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A STUDY OF THE ISOTOPES OF PROMETHIUM
(Thesis)

Vera Kistiakowsky Fischer

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A STUDY OF THE ISOTOPES OF PROMETHIUM

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ABSTRACT

Bombardments of isotopically enriched neodymium samples have been performed with 8.9 Mev protons from the 60-inch cyclotron of the Crocker Radiation Laboratory and with protons at higher energies from the linear accelerator of the Radiation Laboratory of the University of California. Praseodymium has been bombarded at various energies with alpha particles from the 60-inch cyclotron. The half-lives and radiation characteristics of the promethium isotopes produced from these bombardments have been measured. The isotopes were identified chemically and their mass allocations were determined on the basis of their relative yields.

Nuclear shell theory was applied to explain ambiguities and to estimate the decay characteristics of unobserved promethium isotopes. Indirect confirmation of the theory was obtained in the coherence of the results.

The following nuclides were characterized for the first time: Pm^{141}, Pm^{146}, and Pm^{150}. In addition, work on promethium isotopes previously described gave results indicating errors in the assignments of Pm^{143} and Pm^{144} and in the half-life of Pm^{149}. The previous characterizations of Pm^{146}, Pm^{147}, and Pm^{148} were checked. A 42 day negatron emitting nuclide was observed to be an isomer of either Pm^{147} or Pm^{148}. Limits were set on the half-life of Pm^{142}.

A STUDY OF THE ISOTOPES OF PROMETHIUM

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I. INTRODUCTION

Promethium is one of the few elements which is not found in nature, and it is for this reason that a study of its radioactive properties has unusual interest. A knowledge of the history of its discovery and of the explanations offered for its instability are invaluable to further attempts at clarifying the situation. Thus the first portion of this paper will deal with these, as well as summarizing the results obtained by previous experimenters.

A. History of Discovery

By 1925 the periodic table was complete except for the elements, unobserved in nature, at atomic numbers 43, 61, 85, and 87. The contemporary experimental methods were not sufficiently developed to be infallible and some investigators found what they thought were stable isotopes fitting in these gaps.[1] At least twice, discovery of element 61 was claimed and on these occasions the names illinium (Il) and florentium (Fr) were proposed for it.[2] More recent studies with vastly improved methods of rare earth separation, however, indicate that these early "discoveries" were erroneous and that promethium does not have natural occurrence.[3]

The discovery of electron capture removed the requirement of instability greater than one electron mass for decay to a lower atomic number. Thus Jensen[1] was enabled in 1938 to predict on the basis of Mattauch's rule that elements 43 and 61 have no stable isotopes. This becomes obvious for 61 on consideration of Table 1.

Table 1

Isotopes[4] of Elements 60, 61, and 62

Element	Neutron Number														
	80	81	82	83	84	85	86	87	88	89	90	91	92	93	94
$_{60}Nd$	3.3 d K EC	2.42 hr β^+	stable	stable	stable	stable	stable	11 d β^-	stable	1.7 hr β^-	stable	12 min β^-			
$_{61}Pm$			285 d EC		30 yr EC		3.7 yr β^-	5.3 d β^-	47 hr β^-		27.5 hr β^-				
$_{62}Sm$			stable	150 d EC		stable	stable	stable	stable	1000 yr β^-	stable	47 hr β^-	stable	25 min β^-	10 hr β^-

A mass calculation using a modification of the Weizsäcker equation[5] gives A = 146.1 as the most stable mass number for Z = 61. Since 61^{146} is an odd-odd isotope, either 61^{145} (N = 84) or 61^{147} (N = 86) should be the closest to stability; however, Table 1 shows that both of these have neighboring stable isobars. Thus, either one of the naturally occurring isotopes of samarium or neodymium actually decays with a half-life greater than 3×10^9 years or the most stable isotopes of promethium are not stable.

The discovery of element 61 did not come, therefore, with improved methods of analysis alone, but rather with the development of particle accelerators and piles. The first activity attributed to 61 was produced by Pool and Quill[6] in 1937 from a Nd(d,n)61 reaction. This was a 12.5 hr beta decay which they thought to be 61^{144} but which has not been assigned since. It was not until 1941 that Wu and Segre[7] and Kurbatov, Pool, and co-workers[8-10] separately produced activities by various types of bombardments which resemble those recently proven to belong to 61. In an article reporting some supposed activities of 61, Bothe, another early investigator in this field, noted:[11] "In the region of the rare earths, the previous knowledge of artificial radioactivity has been sparser than in other parts of the periodic system, mainly because of the difficulty of obtaining sufficiently pure samples and even less those of well known (rare earth) composition."

In April of 1946 the problem was solved at last! Dr. Coryell disclosed that a 3.7 yr beta emitter from thermal neutron uranium fission had been chemically identified as an isotope of element 61.[12] This was made possible by the recently developed separation procedure

involving adsorption of a sample on the top of a column of cation exchange resin and subsequent elution (selective de-adsorption) with an ammonium citrate-citric acid buffer solution. Preliminary work was done by Ballou,[13] Goldschmidt and Morgan,[14] and Seiler and Winsberg[15] who found the activity and proved it to be either praseodymium, neodymium or 61. Davies[16] discovered the parent of the 3.7 yr isotope to be an 11 day rare earth activity which was confirmed by Hume and Martens.[17] The method of separation of the rare earths by ion exchange was then successfully applied to fission products by Marinsky, Glendenin, and Coryell.[18] They proved that the ~11 day activity belongs to neodymium and that the 3.7 yr activity belongs to its element 61 daughter, completing the official discovery of the element. The name promethium (Pm) (from the Greek God Prometheus, the fire-giver) proposed by this group[19] in 1947 was adopted by the International Congress at Amsterdam in September, 1949.

B. Explanations for Instability

The recent proposal of nuclear shell structure by Maria Mayer[20-22] and others, offered possibilities for an explanation of the absence of promethium in nature. Many excellent articles have been written on this subject, by Ballou,[23] Perlman, Seaborg, and Ghiorso,[24] and Broniewski[25] among others; but, for the sake of compactness, only the papers of Kowarski[26] and Suess[27] will be discussed since together they review the problem well.

Since all isotopes of promethium cluster around the closing of the 82 neutron shell, Kowarski[26] considered the effect that the proximity of the closed shell has on the normal pattern of stable isotopes with odd A. In general, this pattern consists of series of stable isotopes

differing from each other by a neutron-proton pair, since I = N-Z is thus kept a constant as close as possible to the value of I which gives maximum stability for the given values of A ($I_{max.\ stab.}$). I is greater than $I_{max.\ stab.}$ for the first members of such a group, corresponding to a neutron excess, while the reverse occurs for the last members. The proton excess finally forces the addition of two neutrons instead of an n-p pair again making I greater than $I_{max.\ stab.}$ and starting a new series. Usual parity rules make no distinctions between the addition of n-p, n-n, and p-p pairs.

