various nuclear structure calculations. One might therefore expect that a good mass surface would result from a nuclear potential characterized by the parameters arrived at here.[7] In conclusion, we believe this study in conjunction with the work of Wyatt, Wills, and Green suggests that the simple nonlocal nucleon-nuclear potentials when pursued in the Frahn-Lemmer approximation can serve to describe the average behavior of nucleons with nuclei in the range of energies from minus 70 Mev to plus 25 Mev which corresponds roughly to the entire range of concern of classical nuclear physics.

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Mass Assignments and Some Decay Characteristics of Gd^{145}, Eu^{145}, Gd^{146}, and Eu^{146}†

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The nuclides Gd^{145}, Eu^{145}, Gd^{146}, and Eu^{146} were prepared by the interactions of 20- to 40-Mev helium ions with Sm^{144}, and found to decay with half-lives of 25±2 minutes, 5.6±0.3 days, 46±2 days, and 4.4±0.1 days, respectively. The mass number assignments were made on the basis of excitation functions, and chemical evidence of parent-daughter relationships, with special reference to the previously known nuclide Eu^{145}. The most prominent gamma rays appearing in the decay of each of these four nuclides are as follows: in Gd^{145} decay, at 0.80, 1.03, and 1.75-Mev; in Eu^{145} decay, at 0.53, 0.64, and 0.89 Mev; in Gd^{146} decay, at 0.114 and 0.153 Mev; and in Eu^{146} decay, at 0.63 and 0.74 Mev. There is also a strong K x-ray line in each spectrum. In addition, Gd^{145} was found to emit positrons with an end-point energy of about 2.4±0.2 Mev.

INTRODUCTION

RECENT reports by several workers[1–4] suggest the existence of a gadolinium isotope with mass number less than 147, and having a half-life in the range between 25 and 70 days. The mass number of this nuclide has been estimated variously to be 145 and 146.

This paper reports work in which both Gd^{145} and Gd^{146} were prepared and partially characterized under conditions in which the assignment of mass number was relatively unambiguous. In particular, the nuclides in question were prepared by the interaction of 20-Mev to 40-Mev helium ions with Sm^{144}, the mass assignments being made on the basis of the excitation functions for their production.

The observed half-lives were found to be 25 minutes and 46 days, respectively, for Gd^{145} and Gd^{146}, suggesting that Gd^{146} had been responsible for the observations mentioned above.

EXPERIMENTAL

Target Foils

By means of a Zapon painting technique described elsewhere,[5] reasonably uniform deposits of samarium oxide were formed on very pure (99.99%) 0.001-inch aluminum foil. The thickness of the samarium oxide layer in typical target foils was about 400 micrograms per cm^2. A simple backscattering type of beta gauge was used to assure sufficient target uniformity in those experiments in which uniformity was necessary. The samarium oxide enriched in Sm^{144} was obtained from Oak Ridge National Laboratory and had the following isotopic composition (expressed in atom percent): Sm^{144}, 58.9%; Sm^{147}, 13.5%; Sm^{148}, 5.3%; Sm^{149}, 3.2%; Sm^{150}, 1.4%; Sm^{152}, 3.8%; Sm^{154}, 14.0%. Some foils were also prepared using natural samarium oxide (Sm^{144}, 3.1%).

Irradiations

Bombardments were carried out in the 40-Mev external helium ion beam of the Brookhaven 60-inch cyclotron. The bombarding energy was adjusted by means of aluminum absorber foils, employing for this purpose the range-energy relation of Aron *et al.*[6] The full energy of helium ions incident on the target stack was taken to be 40.5 Mev, on the basis of approximate range measurements.

† Research performed under the auspices of the U. S. Atomic Energy Commission.

[1] J. R. Grover, thesis, University of California Report UCRL-3932, September, 1957 (unpublished).

[2] Shirley, Smith, and Rasmussen, Nuclear Phys. **4**, 395 (1957).

[3] Gorodinskii, Murin, Pokrovskii, Preobrazhenskii, and Titov, Doklady Akad. Nauk S.S.S.R. **112**, 405 (1957) [translation: Soviet Phys. Doklady **2**, 39 (1957)].

[4] Gorodinskii, Murin, Pokrovskii, and Preobrazhenskii, The Program and Abstracts of the Eighth Annual Conference for Nuclear Spectroscopy, Leningrad, January 27 to February 3, 1958 (unpublished), p. 22; Gorodinskii, Murin, and Pokrovskii, Izvest. Akad. Nauk S.S.S.R. Ser. Fiz. **22**, 811 (1958) [translation: Bull. Acad. Sciences U.S.S.R. **22**, 805 (1958)].

