Two New Promethium Isotopes; Cross Sections of Some Samarium Isotopes for 14.8-Mev Neutrons*

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When highly enriched samples of Sm^{152} and Sm^{154} are irradiated with 14.8-Mev neutrons, activities having half-lives of 6.5±0.5 min and 2.5±0.5 min are observed. On the basis of yields and cross-bombardments, these are assigned to new isotopes Pm^{152} and Pm^{154}, respectively. Cross-section measurements at 14.8 Mev for $(n,2n)$, (n,p), and (n,α) reactions of samarium are reported, the experimental values being as follows: $Sm^{144}(n,2n)$, 1200±300 mb; $Sm^{154}(n,2n)$, 1500±300 mb; $Sm^{152}(n,p)$, 3.7±0.2 mb; $Sm^{154}(n,p)$, 3.5±0.2 mb; $Sm^{152}(n,\alpha)$, 10±2 mb; and $Sm^{154}(n,\alpha)$, 9±3 mb. While the $(n,2n)$ cross sections are within an order-of-magnitude agreement with statistical evaporation theory, the experimental values for the (n,p) and (n,α) cross sections are several orders of magnitude larger than those calculated from statistical evaporation theory based on the compound nucleus model.

INTRODUCTION

LEVKOVSKII[1] has called attention to the relative variation in (n,p) and (n,α) cross sections (for 14-Mev neutrons) with mass number, A, at constant atomic number, Z. In the cases examined, many of which were taken from the results of Paul and Clarke,[2] the relative cross sections for these reactions decrease by factors of 2, 4, or 8 as A increases, for a given Z. The few exceptions noted were attributed to errors arising from experimental difficulties. This variation in cross section is more pronounced in light elements.

If, instead of considering reactions involving charged particles, one examines the variation with A of $(n,2n)$ cross sections at 14 Mev, one again observes a variation at constant Z, but in this case the cross section usually increases with increasing A. In eight pairs of $(n,2n)$ reactions listed by Paul and Clarke,[2] five show a definite increase in cross section with increasing A; $Ag^{107}-Ag^{109}$, $Br^{79}-Br^{81}$, and $Te^{128}-Te^{130}$ being the three exceptions. Later work[3] has shown that Ag^{109} has a larger $(n,2n)$ cross section than Ag^{107}, as predicted by theoretical calculations according to Blatt and Weisskopf.[4]

In order to determine whether these trends in the (n,p), (n,α), and $(n,2n)$ cross sections also are observed at higher Z, a number of nuclei in the region of closed shell $N=82$ are being studied. The results for samarium are reported here.

EXPERIMENTAL

Cross sections for three samarium isotopes (144, 152, 154) were measured for 14.8±0.9 Mev neutrons from the $T(d,n)He^4$ reaction on the University of Arkansas Cockcroft-Walton accelerator. Natural and enriched samarium oxides were bombarded at total fluxes of approximately 10^{10} neutrons/second, and the radioactive products were identified by their known half-life or by means of cross bombardments, as discussed below.

Natural Sm_2O_3 (99+% pure) was pressed into flat tablets of known weight and area, and weighed thin copper monitor foils were placed in front and back of the tablets. This sandwich arrangement was then irradiated with 14.8-Mev neutrons for periods ranging from a few minutes to an hour. By absolute counting of 9.9-min Cu^{62} in the copper foils, the mean flux through the sample was established, since the cross section for the $Cu^{63}(n,2n)$ reaction is accurately known[3]; a mean value from the literature of 519 millibarns was used in these calculations. The Sm_2O_3 and Cu foils were counted under identical conditions of geometry in a methane-flow beta-proportional counter having a 0.9-mg/cm² aluminized Mylar end-window. Corrections for self-absorption and self-scattering were made using values extrapolated from Nervik and Stevenson[5] and Baker and Katz.[6] All samples were counted under conditions of saturation backscattering. Corrections for air absorption and scattering, scattering from the housing, counting efficiency, backscattering, and geometry were assumed to be identical for sample and monitor. The correction for average window absorption for the four beta groups of Sm^{153} (0.13, 0.645, 0.720, and 0.825 Mev[7]) was determined experimentally by running an absorption curve with Mylar films up to several window thicknesses; an average transmission of 98% for Sm^{153} radiations through the counter window was obtained. A typical decay curve from the bombardment of natural samarium is given in Fig. 1 from which the Sm^{144} and $Sm^{154}(n,2n)$ cross sections were derived. The 8.5-min activity arises from Sm^{143} and the 45-hour half-life