This pattern is perturbed by low binding energy for neutrons to the nucleus immediately after a closed shell. There p-p and even n-n predominate over n-p additions. The pattern of stable nuclei surrounding promethium and the effect of the closing of the 82 neutron shell are illustrated by Table 2.

Table 2

Stable Odd A Isotopes[4] Near Promethium

N-Z = I	N									
	79	80	81	82	83	84	85	86	87	88
25			$_{56}Ba^{137}$	$_{57}La^{139}$			$_{60}Nd^{145}$		$_{62}Sm^{149}$	$_{63}Eu^{151}$
23	$_{56}Ba^{135}$			$_{59}Pr^{141}$	$_{60}Nd^{143}$		$_{52}Sm^{147}$			

Ba^{135} is the last regular member of the I = 23 series and then a n-n step gives $_{56}Ba^{137}$ with I = 25. A n-p step follows giving $_{57}La^{139}$ with a closed 82 neutron shell. The addition of a n-p pair is now no longer equivalent to a n-n or p-p addition since the pairing-off energy of the proton does not balance the very low binding energy for the first neutron

in the new shell. Because a p-p step is also favored over an n-n step, the next stable odd A isotope is $_{59}Pr^{141}$. However, there is now a proton excess and I is much smaller than $I_{max.\ stab.}$ The competition of this effect prohibits another p-p addition, and thus a p-n and an n-n step follow in succession. At $_{60}Nd^{145}$ the effect of the antipathy to open shell neutrons again becomes dominant and therefore the next stable isotope is $_{62}Sm^{147}$ differing by a p-p pair, rather than $_{61}Pm^{147}$ differing by an n-p. This is Kowarski's explanation of why there are no stable promethium isotopes. A similar situation occurs at technetium, and a generalization follows. "If a new series of constant I begins in the region of open-shell neutrons, then there is a tendency toward the elimination of stable isotopes of odd Z."

Suess[27] gave an explanation on slightly different grounds. At the closing of the 50 and the 82 neutron shells there are discontinuities in beta decay energy corresponding to the very large binding energies of the closed shell nucleons. Furthermore, there is no longer an equivalence between even-odd and odd-even nuclei, because the binding energies of a pair of neutrons and a proton are less than those of a pair of protons and a neutron. Thus for all $N > 50 > Z$ and $N > 82$ the beta decay energies for odd-even nuclei are higher than for even-odd. Suess found that at the 82 neutron shell the binding energy for an unpaired neutron is lowered by 2.0 Mev and that for a paired neutron, by 1.3 Mev. The Bohr-Wheeler parabola for an odd value of A gives way therefore at 82 neutrons to two parabolas at higher energies. This splits the line of maximum stability into two parts shifted different amounts toward the configuration of the closed shell nucleus and a

smaller number of open shell neutrons. Thus the most stable configuration for A = 145 is Z = 62 (N = 85) rather than Z_A = 61.3 (N = 85.7), while for A = 147, Z_A = 60.4 (N = 84.6). The effect is negligible, and the most stable configuration is Z = 60 (N = 85). Hence, Pm^{145} and Pm^{147}, the most stable isotopes of promethium, are not the most stable members of their isobar series.

These explanations are both highly empirical, of course, and a deeper understanding can follow only on a further knowledge of shell structure in general and of promethium isotopes in particular.

C. Previous Work on Promethium Activities

Rather than discuss the many experiments on promethium activities in detail, they are summarized in Tables 3 and 4. When necessary in connection with the present experimental study, the material will be amplified. These tables use the notation of Seaborg and Perlman.[4]

One isotope which the present bombardments were not designed to produce will be mentioned here, however. This is Pm^{151} which until October 1951 was postulated to be the 12 min negatron emitting isotope found on neutron irradiation of neodymium.[28,29] Then Rutledge, Cork, and Burson[30] reported its assignment to Nd^{151}. They investigated with a permanent magnet beta spectrometer the activities produced in a sample of neodymium enriched in Nd^{150} and bombarded by neutrons in the Argonne pile. The 12 min activity was found to consist of 1.93 Mev negatrons and 85.4 and 117.1 kev gamma rays (as calculated from the energies of the electrons they ejected from a lead adsorber). Since these show work function differences appropriate for promethium and since absorption measurements showed promethium x-rays associated with the 12 min activity,

the assignment to Nd^{151} follows. A 1.1 Mev negatron emitter with 27.5 hr half-life was also observed. The many gamma rays associated with it (Table 3) were found to have samarium work function differences and thus it was assigned to the Pm^{151} daughter of the 12 min activity (Table 3).

Table 3

Certain and Probable Promethium Isotopes Reported in Literature at Time Present Work Was Begun

A	Class	Type of Radiation	Half-Life	Energy of Particles	Energy of Electromagnetic Radiation	Produced by
143	B	EC, k, γ β^-, (?)[9,31]	285 $\pm$ 3 d[31] >100 d[8,9] ~200 d[7] 350 d[10,32] 1.7 yr[33]	0.7 Mev β^- (abs)(?)[32] No α or β^- [31] 0.74 Mev β^- (abs)[33]	0.95 Mev (abs)[31] 0.67 Mev (abs)[7] 0.7 Mev (abs)[10] 0.72 Mev (abs)[33] K, L x-rays[31,32]	Pr(α,2n)[7-9,31,32] Nd(d,n)[7-10]
145	A_{calc}	EC, e^-(?) K, L x-rays[34]	~30 yr[34] (calc)	low energy[34]		Sm(n,γ)Sm $\xrightarrow{\beta^+}$ [34]
147	A_{ms}[35]	β^- γ	~4 yr[13,28,36] ~3.7[15,18,35] 2.26 yr[15,36,38] (calc from fission yield Sm^{147})	~0.3 Mev (abs)[28] 229 $\pm$ 1 kev (spec)[40] 227 $\pm$ 1 kev (spec)[39] 223 $\pm$ 1 kev (spec)[41,42] ~0.2 Mev (abs F)[13-15,18]	no γ[18,15]	U(n)Nd^{147} $\xrightarrow{\beta^-}$ [13,14,18,28,35,37] Nd(n,γ)Nd^{147} $\xrightarrow{\beta^-}$ [18,28] U^{233} (n, high energy fission)[36]
148	A_{ms}[43]	β^- γ	5.3 d[10,18,43]	2.5 Mev (abs)[18,43] ~2 Mev (abs)[10]	0.8 Mev (abs)[18,43]	U(n)[18,43] Nd(n,γ)Nd^{148} $\xrightarrow{\beta^-}$ [43] Nd(d,2n)[10,43] Nd(p,n)[10,43] Nd(α,p)[43]
149	A_{ms}[44]	$\beta^-\gamma$ e^-(?)[18] x-ray	47 hr[7,11,18,28,45,47] 49 hr[46] 50 hr[33] 55 hr[44]	1.1 Mev (abs)[11,18,28] 0.95 Mev (abs)[7] 0.98 Mev (abs)[45]	~0.2 Mev (abs)[45] 0.25 Mev (abs, low intensity)[18,28] x-rays[18,28,45]	Pu(n)[47] U(n)[18,28,44] Nd(n,γ)Nd^{149} $\xrightarrow{\beta^-}$ [11,18,28]
151	B^{30}	$\beta^-\gamma$[29,30]	27.5 hr[30]	1.1 Mev[30]	615 kev (abs, weak)[30] 64.7, 65.8, 69.6, 99.9, 116.2, 105.0, 163.0, 168.0, 177.0, 208.3, 231.9, 239.9, 275.2, and 340.1 kev (spec)[30]	Nd^{150}(n,γ)Nd^{151} $\xrightarrow{\beta^-}$ [29,30]