[5] Dodson, Graves, Helmholz, Hufford, Potter, and Povelites, *Miscellaneous Physical and Chemical Techniques of the Los Alamos Project*, edited by A. C. Graves and D. K. Froman (McGraw-Hill Book Company, Inc., New York, 1952), p. 1.

[6] Aron, Hoffman, and Williams, U. S. Atomic Energy Commission Report AECU-663, May, 1951 (unpublished).

Chemistry

Whenever the nuclides in question had half-lives of the order of a few hours or more, their chemical separation and identification was effected by means of a standard ion-exchange method.[7] Dowex 50(×12) resin was packed in a glass tube to form a bed 50 cm long and 1.5 cm in diameter. The resin bed was maintained at a constant temperature of 87°C, by means of a concentric outer jacket through which trichloroethylene vapor was continuously passing. The eluting agent was 1.0*M* lactic acid adjusted to a *p*H of 3.4 with ammonium hydroxide. With these conditions it was possible to separate gadolinium, europium, and samarium satisfactorily from each other, even when as much as one to two milligrams of each element were present.

Whenever the half-life of the species of interest was too short to permit its chemical identification by means of ion exchange, it was necessary to use a sodium amalgam extraction method, which is described elsewhere.[2,8] This operation permits a partial separation of europium and samarium from gadolinium, and it requires about ten minutes to perform. Since the separation factor is only about 8 to 1, at least two successive separation steps were always done.

The purified rare earths were precipitated from solution as oxalates and mounted for counting on small disks of filter paper.

Activity Measurement

The photon radiations from the prepared samples were usually detected with a 2×2-inch NaI(Tl) scintillation crystal-photomultiplier combination, and the resulting pulse-height spectra were displayed with the aid of a 100-channel analyzer.

The intrinsic photopeak efficiency curves for this crystal, which were required in order to correct the observed relative photopeak areas to relative gamma-ray intensities, were prepared by interpolation in the total efficiency tables of Wolicki *et al.*,[9] and by interpolation of the peak-to-total values of Bell.[10] The resulting approximate curves were corrected by measuring the intrinsic photopeak efficiencies at 0.662 and 1.276 Mev; the correction was smaller than 10% at worst, the experimental points being a little higher than the uncorrected curves.

Routine counting of *K* x-rays was done with a thin sodium iodide crystal and single-channel analyzer, which was set to admit the *K* x-rays of Pm, Sm, Eu, and Gd.

