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The Yields of Photonuclear Reactions Leading to the Production of Beryllium-7 with Bremsstrahlung up to 60 MeV

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Beryllium, boron, carbon, oxygen, and fluorine targets with natural isotopic compositions were irradiated by bremsstrahlung with four maximum energies, 30, 45, 55 and 60 MeV, and the yields of ^{7}Be from each of these targets were determined by means of chemical separation and gamma-ray spectrometric measurements. The yields were obtained relative to the yield of the $^{12}C(\gamma, n)^{11}C$ reaction at each bremsstrahlung energy. The possible reaction pathways leading to the production of ^{7}Be were discussed. The exponential decrease in the yield value against the difference in mass between the target and the product nuclei was observed.

Knowledge of the magnitude and energy dependences of photonuclear reaction yields is basic to an understanding of the interaction of high-energy photons with nuclei. In a previous work,[1] the yields of a number of radioactive nuclei produced by (γ, xnyp) reactions on titanium and vanadium targets have been determined with bremsstrahlung up to 60 MeV. ^{7}Be decays with a half-life of 53 days upon K-capture with a 477 keV gamma-ray, accompanied by a 10.3 per cent in the decay processes.[2] These properties support the idea of obtaining some evidence of the occurrence of high-energy processes in light nuclei through measuring the ^{7}Be activities. ^{7}Be has been observed as a spallation product in a number of high-energy processes with charged particles.[3-7]

Several investigators[8-13] have also shown that ^{7}Be can be produced with high-energy photons from various light elements. However, many of their reports have been only about brief examinations.

In this work, beryllium, boron, carbon, oxygen, and fluorine targets with natural isotopic compositions were irradiated by bremsstrahlung with four different maximum energies, 30, 45, 55 and 60 MeV, and the yields of ^{7}Be from each of these targets were determined by means of gamma-ray spectrometric measurements. The yields were taken relative to the yield of the $^{12}C(\gamma, n)^{11}C$ reaction at each bremsstrahlung energy. The possible reaction pathways leading to the production of ^{7}Be were discussed. The exponential decrease in the yield value against the difference in mass between the target and the product nuclei has also been observed.

Experimental

Target Materials. Target materials of beryllium in the form of metallic flakes, boron in powder form, a carbon spectrograph rod, silica and lithium fluoride were used. All the materials were of a high purity. A definite amount of each sample (100—500 mg) was wrapped in a thin aluminum foil and then made into a small disk 7 mm in diameter and 3 mm thick.

Irradiation. The linear electron accelerator of the Tohoku University was the bremsstrahlung source. The electron beam, accelerated by the "High Current" accelerating section of the machine,[14] produced bremsstrahlung in a platinum converter 3 mm thick located 3 cm from the beam-exit window. Over the energy range investigated, the average beam current was at least 30 μA, measured at the position of the converter by using a current monitor. The targets were set in a water-cooled target holder on the bremsstrahlung beam axis immediately behind the converter. Irradiations were terminated within 1 to 3 hr. The bremsstrahlung

1) Y. Oka, T. Kato and N. Sato, This Bulletin, to be published.
2) C. M. Lederer, J. M. Hollander and I. Perlman, "Table of Isotopes," Sixth Ed., John Wiley & Sons, New York (1967).
3) P. A. Benioff, *Phys. Rev.*, **119**, 316 (1960).
4) I. R. Williams and C. B. Fulmer, *ibid.*, **154**, 1005 (1967).
5) G. Friedlander, J. Hudis and R. L. Wolfgang, *ibid.*, **99**, 263 (1955).
6) M. Honda and D. Lal, *ibid.*, **118**, 1618 (1960).
7) G. V. S. Rayudu, *Canad. J. Chem.*, **42**, 1149 (1964).
8) K. H. Lokan, *Proc. Phys. Soc. (London)*, **A70**, 836 (1957).
9) M. S. Foster and A. F. Voigt, *J. Inorg. Nucl. Chem.*, **24**, 343 (1962).
10) H. Artus, G. Fricke and D. E. von Stein, *Z. Phys.*, **178**, 109 (1964).
11) H. Artus, *Z. Naturf.*, **20a**, 320 (1965).
12) O. L. Garzón and G. G. Rocco, *J. Inorg. Nucl. Chem.*, **29**, 1 (1967).
13) C. B. Fulmer, I. R. Williams, G. F. Dell and L. N. Blumberg, *Bull. Am. Phys. Soc.*, **12**, 499 (1967).
14) The peak current was at least 100 mA with energies up to 100 MeV. In many cases, the pulse repetition was 200 pps, with a pulse width of 3 μsec.

flux was monitored using a thin sheet of gold (10 μ thick, 7 mmϕ) adjacent to the sample.

