinc (27.0 mg) carriers. Almost all germanium activity was removed by distillation. From the resultant solution, gallium was extracted with isopropyl ether. The aqueous layer was neutralized by the addition of a dilute aqueous sodium hydroxide solution, and the zinc hydroxide precipitate was separated by centrifugation. The precipitate was ignited to obtain zinc(II) oxide. The chemical yield was 12.9%.

Arsenic from Bromine. To the aqueous solution of an irradiated lithium bromide target arsenic(III) and arsenic(V) carriers (total 30.2 mg) were added. Arsenic was precipitated from this solution in the form of the elemental powder by reduction with tin(II) chloride in concentrated hydrochloric acid on warming. After separation by centrifugation, the precipitate was washed repeatedly with water, and dried in a vacuum. The chemical yield was 72.5%.

Rhodium from Silver. An irradiated silver target was dissolved in nitric acid with the addition of rhodium(III) and rhodium(IV) carriers (total 72.9 mg). An aqueous solution of sodium chloride was added to this solution to precipitate silver chloride. The filtrate was further purified by repeated silver chloride scavenging. Finally, metallic rhodium was precipitated by reduction with the addition of zinc dust. The precipitate was washed with dilute nitric acid and with water, and subsequently dried in a vacuum. The chemical yield was 20.4%.

Silver from Indium. After the addition of silver carrier (35.9 mg), an irradiated indium target was dissolved in nitric acid. Silver chloride was precipitated from this solution by the addition of dilute hydrochloric acid.

The precipitate was purified by repeated reprecipitations involving dissolution with aqueous ammonia and precipitation with dilute hydrochloric acid. Finally, the silver chloride was washed with water and dried at 110°C. The chemical yield was 80.5%.

Yield Determination. Yield has been defined as the production rate of a given radioactive nuclide per mole of target nuclide per roentgen. The initial rates of the various activities produced were determined from their photopeak areas from decay curve analyses of the counting rates, and the corresponding saturation rates computed. After correcting for chemical yields, counting efficiencies, branching ratios and internal conversion electrons, the data has been expressed in the form of yields. The decay schemes used were those listed in the Nuclear Data Sheets.¹⁰⁾ A table of internal conversion coefficients¹¹⁾ was also used.

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

Results

Table 1 lists the characteristics and experimental results obtained from 7 gamma-ray emitting products for (γ , α) reactions of the elements studied. The threshold energies for these reactions were

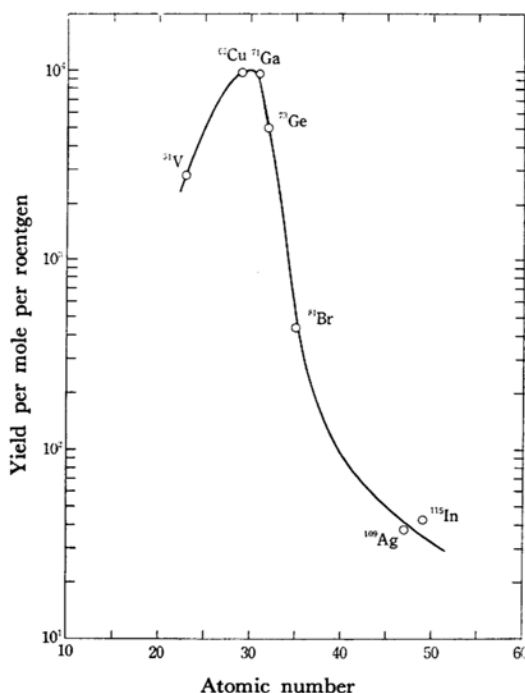


Fig. 1. The yield curve for (γ , α) reaction with 20 MeV bremsstrahlung.

