Excitation Functions for Proton-Induced Reactions with Copper*

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Proton bombardment at energies up to 100 Mev have been carried out in the Harvard University cyclotron using targets of $Cu^{63}O$ and $Cu^{65}O$. Absolute cross sections of the (p, n), $(p, 2n)$, (p, pn), and $(p, p2n)$ reactions of Cu^{63} and the $(p, 3n)$, $(p, 4n)$, (p, pn), $(p, p3n)$, and $(p, p4n)$ reactions of Cu^{65} have been determined using the $C^{12}(p, pn)C^{11}$ and $Al^{27}(p, 3pn)Na^{24}$ reactions to monitor the proton beam. An estimate of the ratio of level densities for odd-odd and even-even nuclei is made.

I. INTRODUCTION

A SERIES of bombardments of the separated isotopes of copper[1] with 100-Mev protons has been carried out in the internal scattered beam of the Harvard cyclotron for the purpose of studying the cross section for formation of various isotopes of copper and zinc as a function of energy. The experimental results show fair agreement with the predictions of the statistical model[2] at low energies and are in qualitative agreement with the predictions of the Serber transparency model[3] at high energies.

II. EXPERIMENTAL TECHNIQUES

A. Target Makeup

A modification of the stacked foil technique was used to determine the excitation functions. The separated copper isotopes were received in the form of the oxide.[1] About 25 milligrams of the oxide powder was packed into a thin, flat aluminum capsule having an inside diameter of $\frac{5}{16}$ inch and a depth of about 0.01 inch. Four to eight of these capsules were interspaced with absorbers and bombarded at initial energies of 100 and 75 Mev. The bombardment periods were short compared to the half-lives of the isotopes investigated. As a check on this method the results obtained using such targets made with ordinary copper oxide were compared with those obtained using 0.001-inch copper foil. No significant differences were noted.

The bombardments were made in the internal scattered beam of the Harvard cyclotron with 180° focusing in the cyclotron magnetic field. The details of this method have been discussed by Hintz and Ramsey.[4] The energy resolution of this method is determined chiefly by the diameter of the target and can be approximated at any energy by

$$\Delta E \sim \Delta E_0 \cdot E_0/E,$$

where E_0 refers to the initial energy. If E_0 is 70 Mev, ΔE_0 is about 1 Mev.

B. Determination of Cross Sections

Absolute cross sections were estimated for all reactions, except those involving Cu^{62}, by placing aluminum foils in the target stacks and using the $Al^{27}(p, 3pn)$-Na^{24} reaction to monitor the proton beam. The cross section for this reaction was measured by comparing it with the $C^{12}(p, pn)C^{11}$ reaction.[5]

No chemical separations were made for the 10.5-minute Cu^{62} activity. The copper oxide was counted through a 370 milligram per square centimeter aluminum absorber to stop any 20-minute C^{11} activity formed from the oxygen, and the relative yield of 10-minute activity was determined from the decay curve. When the target was $Cu^{63}O$ this activity should be almost entirely due to Cu^{62} below 55 Mev. Above this energy there is some contamination by the 10-minute Cu^{59} activity. The slight peak in the $Cu^{63}(p, pn)$-Cu^{62} excitation curve (Fig. 1) at about 80 Mev is probably caused by the $Cu^{63}(p, p4n)Cu^{59}$ reaction. Its cross section is probably less than the corresponding reaction for Cu^{65} because of its higher threshold, but its effect on the (p, pn) excitation curve cannot be estimated since it decays by emitting a positron of unknown energy and all counting was done through

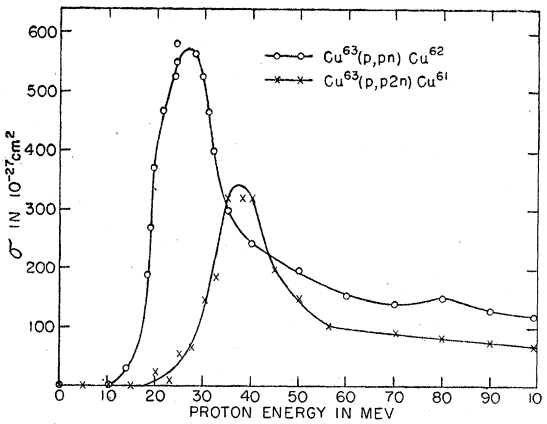

FIG. 1. Cu^{61} and Cu^{62} from Cu^{63}.