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[1] V. N. Levkovskii, J. Exptl. Theoret. Phys. U.S.S.R. **31**, 360 (1956) [translation: Soviet Phys. JETP **4**, 291 (1957)]; J. Exptl. Theoret. Phys. U.S.S.R. **33**, 1520 (1957) [translation: Soviet Phys. JETP **6**, 1174 (1958)].
[2] E. B. Paul and R. L. Clarke, Can. J. Phys. **31**, 267 (1953).
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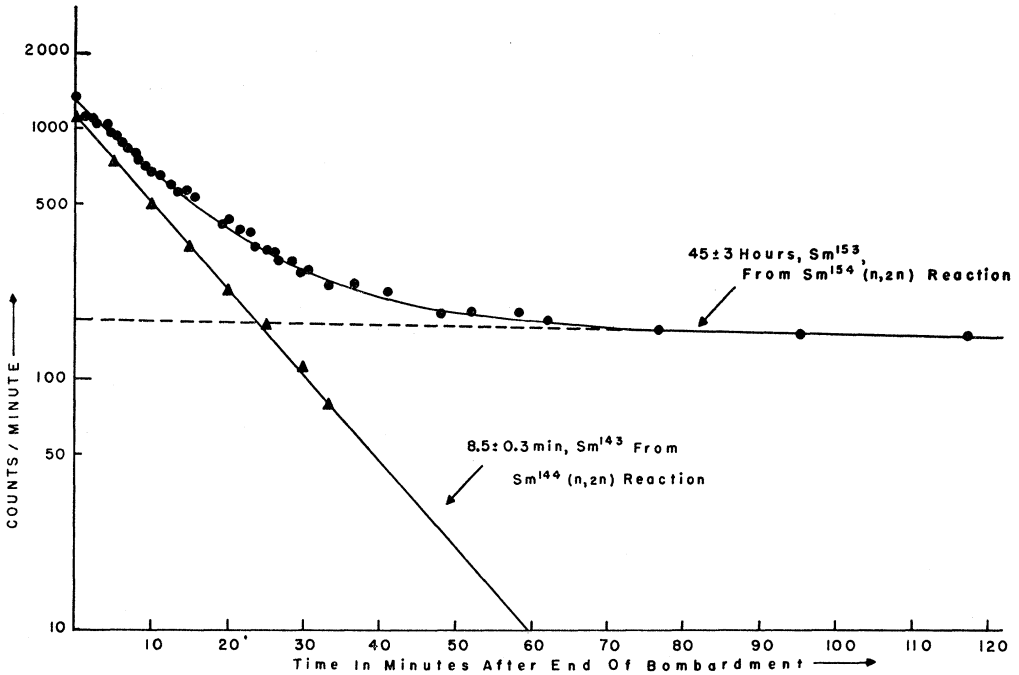

FIG. 1. Gross beta decay from a typical bombardment of natural Sm_2O_3 with 14.8-Mev neutrons for 10 minutes. The curve is resolved into two activities having half-lives of 8.5±0.3 min, assigned to Sm^{143}, and 45±3 hours, assigned to Sm^{153}. The latter activity is shown unresolved (to conserve space) from negligible amounts of intermediate half-lives induced through (n,p) and (n,α) reactions.

corresponds to Sm^{153}, both of which are well-established.[8]

