

Decay of  $\text{Pm}^{149}$  (53 hr)\*

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The radioactive sources were obtained from neodymium oxide, enriched in  $\text{Nd}^{148}$ , irradiated in the Argonne reactor CP-5. After the 1.8-hr activity of  $\text{Nd}^{149}$  had decayed away, the 53-hr activity in  $\text{Pm}^{149}$  remained. Some of the samples were purified by ion-exchange-column techniques before being studied. The radiations were investigated with a  $180^\circ$  focusing beta-ray spectrometer and the Argonne 256-channel scintillation coincidence spectrometer. The magnetic spectrometer resolved two components of the beta spectrum at  $1.064 \pm 0.008$  and  $0.784 \pm 0.010$  Mev. The beta rays were also studied by coincidence absorption techniques. In addition to the branch with a maximum energy of  $0.784 \pm 0.010$  Mev, this method revealed two more at  $0.47 \pm 0.04$  and  $0.19 \pm 0.04$  Mev. The scintillation pulse-height spectrum of the gamma rays revealed the presence of three transitions at  $0.850 \pm 0.008$ ,  $0.582 \pm 0.006$ , and  $0.285 \pm 0.001$  Mev. A fourth gamma ray with an energy of  $0.548 \pm 0.006$  Mev was found in coincidence measurements. These radiations are fitted into a decay scheme comprising the ground state and four excited states in  $\text{Sm}^{149}$  at 0.285, 0.582, 0.833, and 0.850 Mev. The  $\log ft$  values, transition intensities, and possible spin and parity assignments are discussed.

## INTRODUCTION

AN activity with a half-life of 47 hr was first produced<sup>1</sup> by bombarding stable neodymium with deuterons and fast neutrons. Later, it was reported<sup>2-6</sup> that this activity was characterized by a 0.2-Mev gamma ray and beta particles with a maximum energy of 1.05 Mev. The active nuclide was identified<sup>7</sup> as the isotope of promethium with mass 149. Rutledge *et al.*<sup>8</sup> observed *K* and *L* internal-conversion-electron lines for the gamma transition by means of magnetic internal-conversion electron spectrographs. From the positions of these lines and the ratio of their intensities, it was concluded that the transition has an energy of 0.2849 Mev and *M1* (or possibly *M2*) character. Recently, the half-life of  $\text{Pm}^{149}$  has been reported as  $53.09 \pm 0.09$  hr.<sup>9</sup>

There are no stable isotopes of promethium. However,  $\text{Pm}^{149}$  is produced in the beta decay of  $\text{Nd}^{149}$ . Sources of the latter isotope were produced by irradiating neodymium oxide (98.8%  $\text{Nd}^{148}$ ) with thermal neutrons. The half-life of  $\text{Nd}^{149}$  is 1.8 hr and that of  $\text{Pm}^{149}$  is 53 hr; so by allowing the sources to decay for two days before investigating the radiations, it was possible to obtain  $\text{Pm}^{149}$  with unobservable amounts of  $\text{Nd}^{149}$ .

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<sup>1</sup> H. B. Law, M. L. Pool, J. D. Kurbatov, and L. L. Quill, *Phys. Rev.* **59**, 936 (1941).

<sup>2</sup> W. Bothe, *Z. Naturforsch.* **1**, 179 (1946).

<sup>3</sup> J. A. Marinsky, L. E. Glendenin, and C. D. Coryell, *J. Am. Chem. Soc.* **69**, 2, 2781 (1947).

<sup>4</sup> B. Ketelle, Oak Ridge National Laboratory Report ORNL-299, (unpublished), p. 34.

<sup>5</sup> C. Mandeville and M. Scherb, *Phys. Rev.* **76**, 186(A) (1949).

<sup>6</sup> E. Kondaiah, *Phys. Rev.* **81**, 1056 (1951).

<sup>7</sup> J. A. Marinsky and L. E. Glendenin, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), Paper No. 193, National Nuclear Energy Series, Plutonium Project Record, Div. 4, Vol. 9.

<sup>8</sup> W. C. Rutledge, J. M. Cork, and S. B. Burson, *Phys. Rev.* **86**, 775 (1952).

<sup>9</sup> L. R. Bunney, J. O. Abriam, and E. M. Scadden, U. S. Naval Research and Development Laboratory Report USNRDL-TR-305, 1959 (unpublished).