* Assisted by the joint program of the U. S. Office of Naval Research and the U. S. Atomic Energy Commission.

[1] Obtained from Oak Ridge National Laboratory, Oak Ridge, Tennessee.

[2] J. M. Blatt and V. F. Weisskopf, Massachusetts Institute of Technology Technical Report No. 42, May 1, 1950 (unpublished).

[3] R. Serber, Phys. Rev. **72**, 1114 (1947).

[4] N. M. Hintz and N. F. Ramsey, Phys. Rev. **88**, 19 (1952).

[5] Aamodt, Peterson, and Phillips, Phys. Rev. **88**, 739 (1952).

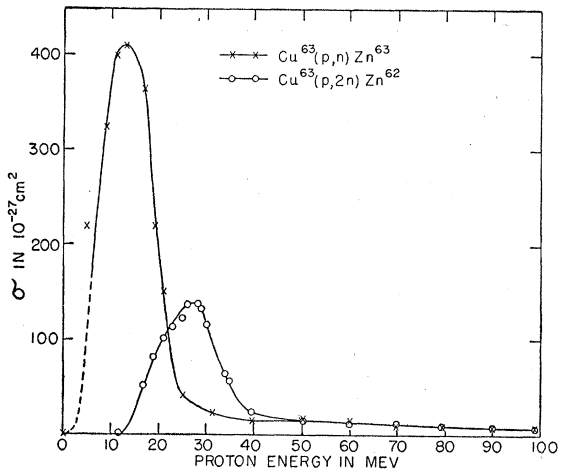

FIG. 2. Zn^{62} and Zn^{63} from Cu^{63}.

an absorber. The cross section for the $Cu^{63}(p, pn)Cu^{62}$ reaction was obtained by comparing it with the Cu^{63}-$(p, 2n)Zn^{62}$ reaction at 26 Mev. The 9.5-hour Zn^{62} decays to Cu^{62} by electron capture (~90 percent) and by a 0.65-Mev positron. Because of the low efficiency of a Geiger tube for x-rays, the observed activity of Zn^{62} when counted through a 370 milligram per square centimeter aluminum absorber consists primarily of the 3-Mev positron of Cu^{62}. At 26 Mev the only other activity observed through the absorber was the 2.4-Mev positron from the 38-minute Zn^{63}, but its activity was very weak compared to the Cu^{62} and Zn^{62} activities. Since these two activities can be counted by the same radiation, a very good estimate of their relative yields can be made from the decay curve. The $Cu^{56}(p, p3n)$-Cu^{62} cross section was then estimated by comparing it directly with the $Cu^{63}(p, pn)Cu^{62}$ cross section.

C. Counting Techniques

The counting samples were mounted on platinum, the active area having about the same diameter as the monitor foils. After drying these were counted by an end window Tracerlab type TGC-d Geiger tube. The monitor foils were mounted in a similar fashion and counted in the same geometry. The observed counting rates were then subjected to the following corrections: (1) counts loss due to counter dead time; (2) background activity; (3) absorption by air and counter window; (4) self-absorption and scattering in the sample material; (5) k capture to electron ratios in the various isotopes; (6) decay of the sample from the bombardment period to the counting period.

No correction was made for backscattering from the sample mounting. Since the targets and monitors were always mounted on the same backing material and since the saturation backscattering is largely independent of energy[6] this effect should cancel in calculating the cross section. Recent work[7] has shown that there may be some difference in the backscattering coefficient for electrons and positrons. This correction was not included here. Since C^{11}, a positron emitter, was used to determine the cross section for Na^{24}, an electron emitter, which was used to determine the cross section of a number of positron emitters, this effect should be largely compensated for. However, owing to differences in backing materials the inclusion of such corrections may increase the values presented here by about 5 percent.