When enriched Sm_2O_3 (97.2% Sm^{152}) was bombarded and counted in the same manner, three activities were resolved as shown in Fig. 2. These are 6.5±0.5 min, 1.6±0.3 hours, and 45±3 hours, the last being identified as Sm^{153} from the $Sm^{154}(n,2n)$ reaction, the sample containing 1.8% Sm^{154}. To insure the absence of any thermalized neutrons, the samples were wrapped in 0.6-mm thick cadmium metal. The 1.6-hour half-life is assigned to Nd^{149} from the $Sm^{152}(n,\alpha)$ reaction. Half-lives of about 1.7 to 2.0 hours have been reported[8] for Nd^{149} which decays with emission of 1.5-Mev beta particles. The 6.5-min activity is assigned to Pm^{152} from the $Sm^{152}(n,p)$ reaction.

Simultaneous bombardment of equal weights of natural and enriched Sm^{152} samples showed that the 6.5-min half-life is not being confused with the 8.5-min half-life of Sm^{143}, since in the enriched sample, the amount of Sm^{144} present was down by a factor of 100, so that the 6.5-min activity, if due to Sm^{143}, should have been reduced by two orders of magnitude. However, the natural samarium sample was only 3 to 4 times more active than the enriched one. The possibility that the 6.5-min activity might arise from an isomeric state in Sm^{151} was ruled out by cross-bombardment of natural Eu_2O_3, in which an (n,p) reaction should produce some 6.5-min activity if this were an isomer of Sm^{151}, but no such activity was observed. Furthermore, since the 6.5-min half-life was not observed when 99.07% enriched Sm^{154} was bombarded, it cannot be an isomer of Sm^{153}, for it would have appeared from an $(n,2n)$ reaction on Sm^{154}. The 27-hour activity[9] of Pm^{151} was not observed

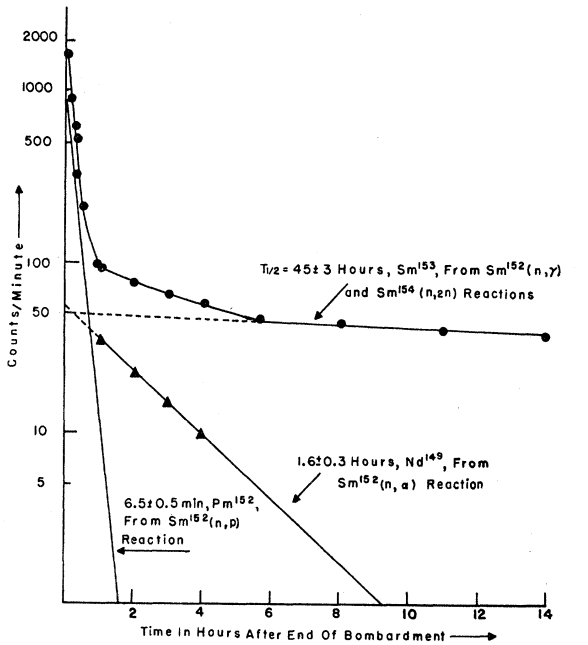

FIG. 2. Gross beta decay from a representative 10-min bombardment of enriched (97%) Sm^{152} by 14.8-Mev neutrons. The curve is resolved into half-lives of 6.5±0.5 minutes, assigned to Pm^{152}, 1.6±0.3 hours, assigned to Nd^{149}, and 45±3 hours, which is Sm^{153} from an $(n,2n)$ reaction on the small amount of Sm^{154} present and/or a small contribution from the $Sm^{152}(n,\gamma)$ reaction.

[8] Nuclear Data Cards (Natural Research Council, Washington 25, D. C.), and Strominger, Hollander, and Seaborg, Revs. Modern Phys. **30**, 585 (1958).

[9] Rutledge, Cork, and Burson, Bull. Am. Phys. Soc. Ser. II, **26**, 38 (1951).