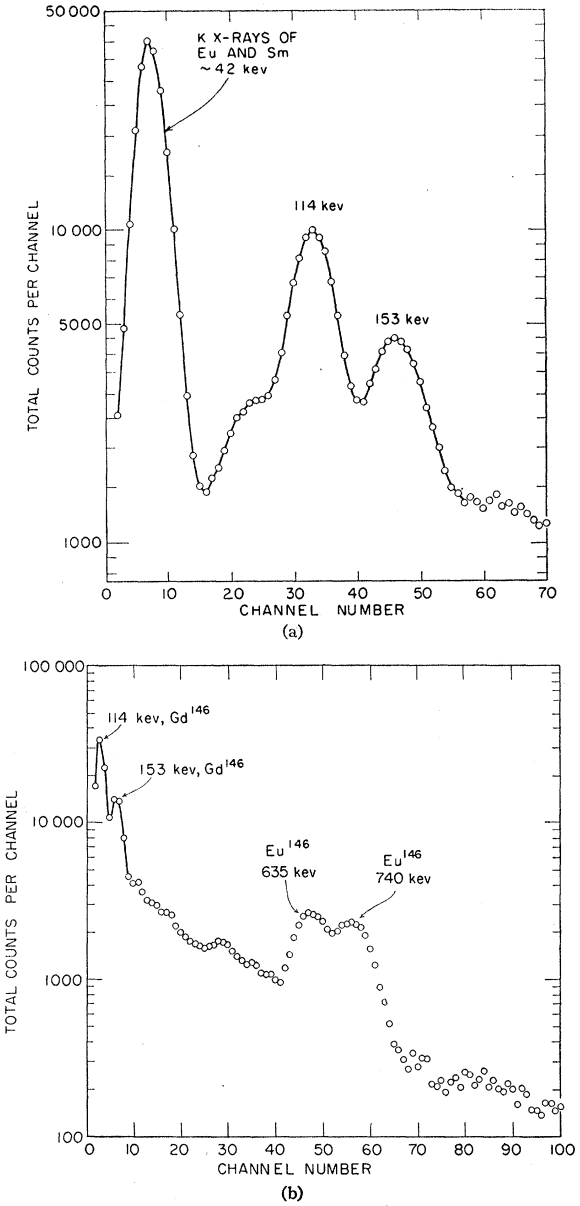

FIG. 1. (a) Gamma-ray spectrum of Gd^{146}–Eu^{146} equilibrium mixture in the energy range 20–220 kev. Taken with a 2×2-inch NaI(Tl) crystal; (b) gamma-ray spectrum of Gd^{146}–Eu^{146} equilibrium mixture in the energy range 100–1300 kev. Taken with a 2×2-inch NaI(Tl) crystal.

RESULTS

Gd^{146} and Eu^{146}

Samarium-144 was bombarded with 28-Mev helium ions, and the gadolinium activities thus produced were radiochemically purified by ion exchange.

The presence, in this preparation, of Gd^{147}, its Eu^{147} daughter, and Gd^{149}, was expected. They were easily identified by means of their known half-lives and gamma-ray spectra. In addition to these expected radiations, there appeared some unassigned gamma rays

[7] W. E. Nervik, J. Phys. Chem. **59**, 690 (1955).
[8] Rasmussen, Thompson, and Ghiorso, Phys. Rev. **89**, 33 (1953).
[9] Wolicki, Jastrow, and Brooks, Naval Research Laboratory Report NRL-4833, October, 1956 (unpublished).
[10] P. R. Bell, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North-Holland Publishing Company, Amsterdam, 1955), p. 133.

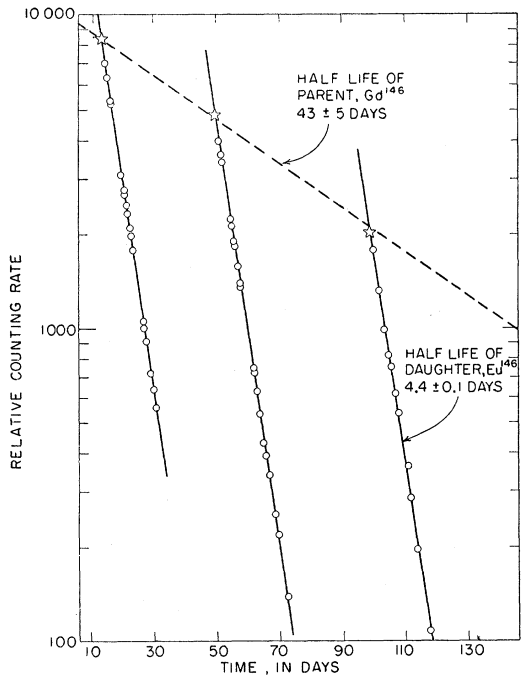

FIG. 2. Results of chemical "milking" experiment showing that the gadolinium parent of 4.4-day Eu^{146} has a half-life of 43±5 days. The points marked with stars represent the mean times of separation of the europium daughter from the reservoir of parent gadolinium.

with energies of 114±2, 153±3, 635±10, and 740±10 kev, all of which eventually decayed with half-lives of roughly 40 days. Also, the 635- and 740-kev gamma rays were clearly growing in for the first few days after purification, and if it is assumed that they result from the decay of a daughter nuclide, the half-life of this daughter would be about 5 days. Figures 1(a) and 1(b) present the gamma spectrum of an equilibrium mixture of this new species and its daughter, taken at 8% geometry and at a time when Gd^{147}, Eu^{147}, and Eu^{149} have decayed away appreciably, but before the contribution from 150-day Gd^{151} has become too objectionable. The small photopeak appearing at about 85 kev [Fig. 1(a)] seems to be a little too large to be due entirely to the escape peak of the 114-kev gamma ray.

The most trustworthy half-life of the gadolinium parent activity was obtained by following the decay of the 635- and 740-kev gamma rays in the equilibrium mixture for about four half-lives; the result was $T_{\frac{1}{2}}=46\pm2$ days.