Table 4

Less Certain Promethium Isotopes Previously Reported in Literature

A	Class	Type of Radiation	Half-Life	Energy of Particles	Energy of Electromagnetic Radiation	Produced by
142		β^+ or EC	<20 min[32] <5 min[31]			Pr(α,3n)[31,32]
144	F	β^+[32]	4.1 ± 0.1 hr[32]	1.3 Mev[32]		Pr(α,n)[32]
146-150	C	β^-,γ[48]	42 ± 1 d[48]	~0.5 Mev (95%), ~2.5 Mev (5%) (abs)[48]	~0.9 Mev (abs)[48] no K or L x-ray[48]	U(p,high energy)[48] fission
146,149,150	E	$\beta^-\gamma$[10,18]	2.7 hr[10,18] 2.3 hr[8,28]	2 Mev (abs)[10]	γ[10,18]	Nd(d,n)[10,18] Nd(p,n)[10,18]
	F	β^-	10.3 d[28]			U(n)[28]
	F	β^-	12.5 hr[6]			Nd(d,n)[6]
	F	β^+ or β^-,γ	16 d[10,18]	1.7 Mev (abs)[10]		Nd(d,n)[10,18]
	F	β^+ or β^-	41 ± 1 hr[9]	2.9 Mev (abs)[9]		Nd(d,n)[9] Nd(p,n)[9] Nd(γ,n)N $\xrightarrow{\beta^-}$ [9] Pr(p,n)N $\xrightarrow{\beta^-}$ [9]

II. EXPERIMENTAL PROCEDURES

A. Discussion

These experiments were undertaken in an attempt to ascertain the mass assignments of the promethium activities. The availability of mass spectrographically separated neodymium isotopes[49] made Nd(p,n)Pm reactions an attractive method for producing the various isotopes. Their mass numbers were to be determined from the relative yields in the different samples.

Since it was felt that the possibilities of Pr(α,xn)Pm bombardments had not been exhausted by previous workers, this method was also utilized. Assignments were to be made on the basis of the changes in cross section with energy and by comparison with the neodymium bombardments.

B. Bombardments

Neodymium Targets

The chemical and isotopic characteristics of the separated neodymium isotopes are given in Table 5. They were received and bombarded in the form of the oxide (Nd_2O_3).

The bombardments were performed by the 60-inch cyclotron of Crocker Laboratory and by the linear accelerator of the Radiation Laboratory, both at this University.

The samples for the 60-inch cyclotron were prepared in most cases by placing 5 to 10 milligrams of the oxide in an envelope of 0.00025 inch platinum. This was then placed in a platinum dish which was covered by a disk of the same foil and clamped in the "pistol grip" assembly described by S. G. Thompson.[50] The powder was placed in an envelope rather than directly in the dish primarily because such small quantities

Table 5

Chemical and Isotopic Characteristics of Neodymium Isotopes[49]

A	% 142	% 143	% 144	% 145	% 146	% 148	% 150	Chemical Purity and % Contaminants
142	93.00 ± 0.03	3.18 ± 0.02	2.89 ± 0.02	0.368 ± 0.008	0.414 ± 0.06	0.084 ± 0.007	0.066 ± 0.008	99.5 La<0.15 Ni<0.08 Mg<0.02 Pt<0.07
143	4.04 ± 0.02	83.93 ± 0.04	8.83 ± 0.05	1.78 ± 0.01	1.16 ± 0.03	0.149 ± 0.003	0.108 ± 0.006	99.5+ La<0.15 Mg<0.02 Ni<0.08
144	0.698 ± 0.090	0.941 ± 0.072	93.45 ± 0.07	2.33 ± 0.04	2.36 ± 0.05	0.147 ± 0.017		99+ La<0.15 Mg<0.02 Ni<0.08 Sm<0.31
145	1.18 ± 0.02	0.840 ± 0.022	4.83 ± 0.05	78.60 ± 0.06	3.71 ± 0.08	0.626 ± 0.011	0.214 ± 0.001	99+ La<0.15 Ni<0.08 Mg<0.02 Sm<0.31
146	0.584 ± 0.037	0.274 ± 0.007	0.903 ± 0.014	1.03 ± 0.01	95.60 ± 0.06	1.42 ± 0.01	0.192 ± 0.050	99+ La<0.15 Ni<0.08 Mg<0.02 Sm<0.31
148	1.21 ± 0.03	0.584 ± 0.006	1.28 ± 0.03	0.742 ± 0.040	2.46 ± 0.03	89.85 ± 0.08	3.88 ± 0.05	99.5 Ba<0.04 Ho<0.31 Fe<0.08 Mg<0.02 Mg<0.04
150	1.10 ± 0.11	0.456 ± 0.048	0.981 ± 0.026	0.419 ± 0.014	1.04 ± 0.05	1.25 ± 0.01	94.76 ± 0.02	99.9 Mg<0.02 Si<0.08

were used. This method, however, also expedited the removal of the sample and protected it from possible water leaks from the cooling system. The target was bombarded in the deflected beam. Differential beam current readings taken during each bombardment yielded an average value of 3 microamperes for the beam intensity. The 10 Mev protons from the cyclotron were reduced to approximately 8.9 Mev by 1.25 inch of air, 0.003 inch of duraluminum and 0.0005 inch of platinum between the snout and the sample.[51] Though the potential barriers of the neodymium isotopes against protons were calculated to range from 9.4 to 9.5 Mev with decreasing mass number, an exploratory bombardment showed that this energy produced promethium activities in adequate if low yields.

The linear accelerator was used in bombardments of neodymium 142 where higher energies were found to be necessary. It was not used in all the bombardments because its average beam current is approximately 0.07 microamperes, a factor of 50 or more lower than that of the 60-inch cyclotron. The 60-inch beam would probably cancel or outweigh any advantage gained in yield by bombarding at higher energies for (p,n) reactions.