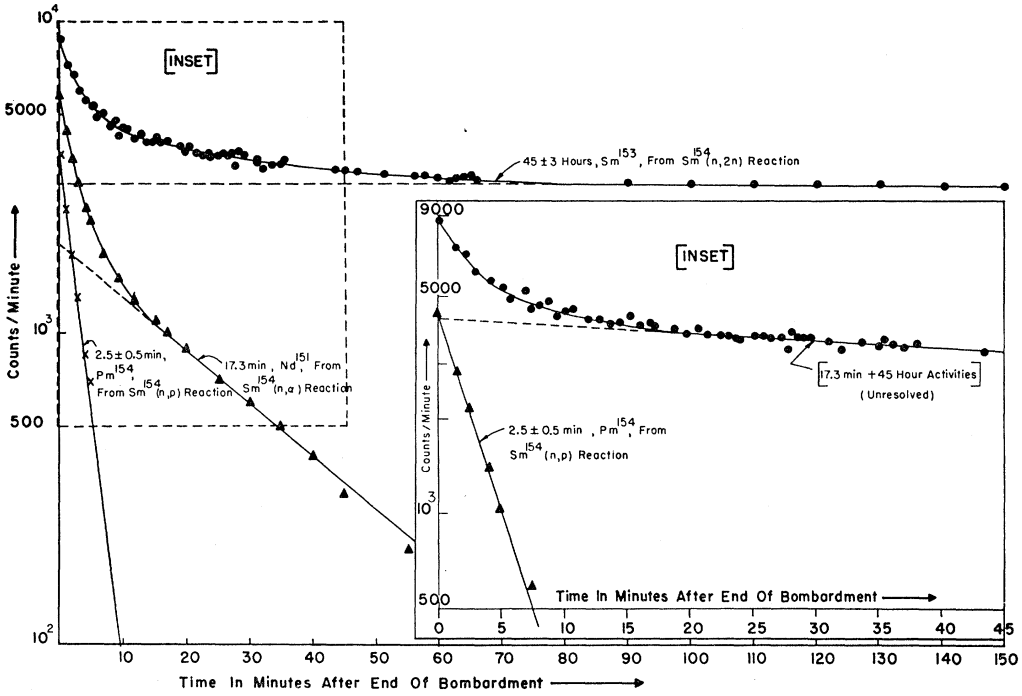

FIG. 3. Gross beta decay from a typical 10-min irradiation by 14.8-Mev neutrons of 99% enriched $(Sm^{154})_2O_3$ (cadmium-wrapped sample) showing half-lives of 2.5±0.5 minutes, assigned to Pm^{154}, 17.3±5.0 minutes, assigned to Nd^{151}, and 45±3 hours, which is Sm^{153} from the $(n,2n)$ reaction.

in any of the irradiations so that (n,np) reactions can be excluded. On the basis of these results, therefore, the 6.5-min half-life has been assigned to Pm^{152}, from the (n,p) reaction on Sm^{152}. An aluminum absorption curve gave a rough estimate of the maximum beta-particle energy for Pm^{152} of 2.2±0.5 Mev. These results and the cross-section measurements are summarized in Table I.

Bombardments of Sm_2O_3 (enriched to 99% Sm^{154}) gave rise to three resolvable activities as shown in Fig. 3. The 45-hour Sm^{153} was observed in high yield from the $(n,2n)$ reaction. The 17.3±5.0 min activity is due mostly to Nd^{151} from the (n,α) reaction with the possibility of some admixture of 23-min Sm^{155} from an (n,γ) reaction. In the calculation of the (n,α) cross section (Table I), it is assumed that all of the 17.3-min activity belongs to Nd^{151}. The 2.5±0.5 min half-life has been assigned to Pm^{154} by the same arguments given for the assignment of 6.5 minutes to Pm^{152} above. Cross bombardments with Eu_2O_3 and Sm^{152} eliminate the possibility of assignment to possible isomeric states in samarium. An aluminum absorption curve gave a maximum beta energy of 2.5±0.5 Mev for Pm^{154}. These data and the cross sections are summarized in Table I.

TABLE I. Summary of results for samarium reactions with 14.8-Mev neutrons.