The radiations were studied with the Argonne 256-channel scintillation coincidence spectrometer and with a  $180^\circ$  beta-ray spectrometer. When "singles" spectra of the gamma rays were being measured with the scintillation spectrometer, a lead collimator was placed between the source and the NaI(Tl) crystal (a  $2\frac{1}{4}$ -in. cube). The collimator (insert Fig. 2) is lined with successive layers of tantalum, tin, and stainless steel in order to suppress x rays that result from fluorescence of the lead. A beryllium filter (1284 mg/cm<sup>2</sup>) was placed between the source and the collimator to absorb beta particles, beryllium being used to minimize the bremsstrahlung. For the coincidence experiments, a "fast-slow" coincidence arrangement was used. The resolving time of this circuit was  $2\tau = 5 \times 10^{-8}$  sec.

## BETA-RAY SPECTRUM

The momentum distribution of the beta particles was obtained with a variable-field  $180^\circ$  beta-ray spectrometer. The Fermi plot calculated from these data is shown in Fig. 1. The spectrum is analyzed into two components. These have end-point energies of  $1.064 \pm 0.008$  Mev and  $0.784 \pm 0.010$  Mev and relative intensities of  $91 \pm 3\%$  and  $9 \pm 3\%$ , respectively. No conclusion could be drawn about the shape of the inner spectrum (curve *b*) because of the scatter of the points. The *K* and *L* conversion lines of the 0.285-Mev gamma ray were also observed. From these data, the ratio of the intensity of the *K* electrons to that of the *L* electrons is found to be  $9.0 \pm 1.5$ , in agreement with the value of  $8.0 \pm 2.5$  obtained by Rutledge *et al.*<sup>8</sup>

## GAMMA-RAY SPECTRA

Preliminary consideration of the gamma-ray scintillation spectrum showed that impurities were present in the source. No detailed discussion of these early spectra are included since chemical purifications were subsequently carried out. However, a peak correspond-

ing to a 0.850-Mev gamma ray was observed in the unpurified sources. The decay of this gamma ray was followed for 15 days, and its half-life was found to be the same as that of the 0.285-Mev gamma ray. After appropriate corrections were made to the data, a value of 0.07 was found for the ratio of the intensity of the 0.850-Mev gamma ray to that of 0.285-Mev gamma ray. Other investigators<sup>8,10</sup> reported a gamma ray with an energy greater than 1 Mev, but none attributable to this activity was found with an intensity greater than 0.1% of the intensity of the 0.285-Mev gamma ray.

Radiation from the decay of 11.2-day  $\text{Nd}^{147}$  was emitted by the sources. This made analysis of the scintillation data difficult in the energy region between 285 and 850 kev. (The  $\text{Nd}^{147}$  resulted from neutron irradiation of  $\text{Nd}^{146}$  present in the source material because of incomplete isotopic separation.) For this reason the  $\text{Pm}^{149}$  and  $\text{Nd}^{147}$  were separated by ion-exchange methods<sup>11</sup> using Dowex-50 resin (see Appendix).

The scintillation spectrum from the promethium fraction (which contains some 27-hr  $\text{Pm}^{151}$ ), is shown in Fig. 2. In addition to the photopeak for the 0.285-

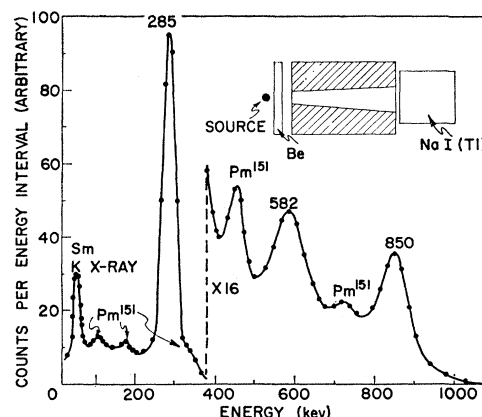


FIG. 2. Scintillation spectrum of the radiation from the Pm fraction of the separated samples.

Mev gamma ray, the one for the 0.850-Mev gamma ray is also seen. A calculation of its intensity relative to that of the 0.285-Mev gamma ray yields the same value (0.07) as for the unseparated source. This information corroborates the assignment of the 0.850-Mev gamma ray to the  $\text{Pm}^{149}$  activity.