Samples for linear accelerator bombardments were prepared by wrapping 5 to 10 milligrams of Nd_2O_3 in a 0.00025 inch platinum foil envelope of approximate dimensions 3/16 x 3/16 inch. This was centered in the hole of a 2.5 x 3.5 x 1/16 inch aluminum backing plate on scotch tape with one thickness of platinum facing the beam. The bombardment time was 10 minutes and thus the scotch tape did not deteriorate to any large extent. This assembly was placed in the beam measuring Faraday cup at the 10° port, aligning the target center with the beam. 1147 mg/cm^2 of aluminum absorbers reduced the energy of the protons from 32 Mev to

a narrow distribution between 10 and 11 Mev. In later bombardments, 700 mg/cm^2 of aluminum absorbers were used to give ~20 Mev protons, or the full energy protons were utilized.

Praseodymium Targets

Ten to twenty milligram samples of spectroscopically pure[52] praseodymium oxide[53] (Pr_6O_{11}) were wrapped in platinum foil envelopes. These were bombarded with alpha particles from the 60-inch cyclotron in a "pistol grip" type assembly in the same manner as the neodymium targets. Varying thicknesses of platinum were used to cover the dish in which the sample was placed. The alpha particles, reduced to approximately 37 Mev by 1.25 inch of air and 0.003 inch duraluminum,[48] had energies shown in Table 6 after passing through the various thicknesses of cover foil.

Table 6

Energy of Alpha Particles Striking the Sample[51]

Platinum Dish Cover (inch)	Energy of Alpha Particle (Mev)
0.50×10^{-3}	35.5
1.25×10^{-3}	32.7
2.25×10^{-3}	26.0
3.00×10^{-3}	18.0
3.25×10^{-3}	15.2

The potential barrier of Pm^{141} against alpha particles was calculated to be 17 Mev, and the thresholds for the (a,n), (a,2n), and (a,3n) reactions were 3.3 Mev, 11.6 Mev, and 19.2 Mev, respectively.

Delivery of Target

The targets were rushed from the bombarding machine to the author's laboratory by the Health Chemistry group. Since the delivery time was five minutes from the linear accelerator and seven from the 60-inch cyclotron, this procedure was adequate for the detection of activities with half-lives above one minute.

C. Chemical Separation

Discussion

At the proton energies used the only probable reactions with neodymium are: (p,n) and (p,2n). The bombardments of praseodymium would be expected to result only in (a,xn) and (a,pxn) reactions. Since the latter result in stable neodymium isotopes for $x \leq 2$ and because of the high purity of the samples, both types of bombardments should yield no rare earth activities other than promethium.

Other activities which might interfere with the studies are the following.

From oxygen:

$$O^{17}(p,n)F^{17} \xrightarrow[66\ sec]{\beta^+}$$

$$O^{18}(p,n)F^{18} \xrightarrow[112\ min]{\beta^+}$$

$$O^{17,18}(a,xn)Na^{22} \xrightarrow[2.6\ yr]{\beta^+}$$

From carbonate:

$$C^{13}(p,n)N^{13} \xrightarrow[10.1\ min]{\beta^+}$$

$$C^{12,13}(a,xn)O^{14} \xrightarrow[76\ sec]{\beta^+}$$

$$C^{12,13}(a,xn)O^{15} \xrightarrow[118\ sec]{\beta^+}$$

From the platinum envelope:

$$Pt(a,xn)Hg^{199} \xrightarrow[44\ min]{IT.}$$

and other reactions producing longer lived gold and mercury isotopes.

Chemical Procedure for Short Half-Lived Isotopes

In most cases the sample was transferred directly from the platinum envelope to a counting dish and covered with a very thin film of zapon. This procedure consumed three minutes for a linear accelerator assembly and four minutes for a 60-inch cyclotron target.

In order to decrease the non-promethium activities, some samples were chemically treated as outlined below.

a. Dissolve in concentrated HCl

b. Dilute and precipitate neodymium hydroxide with NH_3 gas

c. Centrifuge and wash the precipitate

d. Dry on a counting dish

An alternative method was to precipitate the oxalate instead of the hydroxide. Both procedures required an average of ten minutes, and gave 50 percent purification from all but rare earth contaminants. A more complete chemical procedure used on occasion was the following.

a. Dissolve in concentrated HNO_3

b. Dilute and precipitate neodymium fluoride with HF in a lusteroid tube

c. Centrifuge and wash the precipitate

d. Dissolve in concentrated HNO_3 saturated with H_3BO_3

e. Dilute and precipitate neodymium hydroxide with NH_3 gas

f. Centrifuge and wash the precipitate

g. Dry on a counting dish

This required twenty minutes.

Chemical Procedure for Ordinary Bombardments

The first six steps of the second procedure outlined in the last paragraph were carried out with targets from both neodymium and praseodymium bombardments. They should remove all activities from the sample except those of the rare earths, the precious metals and mercury. Separation from these was obtained by dissolving the hydroxide precipitate in concentrated HCl and sucking this solution through a column of anion exchange resin. This was constructed by agitating Dowex A-1 resin[54] with concentrated HCl and then packing it into the tube of a funnel whose end had been drawn to a tip (Fig. 1a). In the concentrated HCl the precious metals and mercury form chloride complexes which are quantitatively adsorbed on the resin.[55] The rare earth ions, however, do not adhere appreciably. After washing any remaining rare earths off the column with an extra volume of concentrated HCl, the acid containing the rare earth sample was diluted and the hydroxide precipitated with NH_3 gas. This was centrifuged, washed, and either prepared for counting or for a cation column separation.

Rare Earth Separation

In cases where rare earth separation was desirable for positive identification of an activity, the method of high temperature elution from a cation exchange resin column was used.