Reaction	Radioactive product	Measured half-life	Observed cross section (mb) Present work	Literature values
$Sm^{144}(n,2n)$	Sm^{143}	8.5±0.3 min	1200 ±300[a]	
$Sm^{154}(n,2n)$	Sm^{153}	45 ±3 hr	1500 ±300	225[b,c]
$Sm^{152}(n,p)$	Pm^{152}	6.5±0.5 min	3.7± 0.2	
$Sm^{154}(n,p)$	Pm^{154}	2.5±0.5 min	3.5± 0.2	
$Sm^{152}(n,\alpha)$	Nd^{149}	1.6±0.3 hr	10 ± 2	8.9[b]
$Sm^{154}(n,\alpha)$	Nd^{151}	17.3±5.0 min	9 ± 3[d]	

[a] Assuming Sm^{143} positron decay is allowed and $\log_{10}(\lambda_K/\lambda_{\beta^+})=0$.
[b] See reference 2.
[c] Presumably a typographical error, since reference 2 reports a calculated value of 2100 mb and a ratio of obs/calc of 1.07; the correct experimental value is believed to be 2250 mb.
[d] Assuming all activity is due to Nd^{151}.

DISCUSSION

In Table II the observed cross sections are compared to values computed from theory as described by Blatt and Weisskopf.[4] The level densities used are based on a Fermi degenerate gas model and are given by

$$\omega(E)=C\exp(2a^{\frac{1}{2}}E^{\frac{1}{2}}), \quad (1)$$

where E is the nuclear excitation energy and C and a are parameters which vary with mass number and which are derived empirically. For the calculations in Table II, C and a were taken from the Fast Neutron Data Project Report.[10] For C, the following relationship was assumed[11]:

$$12C_{\text{even-even}}=2.4C_{\text{even-odd}}=C_{\text{odd-odd}}.$$

[10] Feld, Feshbach, Goldberger, Goldstein, and Weisskopf, U. S. Atomic Energy Commission Report NYO-636, 1951 (unpublished).
[11] G. Brown and H. Muirhead, Phil. Mag. 2, 473 (1957).

TABLE II. Comparison of observed and calculated cross sections.

Reaction	a (Mev^{-1})	θ	$\left(\frac{E_n-S_n}{\theta}\right)$	Q value (Mev)	σ_{cn} (mb)	F_p/F_n	F_α/F_n	Cross section (mb) present work Obs	Calc	$(\sigma_{obs}/\sigma_{calc})$	Cross section (mb) literature Obs	Calc
$Sm^{144}(n,2n)Sm^{143}$	9.0	1.28	6.3		2320			1200	2280	0.526		
$Sm^{154}(n,2n)Sm^{153}$	9.5	1.25	5.8		2360			1500	2330	0.644	225[a,b]	2100[a]
$Sm^{152}(n,p)Pm^{152}$	9.3			−3.2	2350	$5.64\cdot10^{-5}$		3.7	0.133	27.8		
$Sm^{154}(n,p)Pm^{154}$	9.5			−4.0	2360	$2.0\cdot10^{-5}$		3.5	0.0472	74.3		
$Sm^{152}(n,\alpha)Nd^{149}$	9.3			5.3	2350		$4.3\cdot10^{-5}$	10	0.101	99	8.9[a]	0.0064[a]
$Sm^{154}(n,\alpha)Nd^{151}$	9.5			4.8	2360		$2.31\cdot10^{-5}$	9[c]	0.0545	165		

[a] See reference 2.
[b] Probably a typographical error; the correct value is believed to be 2250 mb in reference 2.
[c] Assumed in calculations all activity due to Nd^{151}.

