Yields of Photonuclear Reactions with 320-Mev X-Rays.* I. Experimental Results

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Several medium weight target elements were irradiated by 320-Mev bremsstrahlung. For each target, relative yields were determined for the production of a number of radioactive nuclides.

INTRODUCTION

HIGH-ENERGY photons are known to eject mesons, nucleons, and heavier nuclear fragments from nuclei. Some of these particles seem to be emitted as a result of the direct action of the photons and some seem to be emitted as evaporation fragments from an excited nucleus. The final unexcited nucleus remaining after all the emissions in a given interaction have taken place is often radioactive. When it is, one can determine its production rate by radiochemical techniques. A study of such production rates proves to be helpful in understanding the processes by which the particle emissions take place. In the experiment to be described, the relative production rates or yields of a large number of nuclides have been measured for several target elements of middle atomic weight exposed to 320-Mev bremsstrahlung radiation.

An examination of the distribution of the observed yields reveals strong similarities between the yield patterns observed in this experiment and in bombardments by high-energy particles. Indeed it appears that some of the chief features of observed yield distributions are to a surprising extent independent of both the nature of the incident particle and its energy. In the following paper these features are discussed in some detail and it is shown that they can be accounted for in a semiquantitative way by assuming that the emission of the last few nucleons by a highly excited nucleus is described by the "evaporation" theory of Weisskopf.

The results given here differ, in part, from the high-energy particle-induced reactions (so-called spallation reactions) in that for x-ray irradiation there occur relatively more reactions involving the emission of just one or two nucleons. These emissions are presumably due to those photons in the 320-Mev bremsstrahlung spectrum that have energies of about 20 Mev. This energy corresponds to the giant resonance for photon absorption.

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EXPERIMENTAL PROCEDURES AND RESULTS

The targets used in this experiment were copper, zinc, gallium, germanium, and arsenic. These elements form a group having consecutive atomic numbers. It was decided to study such a group rather than a single element because a large fraction of the photoreactions were expected to lead to stable iostopes and it was felt that the study of just one target element might not provide enough significant information. This particular group of elements was chosen because the periods and radiations in this region of the isotope chart are exceptionally convenient for counting.

The targets consisted of a few grams of material and were exposed at about 2 feet from the x-ray target of the Massachusetts Institute of Technology synchrotron. A typical exposure lasted for several hours, and at its conclusion, quantitative chemical separations were made of the target element and of a number of elements having lower atomic numbers. The precipitates were counted on a set of end-window Geiger counters whose efficiencies had been determined as a function of source thickness and β energy. The counting was usually continued until the rate was very near that of the background. The initial rates of the various activities present were then determined from an analysis of the time-dependence of the counting rate. Corresponding saturation rates were computed and normalized for x-ray beam intensity, which was measured by the amount of 12.9-hr Cu^{64} activity in a standard copper foil irradiated together with the target. After correcting for chemical efficiencies, counting efficiencies and branching ratios, it is possible to express the data in the form of "yields." A yield is the production rate of a given radio-nuclide per g/cm^2 of target element when a standard amount of x-ray energy is being passed through the target per unit time.

The yields determined in this way are listed in Table I in arbitrary units. A number of remarks should be made about the interpretation of the data in this table.

All of the targets except arsenic have more than one isotope. This fact usually makes the assignment of parent isotopes for observed radio-nuclides uncertain. An exception to this ambiguity occurs when the radio-nuclide can be produced by a (γ,n) reaction. For example, the yield of Cu^{62} in a copper bombardment is due either to a (γ,n) reaction on Cu^{63} or a $(\gamma,3n)$

TABLE I. Relative production rates of observed radio-nuclides per g/cm² of target element through which a standard amount of x-ray energy is passed per unit time.