In order to establish the chemical identity of the daughter activity, a preparation of the 46-day gadolinium was repeatedly separated from europium, by means of ion exchange, at intervals of several weeks, and observations were made of the radioactivity in the resulting europium samples. These samples all displayed prominent photopeaks at ~41 (K x-rays), 635, and 740 kev, which decayed with half-lives of 4.4±0.1 days. When appropriate corrections were made for chemical losses and radioactive decay during separation, the parent half-life could be inferred from the activities of the successive daughter samples (Fig. 2); the result was $T_{\frac{1}{2}}=43\pm5$ days, in good agreement with the 46-day half-life which was measured directly.

In an auxiliary experiment, the 635- and 740-kev gamma rays were found to be in prompt coincidence.

The mass number of this gadolinium-europium parent-daughter system was established by measuring the excitation function, by the method of stacked foils, for the production of 46-day gadolinium in the interactions of helium ions with samarium-144. The results are shown in Fig. 3. The threshold and maximum are in the neighborhood of 21 Mev and 34 Mev, respectively, strongly suggesting that the parent nuclide is formed by an $(\alpha,2n)$ reaction. It was established that this activity does indeed arise from the interaction of helium ions with Sm^{144}, rather than with some other isotope of Sm, by bombarding a target stack in which the foils were alternately natural samarium and enriched samarium-144. The 46-day species was unambiguously detectable only in the samarium-144 foils, where it was the predominating activity for bombarding energies well above its threshold. These experiments indicate that the mass number of the 46-day gadolinium isotope (and therefore of its 4.4-day europium daughter) is 146, because it is apparently formed by the reaction $Sm^{144}(\alpha,2n)Gd^{146}$.

The two most prominent gamma rays in the decay of Gd^{146}, appearing at 114 and 153 kev (with relative intensities of 1.0 and 0.4, respectively), may well be identical with the most prominent gamma rays of 114.7, 115.5, and 153.6 kev reported by Shirley *et al.*[2] to be present in an unidentified gadolinium activity which they did not attempt to assign, except to state that the half-life is ≫9 days. It is quite probable that they were actually working with 46-day Gd^{146}.

Gorodinskii *et al.*[4] have reported a gadolinium isotope

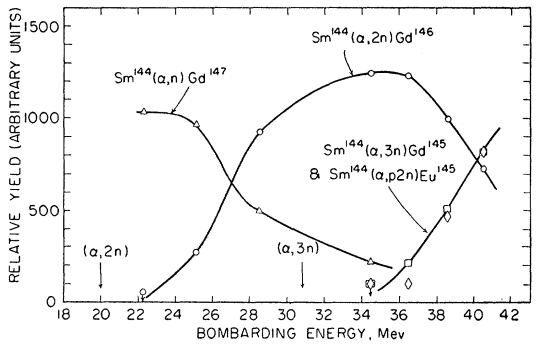

FIG. 3. Excitation functions for the formation of various products in the bombardment of Sm^{144} with helium ions. △, 29-hour Gd^{147}; ○, 46-day Gd^{146}; ◇, 25-minute Gd^{145}; □, Gd^{145} +Eu^{145} observed by means of 5.6-day Eu^{145} a few days after the end of bombardment. The expected reaction thresholds predicted by Cameron's mass table [A. G. W. Cameron, Chalk River Project Report CRP-690, March 1957 (unpublished)] are also indicated.

with a half-life of 52±10 days, appearing among the products of the reactions of 660-Mev protons with tantalum. Moreover, they separated a (4.3±0.6)-day europium daughter from the gadolinium activity, and found the same gamma spectrum for each of these species as is reported in this paper. They found that the 115-kev gamma ray in the decay of their 50-day gadolinium was in reality two gamma rays of 115 kev each, in coincidence with each other. They concluded that the mass number of this decay chain is most likely 146, primarily on the basis of a comparison of their observed gamma spectra with the systematics of the known excited levels of neighboring nuclides (particularly the even-even isotopes of neodymium).

The 38-hour species, which was observed by Hoff *et al.*[11] and tentatively assigned to Eu^{146} on the basis of cross bombardments, was not observed in this work. Also, it was not observed among the products of the reactions of tantalum with 5.7-Bev protons,[1] where one might expect it to be formed in good yield. However, Gusev *et al.*[12] report observing a 1.6-day neutron deficient europium activity among the spallation products resulting from the interactions of 660-Mev protons with tantalum. They also report that the half-life of the parent of this europium isotope is about 12 hours. These results are in apparent contradiction with the results at 5.7 Bev.