A peak which corresponds to a 0.582-Mev gamma ray is also observed. (In the pulse-height distribution for the unseparated sources, this peak was masked by pulses corresponding to 0.532-Mev radiation from the  $\text{Nd}^{147}$ .) The decay of the 0.582-Mev radiation, which was followed for four days, exhibited a half-life of  $(50 \pm 4)$  hr. This result is to be compared with the assumed half-life of 53 hr for the 0.285-Mev gamma ray. (The decay was not followed for more than four days since, at the end of this period, the source was too weak to allow additional data to be significant.) Therefore the 0.582-Mev gamma ray has the same half-life as the 0.285- and 0.850-Mev radiation and is probably also emitted in the decay of  $\text{Pm}^{149}$ .

A value of  $\alpha_K$  for the 0.285-Mev transition was obtained from a scintillation spectrum. This spectrum was from a source which had "aged" for two weeks so that the  $\text{Pm}^{151}$  contamination had decayed relative to the  $\text{Pm}^{149}$ . The spectrum comprises four peaks: the photopeaks of three of the  $\text{Pm}^{149}$  gamma rays and the 40-kev samarium K x rays emitted following the internal-conversion process. (Because of the difference between the half-life of  $\text{Pm}^{151}$  and that of  $\text{Pm}^{149}$ , no peaks corresponding to the characteristic radiation of  $\text{Pm}^{151}$  were observed in this scintillation spectrum.) The value  $\alpha_K = 0.16 \pm 0.05$  was obtained by calculating the ratio of the intensity of the K x rays to the intensity of the 0.285-Mev gamma ray; appropriate corrections for fluorescence yield and counter efficiency were made. This calculation assumes that the x rays result almost entirely from conversion of the 0.285-Mev gamma ray. This assumption is justifiable since the intensities of the high-energy gamma rays are much less than that of the 0.285-Mev gamma ray. In addi-

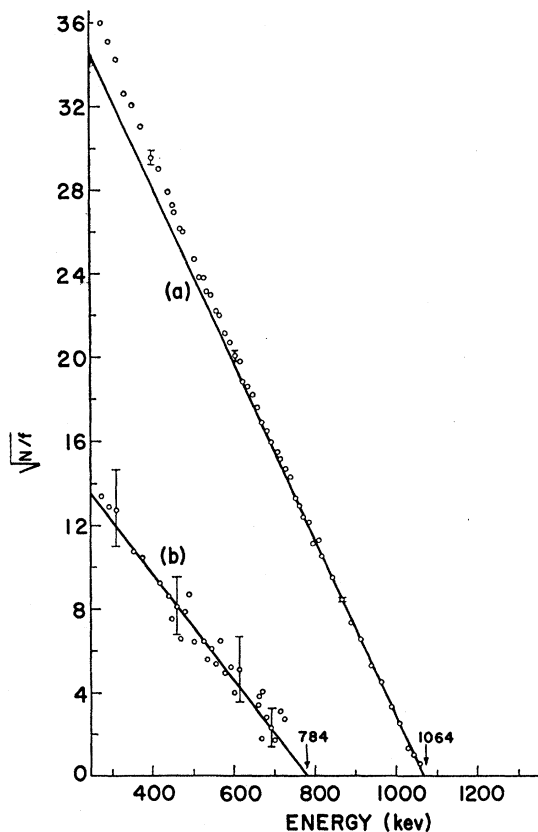


FIG. 1. Fermi plot of the beta-ray spectrum of  $\text{Pm}^{149}$ .

<sup>10</sup> V. K. Fischer, Phys. Rev. **96**, 1549 (1954).

<sup>11</sup> The procedure for this particular separation was furnished through courtesy of K. F. Flynn and J. G. Cuninghame of the Chemistry Division of Argonne National Laboratory.

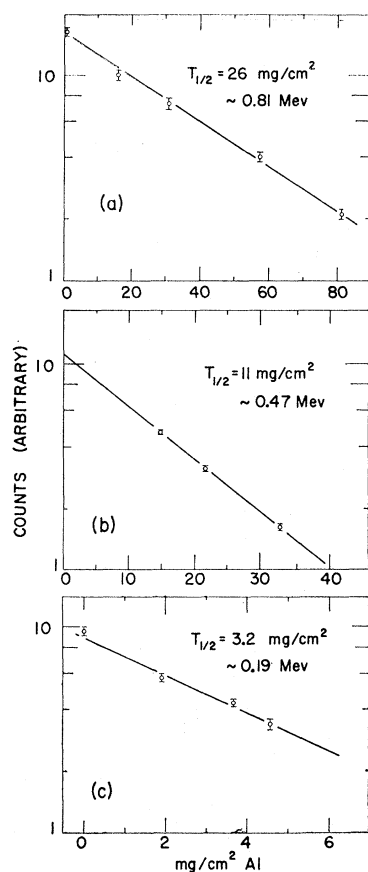


FIG. 3. Absorption of the  $\text{Pm}^{149}$  beta rays in aluminum. (a) Those in coincidence with the 0.285-Mev gamma ray, (b) those in coincidence with the 0.582-Mev gamma ray, and (c) those in coincidence with the 0.850-Mev gamma ray.  $T_{1/2}$  is the "half-thickness" value.