The application of the general method to the rare earths used in these experiments was developed by Dr. K. Street, Jr.[56] Since the particulars for separation of neodymium, promethium, and samarium have not been given elsewhere, they will be detailed in this section. Figure 1b indicates the operation of the apparatus. The characteristics of

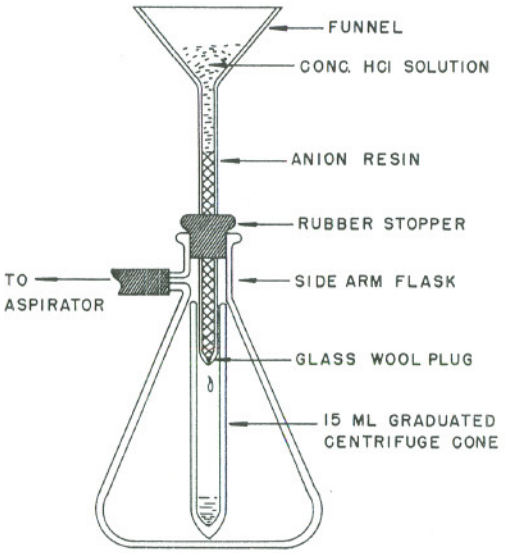

FIG. 1a

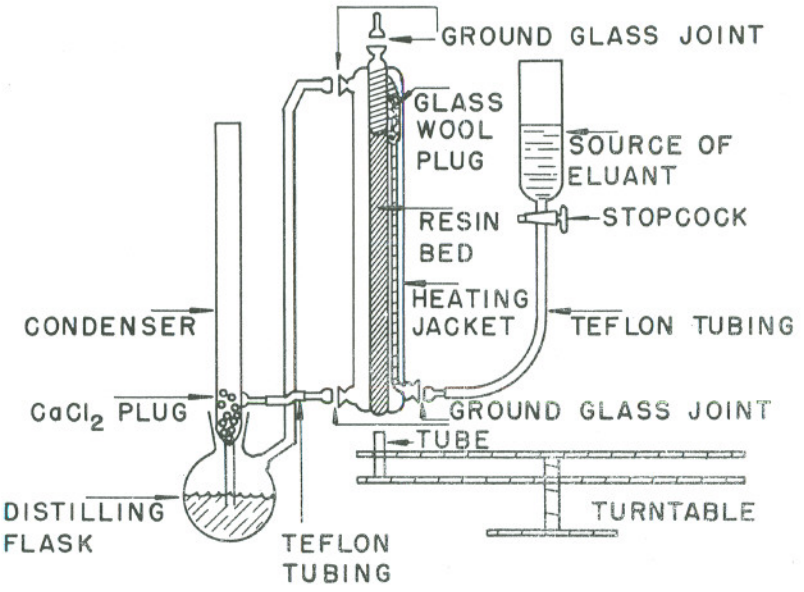

FIG. 1b

(a) Anion column for removing the activities of the precious metals and mercury from a rare earth sample
(b) Cation column for separation of the rare earth elements

the column and the elution follow.

(a) Resin bed:

Dowex 50^{51} resin, medium-fine grade

Height column: 22 cm

Diameter column: 1 cm

Free column volume: 12.4 cc

(b) Drop size: 0.045 cc or 0.0027 free column volumes

(c) Temperature: trichloroethylene condensation point: 87°C

(d) Eluant: pH 3.52 citric acid buffered with ammonium citrate: citrate concentration 0.25 M. Elutions were attempted at other pH's and this alone was found to give both adequate separation and speed.

The hydroxide sample was dissolved in a very small volume of 6 N HCl which was then diluted to 0.1 N hydrogen ion concentration. Approximately 0.2 ml resin was added and the mixture was agitated and heated for 15 minutes. After centrifuging and washing the resin it was placed at the top of the column in as even a layer as possible. The eluant was then allowed to flow through and was collected in 10 x 75 mm rimless glass tubes placed in a perforated automatic turntable, 5λ aliquots were dried on aluminum disks and counted for Geiger activity while 50λ aliquots were submitted for spectrographic analysis.[57]

A calibration run was performed with a sample containing 3 mg neodymium, 2 mg samarium, and promethium tracer (Pm^{147} 3.7 yr, 0.223 Mev β^-). The flow rate was one drop every two minutes or 0.0199 cc/min/cm^2. The elution curve obtained is shown in Fig. 2. While the separation is far from complete, it was more than sufficient for the purposes of these experiments where identification rather than isolation was desired.

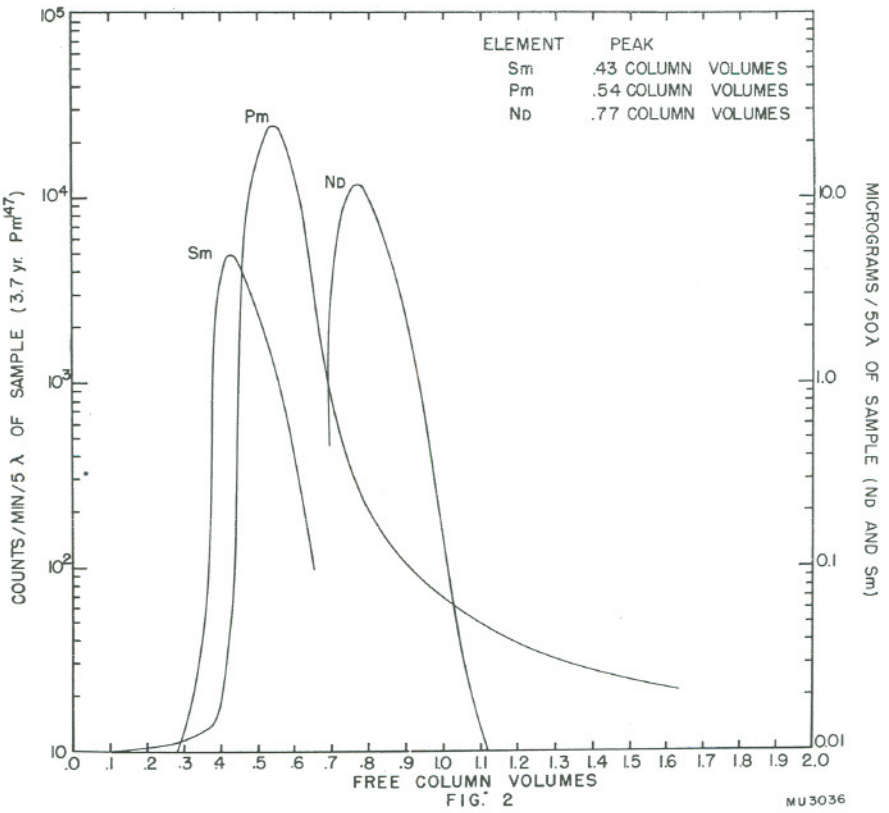

FIG. 2

Column calibration with Pm^{147} tracer and macro amounts of neodymium and samarium determined spectroscopically

In the actual separations the presence of macro amounts of neodymium (added in the case of praseodymium samples) made it possible to use a spectrographic landmark to identify the activity peak. Since in the calibration experiment it required twenty hours for complete elution of the promethium, the drop rate in the actual separations was increased two to four fold. This decreased the elution time to twelve and eight hours, respectively, and permitted the observation of shorter half-lives. The elution peaks were naturally smeared out by this procedure and thus the decay of samples from various parts of the activity peak was compared. If the decay rates were identical, it was accepted as conclusive evidence that the peak contained the activities of one element alone.