Radiochemical Separation. To obtain the ^{7}Be spectrum without any contamination by undesirable activities in the irradiated sample, a chemical separation was necessary for all targets except beryllium. After irradiation, each target was dissolved along with 20 mg of a beryllium carrier, and then subjected to separation procedure. Boron powder was dissolved with nitric acid, and silica was dissolved with hydrofluoric acid. Wet oxidation by potassium iodate in a mixture of sulfuric and condensed polyphosphoric acids[15,16] was efficient in decomposing the carbon targets after they had been finely pulverized in a agate mortar. From these solutions, beryllium hydroxide was precipitated by the addition of aqueous ammonia and subsequent centrifuging. The hydroxide was dissolved with a small amount of nitric acid, and beryllium was extracted as its acetylacetonate into chloroform in the presence of EDTA at a pH of 7. The organic layer was back-washed twice, and then evaporated to dryness. The residual organic substances were decomposed by fuming with a sulfuric and perchloric acid mixture. The beryllium hydroxide was again precipitated from this solution by the addition of aqueous ammonia, and then ignited to obtain beryllium oxide.[17] The final oxide was packed in a small glass tube with an internal diameter of 4 mm for the determination of the chemical yield and for the gamma-ray counting.

Radioactivity Measurements. For the measurements of the gamma-ray spectra, a 3'' dia.$\times$3'' NaI(Tl) detector coupled to an 800-channel pulse-height analyzer made by the Tokyo Shibaura Electric Co., Ltd. was employed. Typical gamma-ray spectra are shown in Fig. 1. In each case, the dacay curve followed in the area under the 477 keV photopeak showed a half-life of 53 days, corresponding to the literature value of ^{7}Be.[2] The initial photopeak activity was obtained and used to determine the reaction yield.

Yield Determination. A yield was defined as the production rate of ^{7}Be per atom of a target nucleus when a standard amount of the bremsstrahlung radiation with a given maximum energy was passed through the target during the irradiation period. From the initial activity of the ^{7}Be, the corresponding saturation rate was computed and then normalized for bremsstrahlung intensity, which was measured by the amount of ^{196}Au activity in a gold foil irradiated together with the target. After correcting for chemical yield,[18] counting efficiencies, branching ratio, and internal conversion electrons, the data were expressed in the form of yields. These yields were expressed relative to that of the

TABLE 1. CALCULATED MASS THRESHOLDS FOR THE PHOTOPRODUCTION REACTIONS OF ^{7}Be FROM ^{9}Be, ^{10,11}B, ^{12}C, ^{16}O AND ^{19}F NUCLEI

Reaction	$E_{th}(-Q)$ (MeV)	Reaction	$E_{th}(-Q)$ (MeV)	Reaction	$E_{th}(-Q)$ (MeV)
^{9}Be(γ, 2n)	20.56	^{12}C(γ, αn)	26.27	^{16}O(γ, 2αn)	33.43
		(γ, td)	43.86	((γ, dtα)	51.02
^{10}B(γ, t)	18.67	(γ, ptn)	46.08	(γ, ptαn)	53.24
(γ, p2n)	27.15	(γ, ^{3}He2n)	46.85	(γ, α^{3}He2n)	54.01
(γ, dn)	36.98	(γ, 2dn)	50.11	(γ, 2dαn)	57.28
		(γ, pd2n)	52.34	(γ, pdα2n)	59.50
^{11}B(γ, tn)	30.12	(γ, 2p3n)	54.56	(γ, 2pα3n)	61.73
(γ, p3n)	38.61				
(γ, d2n)	36.38			^{19}F(γ, 2αtn)	45.13
				(γ, d2α2n)	51.39
				(γ, p2α3n)	53.61

