Photonuclear Reactions of Gallium and Arsenic with 70-Mev Bremsstrahlung*†

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Radiochemical techniques were utilized in the measurement of yields of photonuclear reactions induced by 70-Mev bremsstrahlung in gallium and arsenic targets. For As targets the yields of As^{74}, Ga^{73}, Ga^{72}, Ga^{68}, Zn^{69}, and Zn^{69m} were determined. In the gallium targets the yields of Zn^{69}, Zn^{69m}, Ni^{65}, and Co^{61} were measured. The integrated cross sections for the indicated photonuclear reactions were estimated from the yields. The integrated cross sections of five $(\gamma,3p3n)$ processes were compared. The relative yields of the Zn^{69} isomeric pair depended upon the process of formation.

INTRODUCTION

YIELDS of photonuclear reactions induced by bremsstrahlung of moderate energy, 40 to 100 Mev, provide information about the processes which occur just above the rather thoroughly studied giant-resonance region. Processes which involve the loss from target nuclei of one to eight nucleons can be observed for the middle atomic weight region. Since competition for additional particle boil-off increases with energy, the cross section for any particular reaction is expected to be a peaked function. The determination of the cross section function for $Ca^{40}(\gamma,3p3n)Cl^{34}$ up to 66 Mev[1] has demonstrated such an expected maximum of about 0.3 millibarn at 50 Mev with a width of 6 Mev at half-height.

The yield of radioactivity, $Y(E_1)$, which is the rate of the photonuclear reaction during an irradiation at a particular bremsstrahlung limiting energy, E_1, is given by the expression

$$Y(E_1)=N\int_{E_t}^{E_1}\sigma(E)P(E,E_1)dE, \qquad (1)$$

where N = the number of target nuclei, $\sigma(E)$ = the cross section as a function of the photon energy, E, $P(E,E_1)$ = the number of photons per cm² per unit time between E and $(E+dE)$ for the limiting bremsstrahlung energy E_1, and E_t = the threshold energy. For two photonuclear processes which peak sharply at the energies $E_{\max}(1)$ and $E_{\max}(2)$, the yield ratio for a limiting bremsstrahlung energy well above the peak energies gives an estimate of the ratio of integrated cross sections. A thin-target bremsstrahlung photon spectrum possesses approximately a $1/E$ dependence.[2] For such a spectrum, the ratio of the yields is given by

$$Y(2)/Y(1)=\left[N_2E_{\max}(1)\int\sigma(2)dE\right]\Big/\left[N_1E_{\max}(2)\int\sigma(1)dE\right]. \qquad (2)$$

In cases for which the $\int\sigma(1)dE$ is available, the integrated cross section for process 2 can then be calculated from the yield ratio. The method does require a reasonably accurate evaluation of the values of $E_{\max}$. This method with $E_1=70$ Mev gave 2.4 Mev-mb for the $Ca^{40}(\gamma,3p3n)Cl^{34}$ compared to 2.6 Mev-mb obtained from the cross-section curve.[1] The standard used was the $Cu^{63}(\gamma,n)Cu^{62}$ process with the cross section reported by Berman and Brown.[3]

Photonuclear yields in the range 140 to 320 Mev by Halpern and co-workers[4–6] and by Wolke and Bonner[7] have indicated a substantial contribution to some moderate-energy processes by high-energy photons. Therefore, yields at moderate energies, from the giant resonance up to 100 Mev, are useful in giving the integrated cross section for the portion under the initial cross-section peak. Yields in this region of energy have been reported by Perlman and Friedlander,[8] Perlman,[9] Holtzman and Sugarman,[10] Edwards and MacMillan,[11] and Schupp and Martin.[12]

EXPERIMENTAL

Either stoichiometric chemical compounds or the pure elements with natural isotopic abundances were irradiated in the 70-Mev bremsstrahlung beam of the Iowa State College synchrotron. In each case two or

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TABLE I. Experimental relative yield determinations for 70-Mev bremsstrahlung.