The relative energies of the two prominent gamma rays in the decay of Eu^{146}, and the fact that they are in prompt coincidence, suggests that Sm^{146} may be an example of a "near-harmonic" nucleus.[13] Indeed, this particular nuclear excitation pattern is expected of Sm^{146}, because it is an even-even nucleus having a neutron number in the region $36 \leqslant N \leqslant 88$ (it has 84 neutrons), which is exactly the situation in which the near-harmonic nuclei are known to occur. This consideration guides one to assume that the first excited state of Sm^{146} is at 635 kev instead of 740 kev, and that the second excited state is therefore at 1375-kev excitation (Fig. 4), in order that the ratio (energy of second excited state)/(energy of first excited state)=2.17, to agree with a value of ~2.2 which is observed for the other near-harmonic nuclei which have about 600- to 700-kev excitation for their first excited states (e.g., Cd^{110}).[13] In addition, no 1375-kev gamma ray was observed in the scintillation spectra (aside from a small peak explainable as an addition peak) to correspond to the forbidden two-phonon crossover transition; a conservative upper limit for the crossover branch of <5% can be assigned.

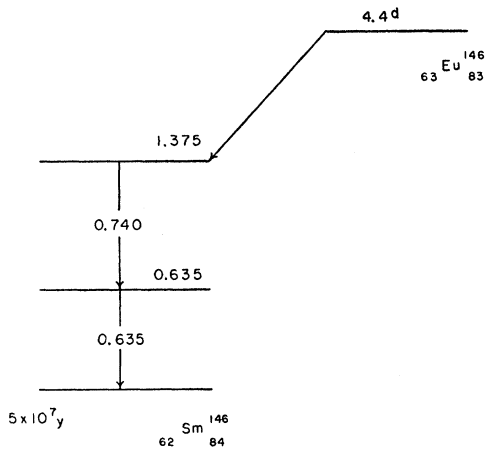

FIG. 4. Proposed decay scheme for Eu^{146}.

Gd^{145}

Samarium-144 was bombarded with 40.5-Mev helium ions, and europium and samarium were quickly separated from the resulting mixture of reaction products by extraction into sodium amalgam. The remaining rare earth products were carried from solution with a precipitate of gadolinium oxalate, and their radiations were examined.

The decay of the K x-ray peak revealed a very prominent component with a half-life of about 25 minutes. Gamma rays with about this same half-life were also observed at 510, 800±10, 1035±15, and 1750±20 kev. The 510-kev photopeak was considerably greater in intensity (~65%) when the sample was sandwiched between absorbers than it was when the upper absorber was omitted, and hence must be largely due to annihilation radiation.

Figures 5(a) and 5(b) present the gamma-ray spectrum observed for this nuclide (using a 3×3-inch NaI(Tl) crystal). The contributions from the gamma spectra of Gd^{146} and Gd^{147} interfere with attempts to observe gamma rays of small intensity in the region below ~300 kev. A channel-by-channel subtraction of two spectra taken about an hour apart yielded a spectrum in which these gamma rays were effectively removed [Fig. 5(b)], which helped considerably in ascertaining that there were no prominent 25-minute gamma rays in this region.

The best-determined half-life is a weighted mean value found by following the decay of K x-rays, positrons, and the 1.75-Mev gamma ray, the result being $T_{\frac{1}{2}}=25\pm2$ minutes.

The combined europium-samarium fraction of the reaction products was extracted from the amalgam, recovered in the form of the precipitated oxalates, and examined with the gamma-ray spectrometer. Apart from a relatively small component of about 40 minutes half-life (probably some 25-minute impurity, at least in part), the main part of the radioactivity decayed with a half-life of about five days. The gamma-ray spectrum revealed that there was much Eu^{146} present,

[11] Hoff, Rasmussen, and Thompson, Phys. Rev. **83**, 1068 (1951).
[12] Gusev, Lilova, Murin, Preobrazhenskii, and Iakovlev, J. Exptl. Theoret. Phys. U.S.S.R. **32**, 1585 (1957) [translation: Soviet Phys. JETP **5**, 1295 (1957)].
[13] G. Scharff-Goldhaber and J. Weneser, Phys. Rev. **98**, 212 (1955); G. Scharff-Goldhaber, Phys. Rev. **103**, 837 (1956).