TABLE 2. THE REACTION YIELDS LEADING TO ^{7}Be RELATIVE TO ^{12}C(γ, n)^{11}C REACTION

Target nucleus	Relative yield			
	30 MeV	45 MeV	55 MeV	60 MeV
^{9}Be	$(2.9_8\pm0.0_2)\times10^{-2}$(2)	$7.1_1\times10^{-2}$(1)	$8.4_3\times10^{-2}$(1)	$(1.3_9\pm0.1_1)\times10^{-1}$(4)
^{10,11}B	$(6.6_4\pm0.1_0)\times10^{-3}$(2)	$(1.0_3\pm0.0_6)\times10^{-2}$(2)	$1.9_3\times10^{-2}$(1)	$(3.3_9\pm0.2_3)\times10^{-2}$(3)
	$(3.1_2+0.0_5)\times10^{-2}$(2) [a]			
^{12}C	$1.3_3\times10^{-3}$(1)	$(2.6_9\pm0.0_9)\times10^{-2}$(2)	$3.2_8\times10^{-2}$(1)	$(3.7_0\pm0.0_7)\times10^{-2}$(2)
^{16}O		$1.0_2\times10^{-3}$(1)		$(3.6_1\pm0.2_4)\times10^{-3}$(2)
^{19}F				$(1.7_6\pm0.0_1)\times10^{-3}$(2)
				$(1.3_4\pm0.0_4)\times10^{-3}$(2) [b]

a) Relative yield per mole of ^{10}B. b) The yield relative to ^{19}F(γ, n)^{18}F reaction.

15) T. Kiba, S. Ohashi, T. Takagi and Y. Hirose, *Japan Analyst*, **2**, 446 (1953).
16) M. Ura, *Nippon Kagaku Zasshi* (*J. Chem. Soc. Japan, Pure Chem. Sec.*), **78**, 316 (1957).
17) In the case of ^{7}Be obtained from boron, the present procedure was insufficient for the complete removal of undesired activities; however, these activities could be removed by copper sulfide scavenging before the final hydroxide precipitation.
18) 50—70%.

$^{12}C(\gamma, n)^{11}C$ process, which was measured by the amount of 0.511 MeV gamma-rays arising from the annihilation of the positrons in a standard carbon disk irradiated together with the target.

Results and Discussion

Reaction Path. Beryllium and fluorine are the single-isotopic elements, and the natural isotopic abundances of the most abundant isotopes for carbon and oxygen are 98.893 and 99.759% respectively. Thus, it has been assumed for the sake of simplicity that each of these elements consists entirely of its most abundant isotope. Boron has the natural composition of two isotopes: ^{10}B(19.61%) and ^{11}B(80.39%). The competitive reactions take place on these two isotopes. Table 1 lists the possible nuclear reactions leading to the production of ^{7}Be in the energy region covered in this experiment, together with their calculated mass thresholds.[19] In many cases, ^{7}Be can be formed through several different reaction pathways. At an excitation energy up to 30 MeV, almost all of the ^{7}Be comes from ^{10}B *via* the (γ, t) process, and at this energy, ^{7}Be can be formed from carbon only by the $(\gamma, \alpha n)$ reaction on ^{12}C. For the production of ^{7}Be from ^{16}O, the only pathwhy which has a mass threshold below 45 MeV is the $(\gamma, 2\alpha n)$ reaction, and it may be seen that this reaction is the most probable pathway over the energy range investigated. All of the reactions leading to the photoproduction of ^{7}Be from the parent ^{19}F have threshold energies greater than 45 MeV.

Reaction Yield. The results obtained in this experiment are summarized in Table 2. The numbers in brackets indicate the number of determinations which were made. The errors in these results are less than 5%. For the $^{12}C(\gamma, n)^{11}C$ reaction, the resonance peak lies below 30 MeV, and the yield curve exhibits a slight increases with an increase in the energy.[20,21] The yields of the reactions investigated here appear likely to be strongly energy-dependent. Since ^{7}Be can be formed through competitive reactions on mixed boron targets, except in the irradiation at 30 MeV, the reaction yields were expressed with preference to the per atomic weight of the target element instead of per mole of the target nucleus. The yields are shown in Fig. 2, plotted against the difference in mass between the target and product nuclei at excitation energies of 30, 45 and 60 MeV. For a given energy, the exponential decrease in the reaction yield *versus* the difference in mass is shown. The gradient of the logarithm of the yield against the difference in mass increases as the excitation energy decreases.