Target	No. of trials	Reactions compared	Chemical sample	Average yield ratio per target nuclei
$Ca(OH)_2$	2	$\frac{Ca—Cl^{34}\ (32.4\ min)}{Ca—K^{38}\ (7.7\ min)}$	AgCl $K(\phi_4B)$[a]	0.12 ± 0.01
$CaCl_2$	2	$\frac{Ca—K^{38}}{Cl—Cl^{34}}$	$K(\phi_4B)$[a] AgCl	0.21 ± 0.01
NaCl	2	$\frac{Cl—Cl^{34}}{Na—F^{18}\ (112\ min)}$	AgCl PbClF	9.4 ± 0.2
NaCl	2	$\frac{Cl—Si^{31}\ (158\ min)}{Na^{23}—F^{18}}$	K_2SiF_6 PbClF	0.079± 0.001
As, As_2O_3, $Na_2HAsO_4\cdot 7H_2O$	7	$\frac{As—Zn^{69}\ (52\ min)}{As—Zn^{69m}\ (13.8\ hr)}$	$ZnHg(SCN)_4$	0.7 ± 0.1
$Na_2HAsO_4\cdot 7H_2O$	6	$\frac{As—Zn^{69}}{Na—F^{18}}$	$ZnHg(SCN)_4$ PtClF	0.022± 0.006
$Na_2HAsO_4\cdot 7H_2O$	2	$\frac{As—As^{74}\ (17.5\ day)}{Na—F^{18}}$	As_2S_3, As_2S_5 PbClF	220 ±30
As, As_2O_3	1	$\frac{As—Ga^{72}\ (14.2\ hr)}{As—Zn^{69}}$	$Ga(8—HQ)_3$[b] $ZnHg(SCN)_4$	3.3
As, As_2O_3	2	$\frac{As—Ga^{73}\ (5.0\ hr)}{As—Ga^{72}}$	$Ga(8—HQ)_3$[b]	0.51 ± 0.03
As, As_2O_3	2	$\frac{As—Ga^{68}\ (68\ min)}{As—Ga^{72}}$	$Ga(8—HQ)_3$[b]	1.9 ± 0.6
Ga	2	$\frac{Ga—Zn^{69}}{Ga—Zn^{69m}}$	$ZnHg(SCN)_4$	1.5 ±0.1
Ga	3	$\frac{Ga—Ni^{65}\ (2.56\ hr)}{Ga—Zn^{69}}$	$Ni(DMG)_2$[c] $ZnHg(SCN)_4$	0.009± 0.002
Ga	2	$\frac{Ga—Ga^{68}\ (68\ min)}{Ga—Zn^{69}}$	$Ga(8—HQ)_3$[b] $ZnHg(SCN)_4$	220 ± 1
Ga	2	$\frac{Ga—Co^{61}\ (1.65\ hr)}{Ga—Ga^{68}}$	$K_3Co(NO_2)_6$ $Ga(8—HQ)_3$[b]	(7.7 ± 0.8)×10^{-5}

[a] (ϕ_4B) is tetraphenylboron anion.
[b] (8-HQ) is 8-hydroxyquinolate anion.
[c] (DMG) is dimethylglyoxime anion.

more radioactive nuclides were produced simultaneously in the same target, and their yields were compared. By the use of a series of target compounds containing suitable elements, the yield of each process in arsenic has been referred to the yield of the $N^{14}(\gamma,n)N^{13}$ reaction, chosen originally as a standard by Perlman and Friedlander.[8] For gallium the processes were all compared to $Ga^{69}(\gamma,n)Ga^{68}$. The yields of this process given by Perlman and Friedlander,[8] substantially identical at 50 and 100 Mev, were used to give the comparison to $N^{14}(\gamma,n)N^{13}$. The method avoids many experimental difficulties in establishing or reproducing the geometry or radiation flux since the target atoms undergoing the two processes experience identical radiation intensities. It was only necessary to maintain a constant beam intensity or to monitor the beam on a relative basis, so long as the bremsstrahlung limit energy was maintained. In case of fluctuations in the beam intensity, indicated by a recording ionization chamber, the yield of each component activity, Y_i^0, for a particular beam

TABLE II. Relative yields and estimated integrated cross sections of photonuclear reactions studied.