D. Chemical Separations

Copper: The copper oxide was dissolved in a minimum of HNO_3, boiled to remove any oxides of nitrogen, diluted to about 20 ml and transferred to an electrolysis cell. Two drops of concentrated HNO_2 and eight drops of concentrated H_2SO_4 were added and the solution plated to exhaustion in about $\frac{1}{2}$ hour with a current of

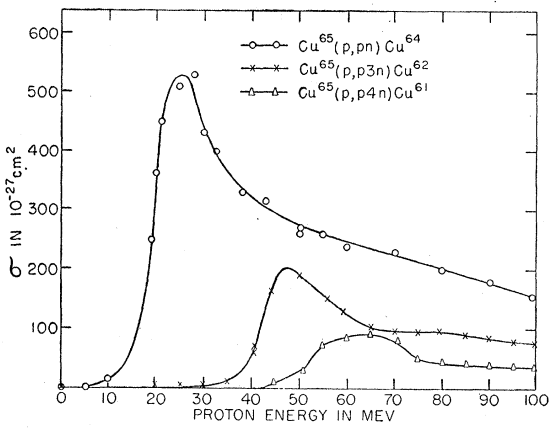

FIG. 3. Cu^{61}, Cu^{62}, and Cu^{64} from Cu^{65}.

25 to 30 milliamperes. The plates were removed, washed in alcohol, dried in air and were ready to count.

Zinc: The copper oxide was dissolved in a minimum amount of concentrated HCl and H_2O_2 to which 10 mg of Zn carrier had been added. The solution was boiled to remove the excess H_2O_2, then made 1N with HCl, and the copper precipitated with H_2S. The filtrate was boiled to remove excess H_2S, and scavanged with ferric hydroxide. The pH was adjusted to 3 and the Zn precipitated with H_2S. The precipitate was washed with 0.01N acid saturated with H_2S, dried under a lamp and counted. To obtain the chemical yield it was ignited to the oxide and weighed.

III. DISCUSSION

The cross section of the reactions studied are plotted as a function of the proton energy in Figs. 1 through 4.

At low energies (E~30 Mev) nuclear reactions are believed to proceed by the capture of the incident

[6] B. P. Burtt, Nucleonics **5**, 2, 28 (1949).

[7] H. H. Seliger, Phys. Rev. **88**, 408 (1952).

particle to form a compound nucleus in an excited state and then evaporate nucleons according to the statistical theory.[2] At high energies ($E\sim100$ Mev) nuclear reactions may be explained by Serber's theory[3] of nuclear transparency. According to this theory the mean free path of the bombarding particle is large compared to the nuclear radius and the particle interacts with the target nucleus by individual nucleon-nucleon collisions inside the nucleus. The average transfer of energy for a 100-Mev incident particle is about 25 Mev per collision. Because of the large mean free path there is a large probability that the incident particle may make only one or two collisions before the particle escapes with a large part of its initial energy. The net results of the process are these: A high-energy nucleon enters the nucleus. One or more neutrons and protons with relatively high energies emerge leaving a residual nucleus in an excited state. This excited nucleus

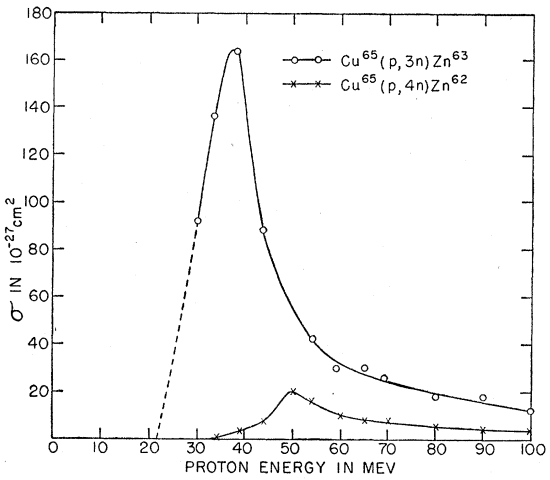

FIG. 4. Zn^{62} and Zn^{63} from Cu^{65}.

then evaporates other nucleons according to the statistical theory.

Considering the rather wide energy spread inherent in the bombardment method below 20 Mev, the Cu^{63}-$(p, n)Zn^{63}$ reaction agrees very well with the statistical theory (Fig. 2, Table I). At 100 Mev the (p, n) cross section is 8 millibarns. According to the Serber model for high energy nuclear reactions, this required that about one percent of the protons entering the nucleus knock out high-energy neutrons leaving the residual nucleus with insufficient excitation energy to evaporate another nucleon. For Zn^{63} this excitation energy must be less than 6 Mev.