The theoretical expression for the (n,p) reaction cross section in the region of 14.8-Mev bombarding energy for intermediate and heavy nuclei is[4]

$$\sigma(n,p)=\sigma_{cn}(E_n)\left[\frac{F_p}{(F_p+F_\alpha+F_n)}\right], \quad (2)$$

where $\sigma_{cn}(E_n)$ is the cross section for formation of a compound nucleus by a neutron of incident energy E_n. F_n, F_p, and F_α are functions proportional to the probabilities for neutron, proton, and alpha-particle emission, respectively.[4] A similar equation for the (n,α) cross section can also be written. Q values for the (n,p) and (n,α) reactions were calculated from Wapstra[12] or from Riddell's table of Levy's empirical atomic masses.[13]

Theoretical estimations of $(n,2n)$ cross sections were obtained from the equation[4]:

$$\sigma(n,2n)=\sigma_{cn}\left[1-\left(1+\frac{E_n-S_n}{\theta}\right)\exp\left(\frac{-E_n-S_n}{\theta}\right)\right]. \quad (3)$$

Here the value of σ_{cn} was taken from graphs given by Blatt and Weisskopf,[4] and $E_n=14.8$ Mev, the incident neutron energy. S_n is the energy required to separate a neutron from the target nucleus. The nuclear temperature, θ, determines the Maxwellian energy distribution of the emitted neutrons. Values of θ were approximated from Blatt and Weisskopf.[4]

The observed results for $(n,2n)$ reactions in the present work agree rather well with the theory and are consistent with the trends mentioned earlier. The fact that $(\sigma_{obs}/\sigma_{calc})$ is less than unity might be explained, according to Blatt and Weisskopf,[4] if the first neutron leaves the compound nucleus before "thermal equilibrium" is established with the nucleus as a whole, and thus it might carry off more energy than would be expected from the Maxwellian distribution, leaving the residual nucleus less highly excited than expected with a consequent reduction of the probability for evaporation of a second neutron.

On the other hand, the deviation between observed and calculated cross sections for reactions in which charged particles are emitted is much greater than in the case of $(n,2n)$ reactions, and the ratio $(\sigma_{obs}/\sigma_{calc})$ is larger than unity by several orders of magnitude.

No reasonable adjustment of the parameters used in this theoretical calculation will bring the theoretical values into agreement with experiment. This effect was observed by Paul and Clarke[2] for heavy nuclei. These authors attributed the deviation to a direct interaction in which the incident neutron interacts strongly with one proton or with only a small part of the nucleus so that particle emission occurs before the energy can be distributed over the entire nucleus.

The importance of direct interactions, in which no intermediate nucleus is formed, in explaining the large (n,p) cross sections has been considered by Brown and Muirhead,[11] who assumed a Fermi gas model of the nucleus, and that nucleon-nucleon direct interactions can occur throughout the nuclear volume. The results from their calculations show, for example, in the case of the $Ba^{138}(n,p)$ reaction a contribution from the compound nucleus process of 0.0 mb and a direct-interaction cross section of about 3 mb (experimental value[2]: 6 mb). For $La^{139}(n,p)$, they calculate a direct interaction cross section of 9 mb (experimental value[2]: 6 mb) and for $Tl^{205}(n,p)$, 3 mb (experimental value[2]: 3 mb). Unfortunately the direct interaction model employed by Brown and Muirhead fails to account for the Levkovskii trends.[1] This failure also has been pointed out by Poularikas and Fink[14] in the case of titanium (n,p) cross sections at 14.8 Mev.

It seems probable that some kind of direct-interaction mechanism must be assumed to explain the (n,p) cross section in samarium, as shown to be true for aluminum and nickel[15,16] in the middle-Z region, but whether this is true or not for the (n,α) process at high Z (Sm) remains an open question, since in the middle-Z region the (n,α) reaction is known to proceed principally via the compound-nucleus mechanism.[17,18]

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[14] A. Poularikas and R. W. Fink (to be published).
[15] Colli, Pignanelli, Rytz, and Zurmuhle, Nuovo cimento **9**, 280 (1958).
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[17] Kumabe, Takekoshi, Ogata, Tsuneoka, and Ōki, Phys. Rev. **106**, 155 (1957).
[18] Kumabe, Takekoshi, Ogata, Tsuneoka, and Ōki, J. Phys. Soc. Japan **13**, 129, 325 (1958).