Reaction	70 Mev (this work)	Yield: $N^{14}(\gamma,n)N^{13}=1$ 50 Mev[a]	140 Mev[b]	320 Mev[c]	Estimated E_{max} (Mev)	Estimated $\int\sigma dE$ (Mev-mb) (based on 70-Mev yields)
$Mg^{24}(\gamma,3p3n)F^{18}$	0.042[d]				50	1.9
$Cl^{37}(\gamma,3p3n)Si^{31}$	0.071[e]				47	3.0
$Ca^{40}(\gamma,pn)K^{38}$	0.45				37	15
$Ca^{40}(\gamma,3p3n)Cl^{34}$	0.054				50[f]	2.4
$Ga^{71}(\gamma,pn)Zn^{69,69m}$	0.50[g]			1.2[h]	35	16
$Ga^{71}(\gamma,3p3n)Ni^{65}$	0.003[g]			0.06[h]	63	0.2
$Ga^{69}(\gamma,2\alpha)Co^{61}$	0.0051[g]			0.029[h]	50	0.23
$As^{75}(\gamma,n)As^{74}$	50			65[h]	17	800
$As^{75}(\gamma,2p)Ga^{73}$	0.0081	0.0079	0.018[h]		40	0.30
$As^{75}(\gamma,2pn)Ga^{72}$	0.016	0.011	0.053[h]		50	0.72
$As^{75}(\gamma,3p3n)Zn^{69,69m}$	0.012	$\leqslant$0.0031	0.50[h]		52	0.56
$As^{75}(\gamma,2p5n)Ga^{68}$	0.030	not detected	0.25[h]		57	1.5

[a] See reference 10.
[b] See reference 6.
[c] See reference 4.
[d] From Schupp and Martin (reference 12).
[e] Using the $Na^{23}(\gamma,3p3n)F^{19}$ value (reference 12) and assuming all Si^{31} by this reaction, i.e., $Cl^{35}(\gamma,3pn)$ process negligible.
[f] Observed by Schupp, Colvin, and Martin (reference 1).
[g] Using the $Ga^{69}(\gamma,n)Ga^{68}$ yield of Perlman and Friedlander (reference 8).
[h] Using the $As^{75}(\gamma,n)As^{74}/Cu^{63}(\gamma,n)Cu^{62}$ ratio at 320 Mev of Debs *et al.* (reference 4), $Cu^{63}(\gamma,n)Cu^{62}=35$, and the assumption that (γ,n) yields are independent of energy.

intensity, I^0, was obtained from the observed counting rate at the end of irradiation, C_i, by

$$C_i/\epsilon_i=\lambda_i Y_i^0 e^{-\lambda_i\tau}\int_0^\tau (Y_i e^{\lambda_i t}/Y_i^0)dt, \qquad (3)$$

where λ_i= the decay constant of the component, τ= the irradiation time, and ϵ_i= the efficiency for counting the component "i." The ratio, Y_i/Y_i^0, was set equal to I/I^0, taken from the recorder, and the integral was evaluated graphically.

After an irradiation the target material was first dissolved. Carriers for the desired elements were added to the target solution under conditions which assured exchange among the various possible chemical forms of the elements together with holdback carriers for other induced activities. The desired elements were separated from the solutions by means of radiochemical separations and procedures which were modified for the specific application from the compilation of Meinke.[13] The radioactive species were precipitated as a final step in the radiochemical separation. Precipitates were collected on tared filter papers, dried, and weighed for the evaluation of the chemical yield. The filter papers, bearing the precipitates, were mounted with cardboard backing and cellophane covering for counting. The decay was followed, usually over several half-lives of the activity, by a conventional end-window G-M counter. Corrections for air, window and cover absorption[14] were applied. Sample self-absorption and self-scattering corrections were made following the procedure of Engelkemeir *et al.*[15] for counting samples less than 15 mg/cm^2 thickness or the procedure of Baker and Katz[16] for thick samples. The nuclear properties were taken from the compilation by Way *et al.*[17]

RESULTS AND DISCUSSION

Experimental yield comparisons have been given in Table I. The target materials and the chemical forms of the counting samples have been indicated. The errors reported represent the standard deviations for the number of experiments listed in column 2. Uncertainties in the decay schemes and in the conversion of counting rate ratios to disintegration rate ratios introduce uncertainties which may easily amount to 20–30%. For the Zn^{69} isomeric pair the beta-ray of the ground state was counted, and the analysis of the decay curve provided the yield ratio of the isomeric pair. The formation of Ga^{68} from As^{75} was confirmed not only by the half-life but by the use of magnetic deflection which demonstrated the presence of a positron component.