At 26 Mev the experimental value for the $Cu^{63}(p, pn)$-Cu^{62} reaction is about four times that for the Cu^{63}-$(p, 2n)Zn^{62}$ reaction (Figs. 1 and 2). This ratio agrees with that observed by Ghoshal[8] although our values for the cross sections are somewhat lower. If the statistical model is valid at this energy, the unusually low yield of the $(p, 2n)$ reaction indicates a relatively low level density for the even-even Zn^{62} nucleus. This appears to be confirmed by the unusually low cross section for the $(p, 4n)$ reaction for Cu^{65} as compared to the $(p, 3n)$ reaction (Fig. 4).

It is not possible to obtain the ratio of the (p, pn) to $(p, 2n)$ reactions for Cu^{65} since Zn^{64} is a stable nucleus. However, the excitation function for the Cu^{65}-$(p, pn)Cu^{64}$ reaction is very similar to the corresponding reaction for Cu^{63} having a peak value of 540 millibarns at 26 Mev (Fig. 3). By analogy to the Cu^{63} reactions the $Cu^{65}(p, 2n)Zn^{64}$ reaction would be expected to have a cross section of about 150 millibarns at this energy. It could not be greater than 250 millibarns or the total cross section including only the observable reactions will be greater than the cross section for the formation of the compound nucleus. In view of this the level density of Zn^{64} must be low compared to that of C^{64}.

Since the $Cu^{65}(p, 3n)Zn^{63}$ reaction must involve the formation of an excited Zn^{64} nucleus, a low-level density for this nucleus would mean a correspondingly low cross section for the $(p, 3n)$ reaction. Experimentally, the $(p, 3n)$ reaction has a peak value of 170 millibarns (Fig. 4) which is about the same as that expected for the $(p, 2n)$ reaction. This is contradictory to the previous conclusion concerning the Zn^{64} level density.

Very little is known about the level densities of the even mass nuclei. According to Harris *et al.*[9] and Hurwitz and Bethe,[10] odd-odd and even-even nuclei have comparable level densities at excitation energies of the order of their neutron binding energies. The level densities are not determined from the ground state but from some characteristic higher state. If the level density then increases according to the formula given by Blatt and Weisskopf,[2] the ratio of the level densities of the two nuclei for high excitation will be given by

$$\omega_{oo}/\omega_{ee}\sim \exp 2a^{\frac{1}{2}}[(E-\epsilon_{oo})^{\frac{1}{2}}-(E-\epsilon_{ee})^{\frac{1}{2}}],$$

where ω_{oo} and ω_{ee} are the level densities of the odd-odd and even-even nuclei, respectively, E is the excitation energy, and ϵ is the energy of the characteristic level above the gound state. If we approximate the difference

TABLE I. Reaction cross sections calculated using constants given by Blatt and Weisskopf—$R=(1.3\times10^{-13})A^{\frac{1}{3}}$cm.

Reaction	Proton energy	σ in millibarns $\frac{\omega_{oo}}{\omega_{ee}}=4$	$\frac{\omega_{oo}}{\omega_{ee}}=28$	$\frac{\omega_{oo}}{\omega_{ee}}=56$	Exp
$Cu^{63}(p, n)Zn^{63}$	12 Mev	370[a]			408
$Cu^{63}(p, 2n)Zn^{62}$	26	247	133	94	138
$Cu^{63}(p, pn)Cu^{62}$	26	294	506	560	570
$Cu^{65}(p, pn)Cu^{64}$	26	321	555	630	525

[a] Calculated using $\omega_{oe}=\omega_{eo}$.

[8] S. N. Ghoshal, Phys. Rev. **80**, 939 (1950).

[9] Harris, Muehlhause, and Thomas, Phys. Rev. **79**, 11 (1950).

[10] H. Hurwitz and H. A. Bethe, Phys. Rev. **81**, 898 (1951).

TABLE II. Q Values for two reactions which may proceed by triton emission.

Reaction	$-Q$, Mev
$Cu^{63}(p, p2n)Cu^{61}$	18.8
(p, dn)	(p, dn)
(p, H^3)	10.4
$Cu^{65}(p,p3n)Cu^{62}$	28.2
$(p, d2n)$	26.0
(p, H^3n)	19.8