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New Germanium Isotope, Ge^{65}†

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A new germanium isotope, Ge^{65}, has been produced by the $(\alpha,3n)$ reaction on Zn^{64}. The mass assignment was established through cross bombardments, "milking" experiments, and excitation function measurements. The half-life of Ge^{65} is 1.5 ± 0.2 minutes. It decays by positron emission to 15-minute Ga^{65}. The ground-state transition occurs in over 90% of the disintegrations and has a positron end-point energy of 3.7 ± 0.4 Mev and a $\log(ft)$ value of 4.8. Gamma rays of 0.67 Mev and 1.72 Mev have been observed with intensities of $0.03/\beta^+$ and $0.02/\beta^+$, respectively.

INTRODUCTION

IN the course of a study of the reactions of Zn^{64} with alpha particles,[1] the $(\alpha,3n)$ reaction was investigated. The product of this reaction is Ge^{65}, a previously unknown nuclide. Decay systematics indicated that the half-life of this nuclide should be of the order of a minute. The threshold of the $(\alpha,3n)$ reaction on Zn^{64} was calculated to be about 33 Mev by use of measured and extrapolated decay energies and masses.[2] The 41-Mev alpha particles obtained from the Brookhaven 60-in. cyclotron could therefore be used to produce Ge^{65}.

A new 1.5-minute germanium isotope, ascribed to Ge^{65}, was found and is described in this study. The experiments performed to establish the identity of this new activity are reported in the next section. The beta and gamma radiations of Ge^{65} are discussed in the following section. In conclusion, the decay scheme of Ge^{65} is discussed.

IDENTIFICATION OF Ge^{65}

The targets used in this experiment consisted of zinc, enriched to 93% in Zn^{64},[3] plated on gold foils. The use of natural zinc made the search for a short-lived activity considerably more difficult due to the very large yield of 20-minute Ge^{67}, formed primarily by the $(\alpha,3n)$ reaction on Zn^{66}. Targets were irradiated in the external beam of the 60-in. cyclotron with 41-Mev alpha particles for periods ranging from a few seconds to two minutes. Immediately after irradiation, the target was chemically processed to separate germanium. The plated zinc was dissolved off its gold backing in concentrated HCl in the presence of Ge carrier. $KClO_3$ was added to reoxidize the germanium to the +4 state, and $GeCl_4$ was distilled from the solution. Special precautions were taken to prevent any droplets from spraying over in the course of the distillation. The distillate was transferred to a vial and counted in a sodium iodide well-counter. The activity of the sample was continuously recorded by means of a Brush recorder. The initial time of counting was approximately 2.5 to 3 minutes after the end of irradiation. At this time the contribution of the 1.5-minute activity to the total counting rate was about 40%.

The decay curves indicated the presence of 40-hour Ge^{69}, 2.5-hour Ge^{66}, 20-minute Ge^{67}, and a new 1.5-minute activity. In addition, the decay curves showed the presence of 78-hour Ga^{67} and 9.4-hour Ga^{66} formed from the decay of the corresponding germanium isotopes. Analysis of six separate experiments, including two experiments in which only annihilation radiation was counted, gives a half-life of 1.5 ± 0.2 minutes for the nuclide in question.

The possibility that the 1.5-minute activity might be due to a gallium isotope, such as 2.5-minute Ga^{64}, was ruled out in a separate experiment. A copper foil was irradiated under the same conditions, and germanium was separated by distillation. The very low counting rates showed that the decontamination from gallium was excellent, and no 1.5-minute activity was observed.

The mass assignment was established by means of a "milking" experiment in which gallium was quickly separated from a germanium sample prepared by the above procedure. A second "milking" was performed about three minutes later. Analysis of the gallium decay

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[2] Strominger, Hollander, and Seaborg, Revs. Modern Phys. 30, 585 (1958): McGinnis, Kundu, Yamada, and Way, Nuclear Data Cards (National Research Council, Washington, D. C.); A. H. Wapstra, Physica 21, 385 (1955).
[3] Obtained from Isotope Research and Production Division, Carbide and Carbon Chemicals Company, Oak Ridge, Tennessee.