tion, because of their higher energies, the conversion of the former transitions would be less than that of the 0.285-Mev gamma ray. The experimental results for the 0.285-Mev transition are tabulated in Table I. Theoretical values<sup>12</sup> for  $\alpha_K$  and the  $K/L$  ratio for several different multiplicities are also listed for comparison. On the basis of its internal-conversion coefficient, the 0.285-Mev transition is considered to be magnetic dipole radiation; the  $K/L$  ratio is not inconsistent with this interpretation.

#### BETA-GAMMA COINCIDENCE MEASUREMENTS

Absorption in aluminum shows that the beta-ray group in coincidence with the 0.285-Mev gamma ray has a maximum energy of about 0.81 Mev (Fig. 3).

TABLE I. The 0.285-Mev gamma-ray transition.

	Experi- mental value	$E1$	Theoretical value $E2$	$M1$	$M2$	Multiplicity
$\alpha_K$	0.16	0.017	0.043	0.10	0.45	$M1$
$K/L$	9.0	7.6	2.6	7.9	5.4	$M1$ or $E1$

<sup>12</sup> M. E. Rose, *The Internal Conversion Coefficients* (Interscience Publishers, New York, 1958).

This component is to be identified with the 0.784-Mev group revolved by the  $180^\circ$  beta-ray spectrometer.

Assignment of the 0.850-Mev gamma ray to the decay of  $\text{Pm}^{149}$  was further substantiated by  $\beta$ - $\gamma$  coincidence measurements. This gamma ray is in coincidence with a beta-ray component which has a maximum energy of 0.19 Mev (Fig. 3). Within the limits of experimental error, this is the same as the value (0.21 Mev) expected from the total disintegration energy of 1.06 Mev.

The maximum energy of the beta transition in coincidence with the 0.582-Mev gamma ray was measured by absorption of the beta particles in aluminum. From these measurements, the maximum energy is found to be 0.47 Mev. Therefore it is concluded that the 0.582-Mev gamma ray is a transition to the ground state since the sum of its energy and that of the coincident beta ray is 1.05 Mev, which is in good agreement with the total decay energy of 1.06 Mev for the  $\text{Pm}^{149} \rightarrow \text{Sm}^{149}$  decay. The intensity of the 0.47-Mev beta-ray component was deduced from a measurement of the ratio of the intensity of the 0.582-Mev gamma ray to that of the 0.285-Mev gamma ray. This ratio, as obtained with the scintillation spectrometer, is 0.04. To correspond to this ratio, the intensity of the 0.47-Mev branch must be 0.3% of the total intensity of the beta rays. The energies and intensities of the beta-ray components are summarized in Table II. Also listed are their calculated  $\log ft$  values and corresponding selection rules.

#### GAMMA-GAMMA COINCIDENCE MEASUREMENTS

The  $\gamma$ - $\gamma$  coincidence experiments revealed a gamma ray which was not observed in other experiments. This radiation has an energy of 0.548 Mev and is in coincidence with the 0.285-Mev gamma ray. The intensity of this transition relative to that of the 0.285-Mev one must be less than 2% since the transition is not evidenced in the "singles" spectrum. No other gamma-gamma coincidences were observed.

#### DECAY SCHEME

The conclusions from the experimental evidence are summarized in the decay scheme, Fig. 4. The fact that there exists a beta branch between the ground states of  $\text{Pm}^{149}$  and  $\text{Sm}^{149}$  is of theoretical importance. Such a transition is expected according to the shell-model theory of nuclei. However, before this investigation, only the 1.06-Mev beta component had been observed,

TABLE II. Beta-ray transitions in the decay of  $\text{Pm}^{149}$ .