The rare earth samples were recovered from the citric acid by heating the eluate with five times its volume of concentrated sulfuric acid with a few drops of 6 N selenium solution until a clear black solution was obtained. Then perchloric acid was added dropwise until the solution became colorless. After cooling and diluting, the rare earth hydroxides were precipitated with NH_3 gas.

D. Radioactivity: Measurements and Calculations

Geiger Counter Measurements

An end-on Amperex tube (type 100C) with a 3.5 mg/cm^2 mica window filled with 33 $\pm$ 5 cm of argon and <1 percent of chlorine was used with a Nuclear Instrument Corporation scaler (AEC Model CGM-7H). It had a plateau from 1200 to 1500 volts and was operated at 1300 volts.

The coincidence correction for this unit was determined as a function of counting rate. A very high activity sample of the 65 hr 2.35 Mev negatron emitter Y^{90} was prepared by separation from a solution

of its parent, Sr^{90}. The counting rate of this sample was determined as a function of time for all shelves until less than five thousand counts per minute were observed. A counting efficiency was assumed for the lowest counting rates and after correcting with this factor, values were obtained by correcting for decay back to a given time. These were compared with the values observed at that time and the coincidence correction was calculated from the difference. Varying the value assumed for the low counting rate counting efficiency until a consistent set of corrections was obtained, the value 0.3 percent per thousand for counting rates up to 35,000 counts per minute resulted. At higher counting rates, the correction increased.

1. Preparation of Sample.-- Samples were prepared by drying the activity-containing neodymium or praseodymium hydroxide or citrate solution on 0.001 inch aluminum disks stamped into dishes. When the absolute counting rate was desired these were supported by 1/16 inch aluminum holders which gave saturation backscattering, the correction factors for which have been reported by Yaffe[58] as a function of energy.

When a cross section determination was desired, the dish was weighed to 0.01 milligrams before and after addition of the sample to give the weight of the sample by difference.

2. Decay.-- The decay of short-lived isotopes was sometimes followed with a traffic-counter type of automatic recorder. Otherwise, each decay point was counted for at least five thousand counts. These determinations had a standard statistical error of 1.4 percent.

A metallic uranium standard was used to correct for variations in counting efficiency as previously described.[32]

3. Counting Efficiency.-- The values for the counting efficiencies of various types of radiation were taken from H. Hicks.[32]

Particles:	100%
L x-rays:	8.0 to 10.0%
K x-rays and gamma rays to 0.5 Mev:[59]	0.5%
Gamma rays above 0.5 Mev:	1.0% per Mev

4. Calibration.-- In order that absolute counting rates might be obtained, the counter geometry was calibrated with a Bureau of Standards Radium D + E negatron standard. This was counted for over 10^5 counts on shelf two with a 3.33 mg/cm^2 absorber on shelf one. After correcting for absorption in the counter window, the air gap, and the aluminum, the counting rate was divided by factors for back-and self-scattering. The corrected value was compared with the calculated disintegration rate and a 3.4 ± 0.3 percent geometry on shelf two was calculated.

5. Self-Scattering.-- Self-scattering factors were obtained as a function of the weight per cm^2 of the sample and the energy of the particle emitted, from curves prepared by W. Nervick.[60]

6. Absorption Measurements.-- In order to determine the energy of particles emitted by the sample, aluminum absorption measurements were taken in the conventional manner. L x-rays were detected by placing enough beryllium above the sample to remove all the particles and repeating the aluminum absorption measurements. After subtracting the L x-ray and hard electromagnetic components, the energies of the particles were obtained from their absorption half-thicknesses. In the cases where the absorption data curve was not linear, energies were determined from ranges.

Copper absorption measurements were used to determine the x-ray half-thicknesses and energies accurately. Enough beryllium was again placed above the sample to absorb all the particles emitted.

Both K x-rays and gamma rays were measured by determining their half-thicknesses in lead. These absorption measurements were performed in an unshielded counter as previously described.[32]

The ratios of the amounts of the various types of radiation emitted by a sample were calculated by extrapolation to zero absorber and application of the following corrections.

Particles: absorption in air gap and counter window, back- and self-scattering

K and L x-rays: absorption in beryllium, air gap and counter window, counting efficiency and Auger effect[59]

Gamma rays: counting efficiency

In the case of electron capture, the decay was assumed to be due to K capture alone and the values in the ratio were divided by the number of K x-rays. In the case of particle emission, the ratio was normalized to the most convenient value. The major sources of error arise in (a) the assumption regarding electron capture and (b) the assumed counting efficiencies.

7. Cross Section Calculation.-- Although the determinations of the beam intensity were very uncertain and in spite of the fact that some bombardments were interrupted by failure of the machine, it was considered worthwhile to calculate approximate cross sections for the 60-inch cyclotron bombardments.

H. Hicks[32] has detailed the calculation for the cross section in the case of a product isotope undergoing electron capture. In the

present work the following modification was used to include various types of decay of the product isotopes.

Electron capture alone or with positron emission:

$$N_{\text{counts/minute}} = (\text{counts/minute of K x-rays}) \times (\text{Auger coefficient}) \times \left(\frac{100}{\% \text{ contribution from electron capture}}\right)$$

Particle emission alone:

$$N_{\text{counts/minute}} = (\text{counts/minute of particle}) \times \left(\frac{100}{\% \text{ backscattering}}\right) \times \left(\frac{100}{\% \text{ self-scattering}}\right) \times \left(\frac{100}{\% \text{ contribution from particle}}\right)$$

All cases:

$$A_{\text{atoms at saturation}} = N \times \left(\frac{100}{\% \text{ counting efficiency}}\right) \times \left(\frac{100}{\% \text{ geometry}}\right) \times (\text{saturation factor}) \times (\text{decay since end of bombardment}) \times (\text{length of bombardment in min})$$

$$n_o \text{ bombarding particles/cm}^2 = (\text{microampere hours of bombardment}) \times \left(\frac{\text{no. of particles/microampere hour}}{\text{area of target in cm}^2}\right)$$

$$n_t \text{ target atoms} = \frac{(\text{grams of sample counted}) \times (\text{Avogadro's number})}{1/n \times (\text{molecular weight of sample } R_nO_m)}$$

$$\sigma_{\text{barns}} = \frac{A}{n_o n_t} \times 10^{24}$$

Examination of Low Energy Electromagnetic Radiation

In a few cases sufficiently active samples could be prepared and were examined by D. Martin[61] on a scintillation spectrometer. Very late in the research a scintillation counter-pulse discriminator gamma

spectrometer became available. Several samples were investigated with this. The energy-current calibration was obtained by the use of the following standards: Cs^{137}, 0.661 Mev gamma rays; Na^{22}, 1.26 Mev gamma rays; and Am^{241}, 59 kev x-rays. Ratios of the various components were obtained by graphical integration.