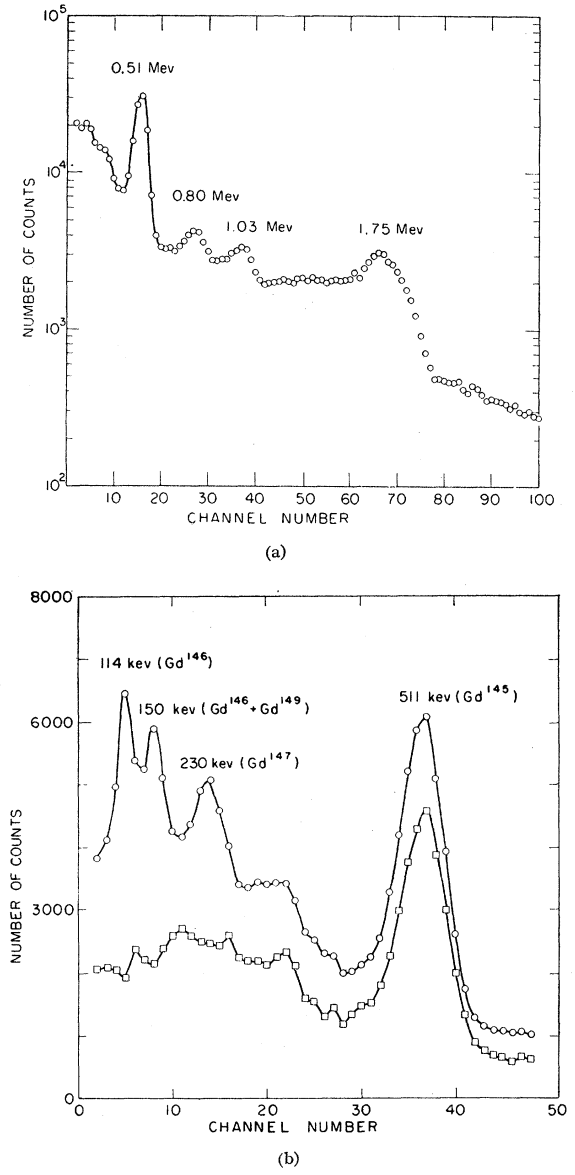

FIG. 5. (a) Gamma spectrum of 25-minute Gd^{145}; (b) Upper curve: gamma spectrum of the gadolinium isotopes produced by the 40-Mev helium ion bombardment of enriched Sm^{144}, taken two hours after the end of bombardment. Lower curve: same spectrum, but with the spectrum taken one hour later subtracted; the result is an essentially pure spectrum for 25-minute Gd^{145}.

as expected, and that in addition there were unassigned gamma rays at ~530, 645±10, and 890±10 kev, all of which also decayed with half-lives of about five days.

Since Eu^{145} has been identified[11] to be a five-day activity arising as the alpha decay daughter of Tb^{149}, the unassigned gamma rays probably belong to Eu^{145}. In order to check this supposition, Sm^{144} was bombarded with 12-Mev deuterons, and the resulting europium reaction products were carefully purified by ion exchange. Gamma rays at 530, 645, and 890 kev, decaying with a half-life of 5.6±0.3 days, were again observed, and were initially the most prominent radiations in the sample. The gamma rays characteristic of Eu^{146} were definitely absent.

In order to establish whether the 25-minute activity was the parent of 5.6-day europium, a fresh preparation of the 25-minute species was separated from europium, using sodium amalgam extraction, and set aside to form a reservoir. At regular timed intervals, aliquots were taken from this reservoir, and from these the europium was rapidly separated using sodium amalgam. These europium samples proved to be reasonably pure preparations of the 5.6-day europium species, displaying the same gamma spectrum (Fig. 6) as that observed for the 5.6-day europium freshly prepared in the deuteron bombardment. After suitable corrections were applied for chemical yield, etc., the half-life of the parent of the 5.6-day europium preparations was inferred to be 20±8 minutes, in reasonable agreement with the 25-minute half-life which was observed directly for the unknown activity.