Energy (Mev)	Intensity	$\log ft$	Spin change	Parity change
1.064	89%	7.2	0, 1	yes
0.784	10%	7.5	0, 1	yes
0.47	0.3%	8.2	2	yes
0.21	0.6%	6.8	0, 1	yes

and it was believed to proceed to the 285-keV level rather than to the ground state of  $\text{Sm}^{149}$ . If the ground state of  $\text{Pm}^{149}$  is assumed to be a  $d_{3/2}$  level as expected from the systematics of shell theory, then the selection rules for the beta transitions (Table II) require that the levels in  $\text{Sm}^{149}$  have negative parities and spins of  $\frac{3}{2}$ ,  $\frac{5}{2}$ , or  $\frac{7}{2}$ —except for the 580-keV level which must have a spin of  $\frac{1}{2}$  or  $9/2$ . The spin of the ground state of  $\text{Sm}^{149}$  has been measured<sup>18</sup> as  $\frac{7}{2}$ . This state is probably  $f_{7/2}$  as predicted by the single-particle shell model. Of the three spins which are consistent with the beta decay to the 285-keV level, only a value of  $\frac{5}{2}$  or  $\frac{7}{2}$  for this level is compatible with the  $M1$  character of the 0.285-MeV transition and the  $\frac{7}{2}^-$  character of the  $\text{Sm}^{149}$

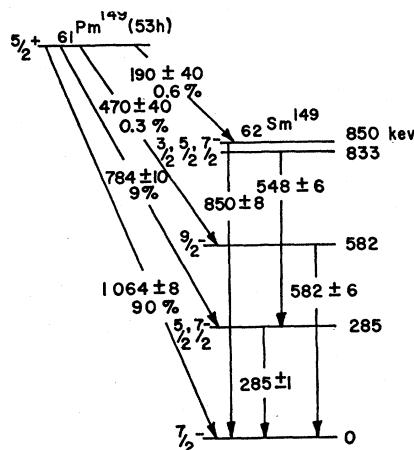


FIG. 4. Decay scheme for  $\text{Pm}^{149}$ . Energies are in keV.

ground state. The level at 580 keV must have a spin of  $9/2$  since, if it were to have the alternative spin  $\frac{1}{2}$ , then the 582-keV transition would be  $M3$  and thus its lifetime should be longer than the observed upper limit ( $<5 \times 10^{-8}$  sec). Any one of the three spins ( $\frac{3}{2}$ ,  $\frac{5}{2}$ ,  $\frac{7}{2}$ ), which are assigned to the 850-keV level on the basis of the beta decay, is consistent with the spins already assigned to the lower levels. However, if it has spin  $\frac{3}{2}$  then the 285-keV level could not have spin  $\frac{5}{2}$  since for that case a transition from the 850-keV level to the 285-keV level would be more probable than one to the ground state. Instead, the ground-state transition is found to be more probable.

<sup>18</sup> G. Bogle and H. Scovil, Proc. Phys. Soc. (London) A65, 368 (1952).

## APPENDIX

The resin was prepared in a column 0.6 cm in diameter and 20 cm high. The source material was dissolved in 0.1N HCl and then deposited on top of the resin. After the source material had been adsorbed by the resin, the latter was washed with 1M lactic acid which had been adjusted with ammonium hydroxide to a pH of 3.25. The lactic acid flowed through the resin at the rate of 0.15 ml/min and was collected in test tubes. The tubes were changed every 12 min, so that each contained about 2 ml of solution. The radiation emitted from each solution was counted by use of a NaI(Tl) crystal and a single-channel analyzer. The

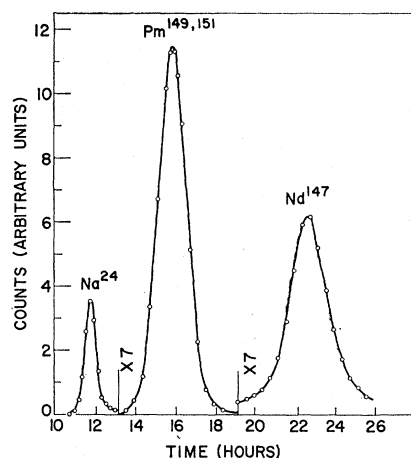


FIG. 5. Temporal relation between the activities separated by the ion exchange column. Time zero is when the lactic acid was added to the column.

counting rates obtained in this manner were then plotted as a function of the time that the solution had been collected in the tube. This plot is shown in Fig. 5. Three peaks, which represent activities that were eluted from the column at different times, are seen in the figure. The pulse-height distribution of the radiation which corresponded to the maximum counting rate for each peak was analyzed with the 256-channel scintillation spectrometer. In each of the three cases, the pulse-height spectrum of the emitted radiation was different from those associated with the other two fractions. The spectra were identified as being those characteristic of  $\text{Na}^{24}$ ,  $\text{Nd}^{147}$ , and  $\text{Pm}^{149} + \text{Pm}^{151}$ .