Target	Observed nuclide	Yield	Target	Observed nuclide	Yield
Arsenic	As^{74}	1050	Gallium	Zn^{69}	16
	As^{72}	100		Cu^{67}	1.7
	Ga^{73}	1.3		Cu^{64}	13
	Ga^{72}	3		Cu^{61}	3.2
	Ga^{68}	13		Ni^{66}	0.2
	Zn^{72}	0.1		Ni^{65}	0.8
	Zn^{69}	1.9		Co^{61}	0.4
	Cu^{67}	0.3		Co^{55}	0.09
	Cu^{64}	2.5		Mn^{56}	0.4
	Cu^{61}	0.75	Zinc	Zn^{63}	540
	Ni^{66}	0.016		Zn^{62}	34
	Ni^{65}	0.11		Cu^{67}	13
	Co^{61}	0.17		Cu^{64}	22
	Co^{55}	0.01		Cu^{62}	73
Germanium	Ge^{75}	130		Cu^{61}	20
	Ge^{69}	310		Ni^{66}	0.16
	Ga^{73}	26		Ni^{55}	0.39
	Ga^{72}	18	Copper	Cu^{64}	270
	Ga^{68}	20		Cu^{62}	470
	Zn^{69}	3.8		Cu^{61}	83
	Cu^{67}	1.2		Ni^{57}	0.83
	Cu^{64}	3.6		Co^{61}	3
	Cu^{62}	5.2		Co^{55}	0.4
	Cu^{61}	2.3		Mn^{56}	1.4
Gallium	Ga^{68}	600		Mn^{52}	0.6
	Ga^{66}	18		Cr^{49}	0.24

reaction on Cu^{65}. But measured (γ,n) yields are roughly twenty times larger than $(\gamma,3n)$ yields, so that it is quite certain that almost all of the Cu^{62} comes from the parent Cu^{63}. Assuming that all of the observed Cu^{62} does indeed come from Cu^{63}, it is possible, by taking the abundance of Cu^{63} into account, to convert the yield in the table into a more useful quantity. That is, one can obtain a yield per g/cm² of *parent* isotope instead of a yield per g/cm² of target element.

Unfortunately, for yields of reactions more complicated than the (γ,n) reaction, the proper assignment becomes less apparent and in fact rather uncertain. The general problem of yield assignments is connected with that of the interpretation of the yields and is discussed in the following paper.

It should be pointed out here that the yields in the table are not all necessarily primary yields because the observed radio-nuclides are not all "shielded" nuclides. In spite of this, most of the measured yields are probably predominantly primary because the experimental results indicate that the yields tend to decrease very rapidly for nuclides away from the stable valley of the nuclides. This would imply that the contributions of any "fed-in" yield are smaller than the original yield of any radio-nuclide being considered.

In addition to possible errors of the type just considered, that can come about from misinterpretations of the data, the experiment suffers from a number of more conventional sources of error. An observed yield of 0.1 corresponds, for reasonably short periods and reasonable decay schemes, to an initial counting rate of about ten counts a minute. For most of the data the yields are larger than 0.1 and the statistical errors on the observed counting rates are quite small. But the "initial rates" are not unambiguously determined except for samples decaying with only one period. The back-extrapolation of counting rate data to determine the initial rates is rather sensitive to the assumed periods and somewhat more sensitive to statistical errors than one might at first suspect. In every case the analysis of the decay curves was based on the half-lives compiled in *Nuclear Data*.[1] Most of the initial rates determined from the decay curves are probably good to ± 20 percent although some of the lower rates are not known as well. The reliability of the initial rate determinations was tested empirically by having a number of people independently analyze some of the decay curves into component periods.

The "initial rate" data were converted into "saturation rates" without much additional error, but the corrections for Geiger counter efficiencies and the uncertainties in assumed branching ratios tend to increase the probable error of a typical yield datum to about ± 50 percent. This would be a large error in some experiments, but in the present experiment the yields cover a range of over 10^5 and an error of even a factor of 2 represents only $\frac{1}{16}$ of this range on a logarithmic scale.

In certain cases, activities produced by either fast or slow neutrons in the synchrotron chamber could have contributed to the yields in the table. But measurements of the slow and fast neutron fluxes at the position of our targets showed that contributions to the observed yield by neutron-induced reactions were negligible. It should also be mentioned that the targets were kept thin enough so that there was no appreciable distortion of the x-ray spectrum as the x-ray beam passed through the target.

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APPENDIX OF CHEMICAL PROCEDURES

The target material is dissolved, and a standard amount of carrier solution of each of the elements to be separated is added. The mixture is then processed according to one of the procedures outlined below, the final precipitates being weighed before being counted. The chemical yield of any given element is calculated by determining the weight of precipitate obtained from a known amount of the carrier solution.

Specific Separations

(1) *Chromium, Manganese, Cobalt, Nickel, and Copper from Copper*

Copper turnings are used as the target material. They are dissolved in HCl-H_2O_2 and carriers are added. The copper is then reduced to the metal with powdered iron, and centrifuged from the solution. After oxidation with H_2O_2 the iron is extracted from

[1] *Nuclear Data*, National Bureau of Standards Circular 499 (U. S. Government Printing Office, Washington, D. C., 1950).

the solution with dichloroethyl ether and discarded. A treatment with NaOH-Na_2O_2 precipitates nickel, cobalt, and manganese, and extracts the chromium in the form of chromate, which is precipitated with barium. The manganese is left in the form of MnO_2, and an extraction with concentrated HNO_3 separates it from nickel and cobalt. The MnO_2 is dissolved in HCl, reprecipitated with Na_2O_2, extracted with HNO_3, filtered and dried at 110°C. Nickel is precipitated with dimenthylglyoxime and then cobalt is precipitated with α-nitroso β-naphthol reagent.