Measurement of Particles

1. Alpha Particles.-- Samples which might have contained alpha emitters were counted for the period of an hour on a conventional alpha counter.

2. Beta Particles - General.-- A crude beta ray spectrometer was used to identify the charge of the particles emitted and to give a rough estimate of the energies. It was also used with a traffic counter to measure the decay rate of beta particles at a given charge and energy.

3. Beta Particles - Energy.-- The negatron spectra of two samples were examined on the double-focusing beta spectrometer with radius of 25 cm described by G. D. O'Kelley.[62] Since a minimum of 10^7 counts/minute was required for a good measurement, and since the samples decayed with a 3 hr half-life, it was not possible to prepare a carrier free sample by column separation. Thus just enough neodymium hydroxide (carrying the promethium) to give sufficient activity was dried on a 1.5 mg/cm^2 mica film glued to the sample holder ring and covered with a thin Tygon film. This amount of backing would badly distort the low energy end of the beta spectrum but would not be expected to appreciably effect particles with energies greater than 1 Mev.

The K and L electrons of Cd^{109} and the K electron of Cd^{137} were used as known points from which the spectrometer current was calibrated

in terms of Hρ.

The sample was counted at 0.2 milliampere intervals from 1.0 milliampere until a constant background value was reached (Fig. 7). After subtracting the background, all the data were corrected for a 161 min decay. A Fermi-Kurie plot $\left[\sqrt{N(H\rho)/f(Z,\eta)} \text{ vs } E\right]$ (Fig. 8) was made using the formulae given by Feister.[63] The relativistic Fermi function, $f(62,\eta)$, was found by graphical interpolation between values given for $f(60,\eta)$ and $f(70,\eta)$ in the same article. The curve was resolved by successively subtracting the two longer components as indicated.

$$\sqrt{\left[\frac{N(H\rho)}{f(62,\eta)}\right]_{\text{total}} - \left[\frac{N(H\rho)}{f(62,\eta)}\right]_{\text{component}}} = \left[\sqrt{\frac{N(H\rho)}{f(62,\eta)}}\right]_{\text{difference}}$$

The "difference" was then replotted. The third component was known to have a 56 hr half-life and so the second "difference" was corrected for such a decay from the previous 2.7 hr correction.

The endpoints of the components gave their energies, while the ratio of the amounts of each present were obtained by comparing the squares of their values of

$$\sqrt{\frac{N(H\rho)}{f(62,\eta)}} \text{ at } E = 0$$

III. DISCUSSION OF RESULTS

A. Proton Bombardments of Neodymium Enriched in Nd^{150}

In the present work 161 min (2.7 hr) and 54 hr activities were produced by bombardment with protons from the 60-inch cyclotron of neodymium enriched in Nd^{150}.

Previous experimenters had not assigned a half-life to Pm^{150}. An isotope with 2.7 hr half-life emitting a 2 Mev negatron[10,18,28] was reported[61] and assigned tentatively to Pm^{146} and to an isomeric state of Pm^{149} or Pm^{147}.[10,18] The 2.7 hr activity was produced by Nd(d,n), Nd(p,n) and Nd(α,p) bombardments and was thought to be promethium since its chemistry identified it as a rare earth.[10,18]

An activity with a half-life generally reported as 47 hr was proven to belong to Pm^{149} by ion exchange separation and mass spectrographic analysis.[44] It is found among the products of thermal neutron uranium fission[18,28,44] and from the $Nd(n,\gamma)Nd^{149} \xrightarrow{\beta^-}$ reaction.[11,18,28] An approximately 1.0 Mev negatron[7,11,28] and a low intensity 0.25 Mev gamma ray[18,28] comprise its radiations. Inghram and co-workers[44] found a 55 hr half-life value for the mass spectrographically separated activity.

The 161 min and the 54 hr activities observed in the present experiments have both been characterized in detail and have been shown chemically to belong to promethium isotopes. On the basis of cross section considerations and previous work, assignments respectively of Pm^{150} and Pm^{149} have been chosen (see Table 9).

Measurements

The decay of isotopes produced from neodymium samples enriched in Nd^{150} bombarded with protons from the 60-inch cyclotron was composed of

161 ± 1 min and 54 ± 2 hr components (Fig. 3). These values are the average of five determinations.

Aluminum absorption measurements shortly after bombardment, with the data corrected for 161 min decay (Fig. 4), yielded a curve showing a hard beta particle with half-thickness 135 mg/cm^2 (2.1 Mev) (range 1077 mg/cm^2, 2.2 Mev). Subtracting this component from the total absorption data curve and correcting the difference to a 54 hr decay, a particle with half-thickness 40 mg/cm^2 (0.9 Mev) (range 322 mg/cm^2, 0.8 Mev) was observed. Three determinations gave these values.

Lead absorption measurements also shortly after bombardment and corrected for 161 min decay (Fig. 5) showed electromagnetic radiation with half-thickness ~46 mg/cm^2 (40 kev), ~1.36 g/cm^2 (0.33 Mev), and ~11.38 g/cm^2 (1.4 Mev). These values represent averages of three such absorptions. Forty kev is within experimental error of the energy of the K x-rays of samarium.

Aluminum absorption measurements at least 36 hr after bombardment (Fig. 6) showed a single beta particle with half-thickness 52 mg/cm^2 (1.0 Mev) (range 476 mg/cm^2, 1.1 Mev). Again three absorptions gave these values.

The electromagnetic radiation of the sample after the 161 min activity had decayed out was too low for absorption measurements. Examination with a scintillation spectrometer[61] indicated a very low intensity gamma ray with an energy of approximately 0.3 to 0.35 Mev and samarium K x-rays.

When the 161 min activity was examined on a crude beta spectrometer, a complex negatron spectrum containing some particles of at least 3 Mev