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

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

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

Nuclide		E_{th} ($-Q$, MeV)	Observed gamma-ray			Results obtained	
Parent (Natural abundance, %)	Product (Half-life)		Energy (MeV)	Branching ratio (%)	Type of multipole transition	$\mu\text{Ci/mg}^a$	Yield ($\text{mol}^{-1} \cdot \text{R}^{-1}$)
^{51}V (99.75)	^{47}Sc (3.4 d)	10.27	0.160	100	$M1+E2$	1.99×10^{-3}	2.8×10^3
^{65}Cu (30.9)	^{61}Co (99 min)	6.75	0.068	99	$M1+E2$	7.23×10^{-3}	9.7×10^3
^{71}Ga (39.6)	^{67}Cu (61 hr)	5.15	0.184	41	$M1$	2.70×10^{-3}	9.6×10^3
^{78}Ge (7.67)	^{60m}Zn (14 hr)	5.89	0.435	100	$M4$	1.11×10^{-2}	5.0×10^3
^{81}Br (49.48)	^{77}As (39 hr)	6.46	0.246	2.81	$M1+E2$	1.97×10^{-4}	4.3×10^2
^{109}Ag (48.65)	^{105}Rh (36 hr)	3.28	$0.319+0.306$	24.8	$M1+E2$	8.29×10^{-4}	3.7×10^1
^{115}In (95.77)	^{111}Ag (7.6 d)	3.78	0.340	6	$M1+E2$	5.70×10^{-5}	4.3×10^1

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

calculated on the basis of the mass differences.¹²⁾ The yield value, shown in the last column of Table 1, are 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 (γ, α) processes which occurred during irradiation. The yield values were also plotted against atomic number of the parent nuclides in Fig. 1 in which the symbol noted near each plot indicates the parent nuclide. The yield curve exhibits a maximum in the region of copper and zinc, and drops rapidly with increasing atomic number. It is seen that the yield levels off in the region of silver and indium.

Discussion

The shape of the yield curve, shown in Fig. 1, is very similar to that of the curve for the photo-alpha yield as a function of atomic number with 23 MeV bremsstrahlung presented by Greenberg *et al.*⁵⁾ The curve is also similar to that for the (γ, p) reaction yield reported previously.²⁾ The (γ, α) reaction yield is lower by a factor varying from 20 to 10^3 in the region of $Z < 50$. The absorption cross-section for gamma-rays increases with increasing atomic number, and the potential barrier also increases. From the relative effects of these factors, the yield curve shows an initial rise and then, through a maximum point, drops rapidly with increasing atomic number. For silver and indium targets, however, the yields are much

higher than those expected from the rapid drop until the bromine target. The presence of direct effects in the photoproduction of alpha-particles from heavier elements has been suggested by several workers.^{8,9)} Erdős *et al.*⁶⁾ reported that the photoalpha yield curve levels off at a roughly constant value for elements with atomic number greater than 50. Furthermore, disagreement between the calculated evaporative spectra of emitted photoalpha particles and the experimental ones was pointed out by Meneghetti *et al.*⁹⁾ An excess of energetic alpha-particles, especially in indium and gold irradiated with 35 MeV bremsstrahlung gamma-rays, has been attributed to direct effects. The leveling-off behavior of the yield curve in the region of silver and indium obtained in this experiment suggests that a mechanism such as the direct photoelectric effect becomes more important for heavier elements.

Experimental results show that the (γ, α) reaction yield of an element amounts to only a small fraction of the total photonuclear yield. The relative yields of the (γ, α) reaction to those of the (γ, p) and (γ, n) reactions in the region of copper and zinc have been determined as

$$Y_{(\gamma, \alpha)} : Y_{(\gamma, p)} : Y_{(\gamma, n)} = 1 : 20 : 500$$

In the region of silver and indium, the relative yields are

$$Y_{(\gamma, \alpha)} : Y_{(\gamma, p)} : Y_{(\gamma, n)} = 1 : 10^3 : 3 \times 10^5$$

Since the yield of a photonuclear reaction is a strong function of the maximum bremsstrahlung energy, the relative probability of producing each activity as the result of photonuclear processes with various energies is to be investigated in detail through further experiments.

12) Nuclidic mass values used were those listed in G. Friedlander, J. W. Kennedy and J. M. Miller, "Nuclear and Radiochemistry," 2nd Ed., John Wiley & Sons, New York (1964), p. 533.