in the characteristic levels of the even-even and odd-odd nuclei by the difference in their neutron binding energies (~5 Mev for Cu^{62} and Zn^{62}), it can be seen that the ratio of level densities is large for low excitation energies and small for higher ones. For energies of about 10 Mev, the ratio is of the order of 10. Such an energy dependence would provide a possible explanation for the high cross section for the $(p, 3n)$ reaction as well as the low cross section for the $(p, 2n)$ reaction. In the $(p, 2n)$ reaction the even-even nucleus is the final one and must have an excitation energy of less than about 8 Mev or it will evaporate additional nucleons. This means a relatively low-level density compared to the odd-odd nucleus and consequently a low yield for the $(p, 2n)$ reaction. In the $(p, 3n)$ reaction the even-even nucleus is an intermediate one and must have an excitation energy of more than 12 Mev if it is to evaporate another neutron. This higher excitation energy means that the level density is relatively high and consequently there will be a relatively higher yield of this excited nucleus. The next step involves competition between an odd-even and an even-odd nucleus which have approximately the same level densities.[2] In this situation the emission of a neutron will be the preferred process resulting in a high yield for the $(p, 3n)$ reaction.

These reactions are also dependent on the level density of Ni^{62} and Ni^{64} since the $(p, 2p)$ reaction is a competing one in each case. Since these nickel isotopes are even-even as well as magic number nuclei, their level density should be quite low. Actually, these reactions are not very sensitive to the level density of these nuclei, as the probability of evaporating two protons is small.

It is possible to use the experimental values of the (p, pn) and $(p, 2n)$ reactions to obtain an approximation of the average ratio of level densities for the odd-odd and even-even nuclei below the nucleon binding energy. The values appearing in Table I were calculated according to the statistical model using the following three assumptions. First, it was assumed that neutrons and protons were the only particles involved. The inclusion of alpha particles would decrease the values by a small amount but would have little effect on the ratio of the (p, pn) to $(p, 2n)$ reactions. The evaporation of deuterons would increase the (p, pn) reaction but this type of process appears to be negligible.[11] Second, it was assumed that ω_{oe} was equal to ω_{eo}. The agreement between the observed and calculated cross sections for the (p, n) reaction seems to justify this assumption. Third, it was assumed that there was a constant ratio between ω_{oo} and ω_{ee}. This is certainly not correct according to the previous agrument, but it does give an indication of the level density ratio at low excitation energies. The cross sections for the (p, pn) and $(p, 2n)$ reactions are calculated using three values of ω_{oo}/ω_{ee}. A value slightly larger than 28 will give the observed ratio of (p, pn) to $(p, 2n)$ reactions. The calculated values for the cross sections are then in fair agreement with the experimental results.

The observed ratio of the (p, pn) and $(p, 2n)$ reactions at low energies makes the even higher ratio at 100 more understandable. At high energies the reaction should occur primarily by an initial knockout process whereby the incident particle makes only one or two collisions inside the nucleus, resulting in the ejection of one high-energy neutron or proton and leaving a residual nucleus with a few Mev of excitation energy which permits it to evaporate one more nucleon. Since the incident particles are protons, a high-energy neutron can only be produced by a proton-neutron collision inside the nucleus while a high-energy proton can be produced by either a proton-neutron or a proton-proton collision. Therefore the ratio of protons to neutrons in the initial step should be about 2 to 1. Then, if the two residual nuclei (Cu^{63} and Zn^{63}) evaporate secondary neutrons and protons in the same ratio as at lower energies, the (p, pn) reaction should be favored over the $(p, 2n)$ reaction by roughly a factor of 10. The experimental value is about 15. However, this is high due to the contribution of Cu^{59} at this energy.

The previous discussion has neglected competing reactions involving the emission of deuterons and tritons. The ratio of these particles to single nucleons should be small because of their greater difficulty in penetrating the Coulomb barrier and their small probability of existence at the nuclear surface. In general, this appears to be true for elements of intermediate and heavier atomic weight and for fairly high excitation energies.[11] However, below the threshold for the individual nucleon reaction the corresponding processes of this type must account for the total yield of a particular reaction.

The energy resolution of this bombardment method is not sufficiently narrow to distinguish between the threshold of such reactions as (p, d) and (p, pn). However, reactions involving tritons have thresholds about 6 Mev below the corresponding reaction involving a deuteron and a neutron. Thus, there is a narrow region where a particular reaction can proceed only by the emission of a triton. The Q values calculated from mass[12] and radioactivity[13] data for two reactions which

[11] P. J. van Heerden (private communication).

[12] Collins, Nier, and Johnson, Phys. Rev. **86**, 408 (1952).