An excitation function was measured for this 25-minute activity by means of the method of stacked foils, with the results shown in Fig. 3. After the 25-minute material had decayed completely, another excitation function was measured in the same stack of foils, for the formation of the 5.6-day europium daughter (making use of the prominent and easily observed 890-kev gamma ray). When a suitable scale factor was applied, the two excitation functions were identical, within experimental uncertainties. The threshold appears to be in the neighborhood of 33 Mev, consistent with the assumption that the decay chain in question originates with the $(\alpha,3n)$ reaction. Assurance that these species are formed by reaction with Sm^{144} was provided by bombarding a target stack in which the

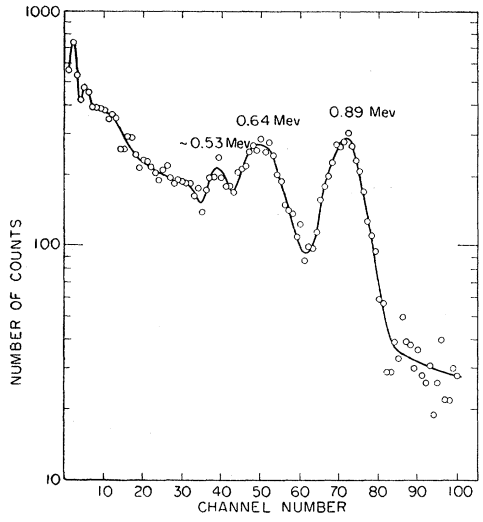

FIG. 6. Gamma spectrum of 5.6-day Eu^{145}. This specimen was prepared by chemically "milking" the 25-minute Gd^{145}. The two small peaks at ~120 and 150 kev are due to a small amount of Gd^{146} impurity.

TABLE I. Summary of observations.

Nuclide	Half-life	Photon radiations (Mev)	Relative intensity	Remarks
Gd^{145}	25 ±2 min	K x-rays	1.0	This nuclide emits positrons with an end-point energy of 2.4±0.2 Mev. The 1.75-Mev gamma ray may be an unresolved doublet.
		0.510	{1.1 if pure γ 0.6 if pure β^+	
		0.800±0.010	0.13	
		1.035±0.015	0.14	
		1.750±0.020	1.4	
Eu^{145}	5.6±0.3 days	K x-rays	1.0	
		~0.53	~0.03	
		0.645±0.010	0.24	
		0.890±0.010	0.6	
		~1.30?	weak	
		~1.65?	weak	
Gd^{146}	46 ±2 days	K x-rays	<2.4	These intensities are normalized to Eu^{146} (given below) via measurements on the equilibrium mixture.
		0.114±0.002	1.0	
		0.153±0.003	0.4	
Eu^{146}	4.4±0.1 days	K x-rays	1.0	The 0.635- and 0.740-Mev gamma rays are in coincidence, and are of nearly equal intensity.
		0.635±0.010 0.740±0.010	1.8 (sum)	
		(1.38)	<0.04	

target foils were alternately enriched samarium-144 and natural samarium. The 5.6-day europium activity, which was identified by means of its very prominent 890-kev gamma ray, was detected definitely and unambiguously only in the foils prepared from enriched samarium-144. The mass number of the 25-minute activity is thus most probably 145. Also it is quite likely that it is a parent of the previously identified 5-day Eu^{145}, and is therefore Gd^{145}, its formation being consistent with the reaction $Sm^{144}(\alpha,3n)Gd^{145}$.

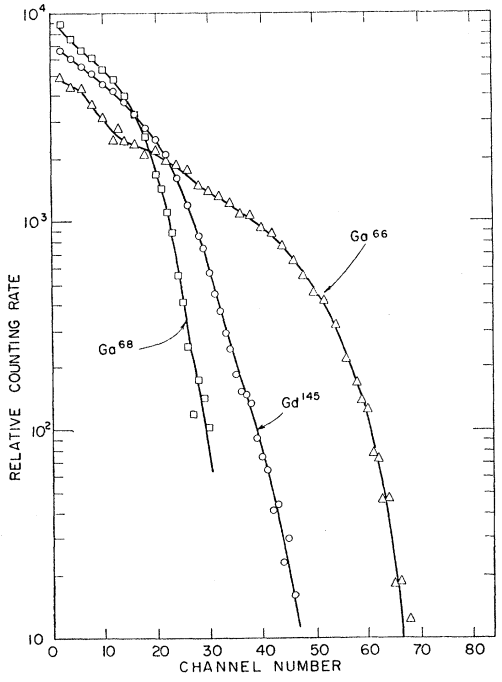

FIG. 7. Positron energy spectrum of 25-minute Gd^{145} observed with a plastic scintillator. Also included are the spectra of Ga^{66} and Ga^{68} which were used for calibration. All spectra are normalized to equal total number of counts.