(2) *Chromium, Manganese, Cobalt, Nickel, Copper, Zinc, and Gallium from Gallium*

Acid-soluable gallium oxide prepared by the decomposition of the nitrate at 250°C is used as the target. The target is dissolved in concentrated HNO_3 and carriers are added. Then the nitrates are converted to chlorides by fuming with HCl. Gallium is extracted with dicholoroethyl ether from 6*N* HCl solution and back extracted into water; an aliquot is precipitated with 8-hydroxyquinoline ("oxine") for counting.

Copper is reduced to metal with granulated tin. Zinc and chromium are separated by treatment with Na_2O_2. The chromium as chromate is precipitated with barium, and the zinc is precipitated with mercuric thiocyanate reagent. Manganese, cobalt, and nickel are separated as in Part (1).

(3) *Manganese, Cobalt, Nickel, Copper, Zinc, and Gallium from Germanium and Arsenic*

Germanium oxide or arsenious oxide are used as targets. The oxide is dissolved in a minimum of concentrated NaOH, after which the solution is made slightly acid with HCl. The carriers are added and precipitated with excess potassium ferrocyanide. An aliquot of the supernatant solution may be precipitated with H_2S to obtain germanium or arsenic activities.

The precipitated ferrocyanides are fumed with H_2SO_4 and the residue is dissolved in HCl. Copper is precipitated with granulated tin, which also reduces iron to the divalent state. Gallium is extracted with dichloroethyl ether, back extracted into water and precipitated with oxine.

Iron is then oxidized with H_2O_2, extracted with dichoroethyl ether and discarded. Zinc is separated by treatment with Na_2O_2, and precipitated for counting with mercuric thiocyanate. Manganese, cobalt, and nickel are separated as in Part (1).

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Yields of Photonuclear Reactions with 320-Mev X-Rays. II. Interpretation of Results*

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It is convenient to separate the yields of radio-nuclides obtained in the work described in the preceding paper into two groups. The first group consists of the yields of nuclides which are only one or two mass units lighter than the target. All of the yields in this group, and especially those corresponding to (γ,n) reactions, are relatively large. They are due mainly to photons in the giant resonance region ($h\nu\sim 20$ Mev) of the x-ray spectrum, and account for most of the nuclear events produced in medium-weight targets by 320-Mev x-rays.

The yields of those radio-nuclides which are more than a few mass units lighter than the target are in many ways more interesting than the ones in the first group. They are found to exhibit a simple pattern very similar to those obtained in particle-induced high-energy reactions. It is shown that such patterns are at least qualitatively consistent with models of high-energy nuclear reactions in which the last few particles emitted from a struck nucleus, leave by "evaporation." Indeed the evaporation of these last particles exerts so strong an influence on the form of the observed yield pattern, that it becomes very difficult to say anything about either the nature of the original nuclear events or the emission of the first few particles on the basis of a study of yield patterns.

Finally, a rough quantitative comparison is made of the yields of radio-nuclides and the reported yields of neutrons and other particles emitted from medium-weight nuclei irradiated with 320-Mev x-rays.

IN the preceding paper, a number of relative yields are reported for the formation of radio-nuclides from the irradiation of medium weight elements by high-energy x-rays. In the following two sections, the yields for radio-nuclides which are more than a few mass units lighter than the target are shown to fall into simple patterns. These patterns are described quantitatively in Sec. 3 and it is shown that they are evidence for a major role of nuclear evaporation in high-energy photoreactions. It is found in Sec. 4 that the pattern described here is quantitatively very similar to patterns constructed from data of high-energy particle-induced reactions. An attempt is made in Sec. 5 to account for all of these patterns in a semiquantitative way by assuming that the last few particles ejected in a high-energy reaction are evaporated. Section 6 deals with those photoreaction yields in which the observed nuclide is only a few mass units lighter than the target. Finally, in Sec. 7, the data of this experiment are compared to data for neutron production and meson production by high-energy x-rays.

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1. YIELDS FROM ARSENIC

When the work described in the preceding paper was undertaken, only a few measurements had been made of the distribution of the residual nuclides in high-