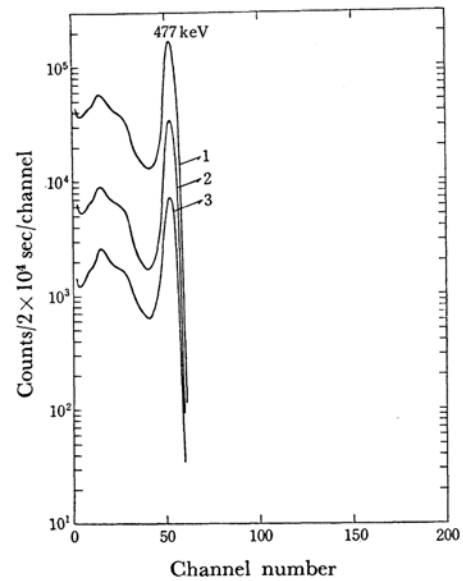

Fig. 1. Gamma-ray spectra of ^{7}Be.
1: Carbon 1.76 g, 32.1 days after irradiation
2: Beryllium 47 mg, 37.5 days after irradiation
3: Boron 77 mg, 49.4 days after irradiation
Irradiation: 60 MeV (45 μA), 3 hr
Measurement: 3″dia.×3″NaI(Tl), source distance=8 cm

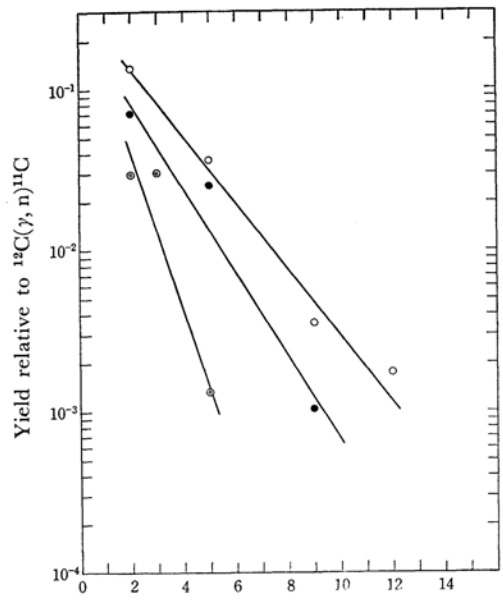

Fig. 2. Relative yield *versus* mass difference between target and product nuclei.
○: 60 MeV ●: 45 MeV ◎: 30 MeV

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

20) W. C. Barber, W. D. George and D. D. Reagan, *Phys. Rev.*, **98**, 73 (1955).

21) B. C. Cook, J. E. E. Baglin, J. N. Bradford and J. E. Griffin, *ibid.*, **143**, 724 (1966).

Walker *et al.*[22] measured the reaction yields for the production of ^{18}F from ^{19}F to ^{32}S targets with 240 MeV bremsstrahlung, and found their exponential decrease when plotted against the number of emitted nucleons.

A similar tendency has been shown in various works of proton-induced spallation reactions,[23-25] and a possible mechanism which would result in this behavior has also been considered. When the total reaction cross-section is distributed among many of the product nuclei, a yield generally favors the product nuclide which is close to the line of the stable nuclei on the isotopic chart. If this is the case, the yield shows the exponential decrease against the difference in mass, as has been determined here. The foregoing presentation may prove useful in a general consideration of photonuclear properties in the energy region up to 60 MeV.

For analytical purposes, the lower limits of detection for beryllium and for boron under the present experimental conditions[26] can be estimated to be 2 and 10 μg respectively. Obviously, these limis of detection will be reduced by a higher bremsstrahlung intensity and by lengthening the time of irradiation.

22) T. G. Walker and W. T. Morton, *Proc. Phys. Soc. (London)*, Vol. LXXV, 4 (1960).

23) S. G. Rudstam, P. C. Stevenson and R. L. Fowler, *Phys. Rev.*, **74**, 442 (1952).

24) S. G. Rudstam, *Phil. Mag.*, **44**, 1131 (1953).

25) R. G. Korteling and A. A. Caretto, Jr., *J. Inorg. Nucl. Chem.*, **29**, 2863 (1967).

26) At the end of a one-hour irradiation with a 60 MeV bremsstrahlung (average beam current: 45 μA), the counting geometry is 10% with a 3'' dia.$\times$ 3'' NaI(Tl) crystal.