An approximate value for the end-point energy of the positron spectrum was measured using a 2×2-inch plastic scintillator with a 100-channel pulse-height analyzer. The pulse-height spectrum displayed by the decay of a freshly prepared Gd^{145} sample was observed, first directly, and then with sufficient beryllium absorber interposed to stop all of the positrons. A spectrum which was obtained by subtracting the second spectrum from the first (after correcting for decay) was taken to represent the approximate positron spectrum of Gd^{145}. This spectrum was compared to the positron spectra of Ga^{68} and Ga^{66}, which have positron end-point energies at 1.89 Mev and 4.15 Mev, respectively. All three spectra were obtained in the same equipment at approximately the same time, with the results shown in Fig. 7. The positron end-point energy of Gd^{145}, which was estimated by various interpolations between the known spectra, is 2.4±0.2 Mev. There appear to be no other obvious components in the spectrum, within the rather broad limits imposed by this admittedly crude experiment.

The intensity of the 1.75-Mev gamma ray is so great (see Table I) that the bulk of the decay of Gd^{145} must populate states which include this energetic transition in their de-excitation. Moreover, the energy and logft value (~5.5) of the positron group are such that nearly all of the observed K x-rays must arise from electron capture transitions to the same level or levels as are populated by the positrons, in order to satisfy the theoretical K-capture to positron ratio for allowed transitions.[14]

Even if one assumes that the relative positron abun-

[14] M. L. Perlman and M. Wolfsberg, Brookhaven National Laboratory Report BNL-485 (unpublished).

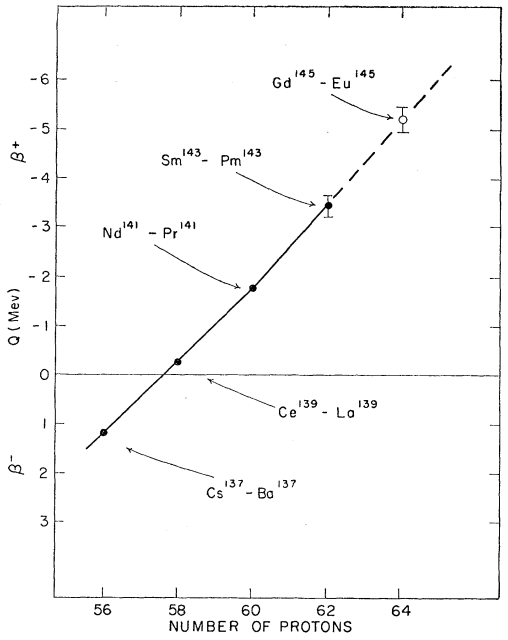

FIG. 8. Plot of beta-decay energies between the odd mass isotones which have 81 and 82 neutrons. The form of the plot follows that of Way and Wood,[15] and the data for the solid points are taken from Strominger, Hollander, and Seaborg [Revs. Modern Phys. **30**, 585 (1958)]. The decay energy observed for Gd^{145}, plotted as an open circle, is consistent with reasonable extrapolations of the solid points.

dance is overestimated by a factor of two (an unreasonably large error) due to a contribution to the annihilation photopeak by a nuclear gamma ray of about 500 kev, one still must conclude that the principal positron group populates some state or states decaying by means of the 1.75-Mev gamma ray, because it is not possible to arrange the decay scheme in any other way which will also permit such a large intensity of 1.75-Mev gamma rays.

If this conclusion is correct, then the total decay energy of Gd^{145} is 5.2 ± 0.3 Mev or greater. However, since there are no other gamma rays present with an intensity comparable to that of the 1.75-Mev gamma ray, the decay energy is unlikely to be higher than the calculated value.

We may check these conclusions by employing the beta-decay systematics of Way and Wood[15] to estimate the expected total decay energy of Gd^{145}. The result is presented in Fig. 8, in which are plotted all of the known beta-decay energies occurring between isotones of 81 and 82 neutrons among odd-mass nuclides. The open circle corresponding to the decay of Gd^{145} appears to be reasonably placed with respect to the other points.

Table I summarizes the observations which were made concerning the decay of all of the nuclear species described in this paper. The errors in the estimated relative gamma-ray intensities probably do not exceed 25% of the quoted values for the more intense photopeaks, and are correspondingly higher for gamma rays of lesser intensity.

ACKNOWLEDGMENTS

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[15] K. Way and M. Wood, Phys. Rev. **94**, 119 (1954).