In Table II the relative yields have been expressed on an isotopic basis, relative to $N^{14}(\gamma,n)N^{13}$. The rather high yields for processes in which Z changes by two or more units and A by 4 or more units at these moderate energies [e.g., $As^{75}(\gamma,2p5n)Ga^{68}$ in comparison to $As^{75}(\gamma,2p)Ga^{73}$] indicate that α-particle emission occurs in the most probable reaction mode.

The integrated cross sections, estimated by Eq. (2), are included in Table II. For these values the

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$Cu^{63}(\gamma,n)Cu^{62}$ process was used for the comparison process since it has been studied more than any other photonuclear process. For the comparison a yield of 35 was taken for $Cu^{63}(\gamma,n)Cu^{62}$, and the cross-section function obtained by Berman and Brown[3] was used. The estimates of E_{max} for the reactions have been included; these values were taken about 10 Mev above the Q for the process plus a reasonable Coulomb barrier for the most likely reaction mode.

Yields reported by Holtzman and Sugarman[10] at 50 Mev for As are included in Table II. Their yields for Ga^{73} and Ga^{72} were comparable to the ones from the present work. At the somewhat lower energies, however, they did not detect the presence of Zn^{69} or Ga^{68}. Yields for 140 Mev and 320 Mev have been estimated from the results reported by Sugihara and Halpern[6] and by Debs *et al.*,[4] respectively. The $As^{75}(\gamma,n)As^{74}$ yield, calculated from the data of Debs *et al.*, agrees with the value from the present work as well as may be expected. Since the (γ,n) processes are mostly associated with the giant resonance in the vicinity of 20 Mev, their yields do not depend upon the bremsstrahlung limit energies which greatly exceed this value. However, the yields of the multiple nucleon reactions have apparently increased strongly with energy. Such results imply that the cross-section functions possess a high-energy tail. Indeed, results of Sugihara and Halpern indicate that $\int \sigma dE$ from 140 to 320 Mev is considerably greater than the area under the peak which is inferred from the present work.

The yields of five $(\gamma,3p3n)$ processes at 70 Mev are included in this work. Such yields are expected to decrease as the Z of the target nuclei increases because of the higher barrier. However, from the results it is apparent that yields are not simple functions of Z. An even-even target nucleus may have a lower density of levels and therefore might have a lower capture probability for photons. However, the yields of the two even-even targets, Mg^{24} and Ca^{40} (the latter being doubly magic), are relatively high. It is noted that the residual nuclei for each of these targets are 0.5–1.0 mass unit lighter than the stable valley, close to the maximum of the yield curve which was described by Halpern *et al.*[5] The very low yield for $Ga^{71}(\gamma,3p3n)Ni^{65}$ may result from the fact that the residual nuclide is 1.3 mass units heavier than the valley of stability and is therefore rather far from the maximum of the yield ridge.

The relative yields of the Zn^{69} isomeric states from Ga^{71} are consistent with the general rule that the isomer requiring the lower spin change from the target in the photonuclear process is favored. The fact that the ratio of the Zn^{69} isomers, Zn^{69}(52 min, $I=\frac{1}{2}$)/Zn^{69m} (13.8 hr, $I=9/2$), produced from $As^{75}(I=\frac{3}{2})$ is less than that produced from $Ga^{71}(I=\frac{3}{2})$ indicates a more complete excitation of states in the more complicated reaction. When the Zn^{69} isomeric states[18] are produced by neutron capture in $Zn^{68}(I=0)$, values for Zn^{69}/Zn^{69m} of 13.6, 6.1, and 2.4, respectively, were observed for thermal, resonance (5.2 kev), and fast (1 Mev) neutrons.

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