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

may proceed by emission of tritons are given in Table II. The Cu^{65} targets did show a yield of Cu^{62} below 25 Mev, but this can be largely accounted for by the Cu^{63} contamination. The maximum yield of the (p, H^3n) reaction in this region is of the order of 1 millibarn. The Cu^{63} targets have a cross section of 1.2 millibarns for the production of Cu^{61} at 15 Mev. The energy uncertainty here is ± 2.5 Mev so this may be partly due to the (p, dn) reaction. At 10 Mev the cross section is about 0.1 millibarn and should be entirely due to the (p, H^3) reaction.

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The Radiations of U^{240} and Np^{240}†

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The beta and gamma radiations of U^{240} and Np^{240} have been examined with magnetic lens and scintillation spectrometers. $U^{240}(t_{\frac{1}{2}}=14.1\pm0.2$ hours) emits a single beta-ray group of maximum energy 0.36 Mev. No gamma rays were observed. $Np^{240}(t_{\frac{1}{2}}=7.3\pm0.3$ minutes) emits four beta-ray groups with maximum energies of 2.156, 1.59, 1.26, and 0.76 Mev and relative intensities of 52, 31, 11, and 5.4 percent, respectively. Gamma rays having energies of 1.40, 0.90, and 0.56 Mev were observed. These data can be fitted into a simple decay scheme. On the basis of cycles involving the total decay energies of these isotopes, plus published data on alpha- and beta-transition energies and photoneutron thresholds of related nuclides, the binding energy of the last neutron in U^{240} is calculated to be 5.92 ± 0.15 Mev, and that of the last neutron in Np^{240} to be 4.98 ± 0.15 Mev.

INTRODUCTION

THE isotopes U^{240} and Np^{240} were found by Hyde and Studier[1] in uranium which had been irradiated at high neutron flux in a pile, the U^{240} having been produced by the neutron capture sequence $U^{238}(n,\gamma)U^{239}(n,\gamma)U^{240}$. The U^{240} and its daughter Np^{240}, both found to be beta emitters, were identified by standard radiochemical techniques. The half-lives were reported as 14 ± 2 hours and 7.3 ± 0.3 minutes, respectively. A later report[2] from the same laboratory gave a revised U^{240} half-life of 17 hours. Further characterization of these isotopes was limited by the low attainable specific activity of the U^{240} and by the presence of a relatively large amount of U^{237} which was also produced during the irradiation by the reaction $U^{238}(n,2n)U^{237}$.

With the availability at this laboratory of high-neutron-flux devices and of the means of retrieving portions of target materials placed in or near them, it has been possible to obtain U^{240} sources of sufficient strength for beta-ray spectrometry and without serious U^{237} interference. This report describes the results of an investigation of the radiations of the U^{240}-Np^{240} chain, conducted on a series of such sources.

APPARATUS AND SOURCE PREPARATION

The beta counting for measurement of half-life and for verification of parent-daughter relationship of the observed uranium and neptunium activities was done with continuous-flow methane gas proportional counters. These counters have a dead-time loss of about 1 percent at 10^5 counts per minute so that sample decay can be followed through factors of the order of 10^4 in counting rate.

The beta-ray spectrum of the U^{240}-Np^{240} equilibrium mixture was measured with a magnetic lens spectrometer.[3] The baffles were adjusted to give a resolution of 3.5 percent. The electron detector was an end-window Geiger tube with a 0.5-in. diameter aperture and a 2.4 mg/cm² mica window having a low energy cutoff at about 31 kev.

The gamma-ray data were obtained with a scintillation spectrometer consisting of a NaI(Tl) crystal attached to an RCA type 5819 photomultiplier. A 1-in. aperture lead collimator was placed between the source and the crystal. Two different scintillation crystals were used: a large one, 2 in. in diameter and 2 in. high, for examination of high-energy gamma rays and general coverage of the entire spectrum, and a smaller one, about 0.25 in. thick, for more detailed examination of the low-energy portion of the spectrum. The pulses were sorted with a ten-channel analyzer.

The uranium activity was isolated from the irradiated

† Work done under the auspices of the U. S. Atomic Energy Commission.

[1] E. K. Hyde and M. H. Studier, Argonne National Laboratory Reports ANL-4143, April 15, 1948, and ANL-4182, August 4, 1948 (unpublished).

[2] Studier, Magnusson, Siddall, and Huizenga, Argonne National Laboratory Report ANL-4667, May 1, 1951 (unpublished).

[3] L. M. Langer, Phys. Rev. **77**, 50 (1950).