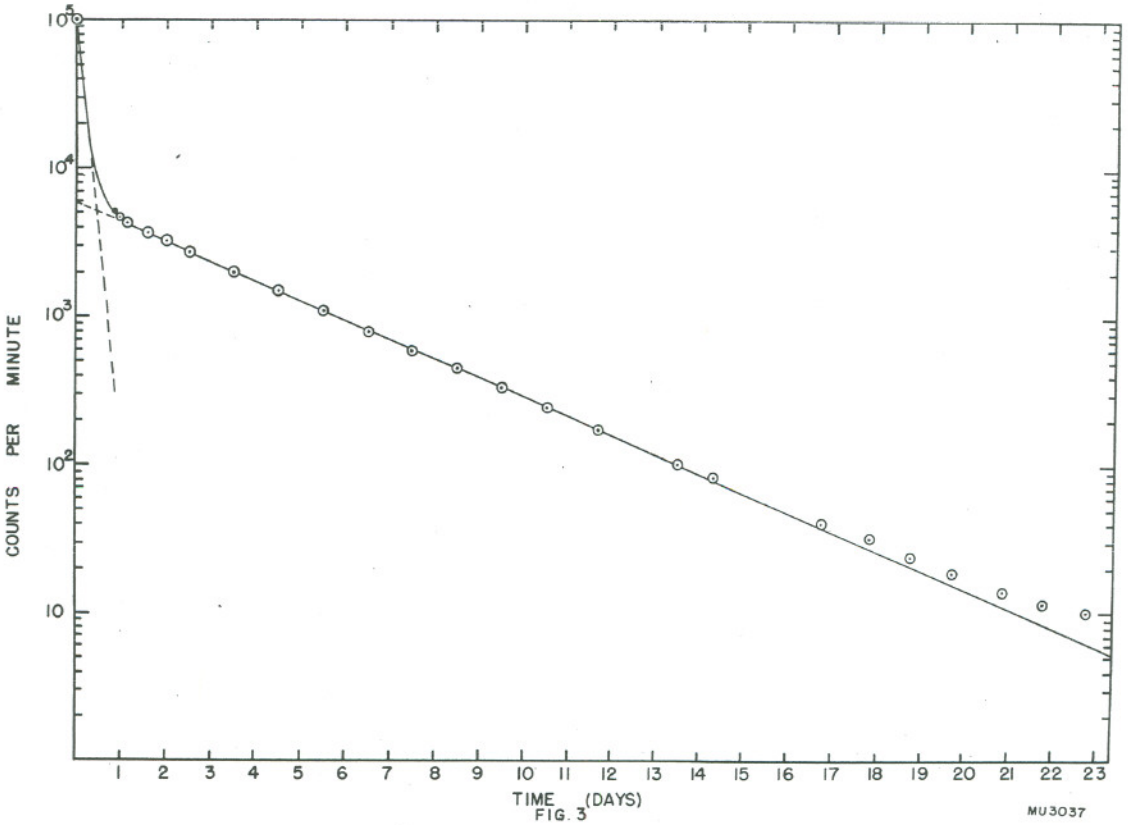

FIG. 3

Decay of the 161 minute and 54 hour activities from Nd^{150} + p bombardments

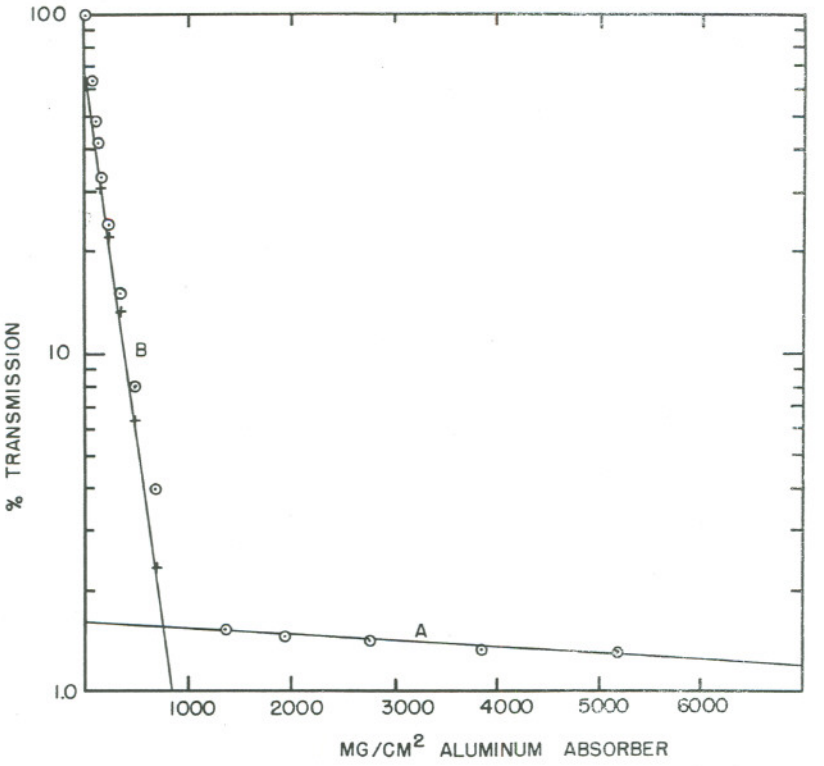

FIG. 4a

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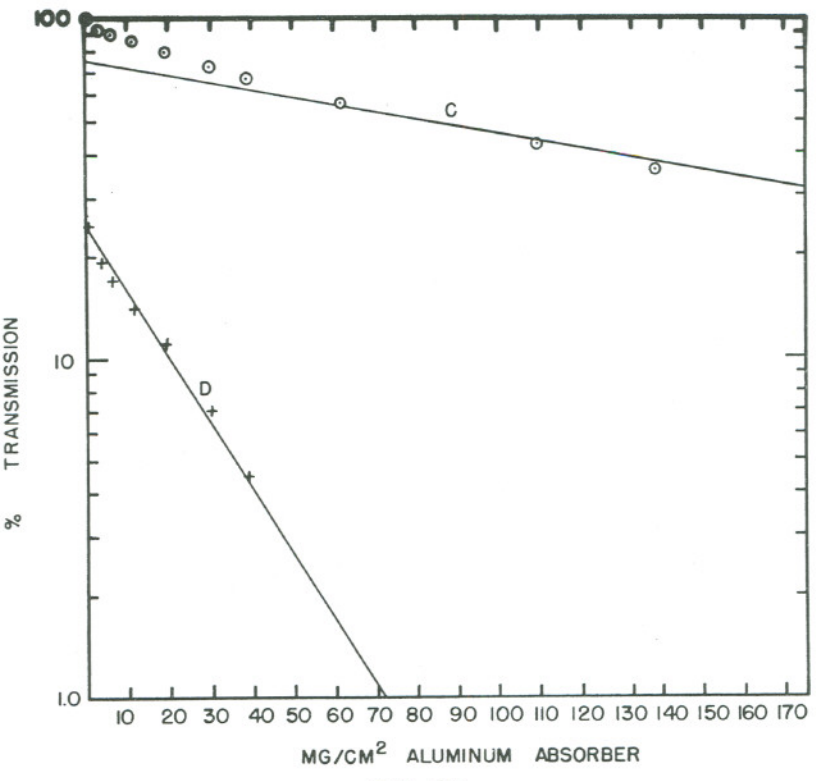

FIG. 4b

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Aluminum absorption data curve of a mixture of the 161 minute and the 54 hour activities from Nd150 + p bombardments

A. Absorption data corrected for 161 minute half-life decay. Gamma ray (A), 2.05 Mev negatron (B)

B. Same absorption data but uncorrected for decay. Sum of (A) and (B) corrected to the value they would have without undergoing 161 minute decay (C), 1.05 Mev negatron (D)

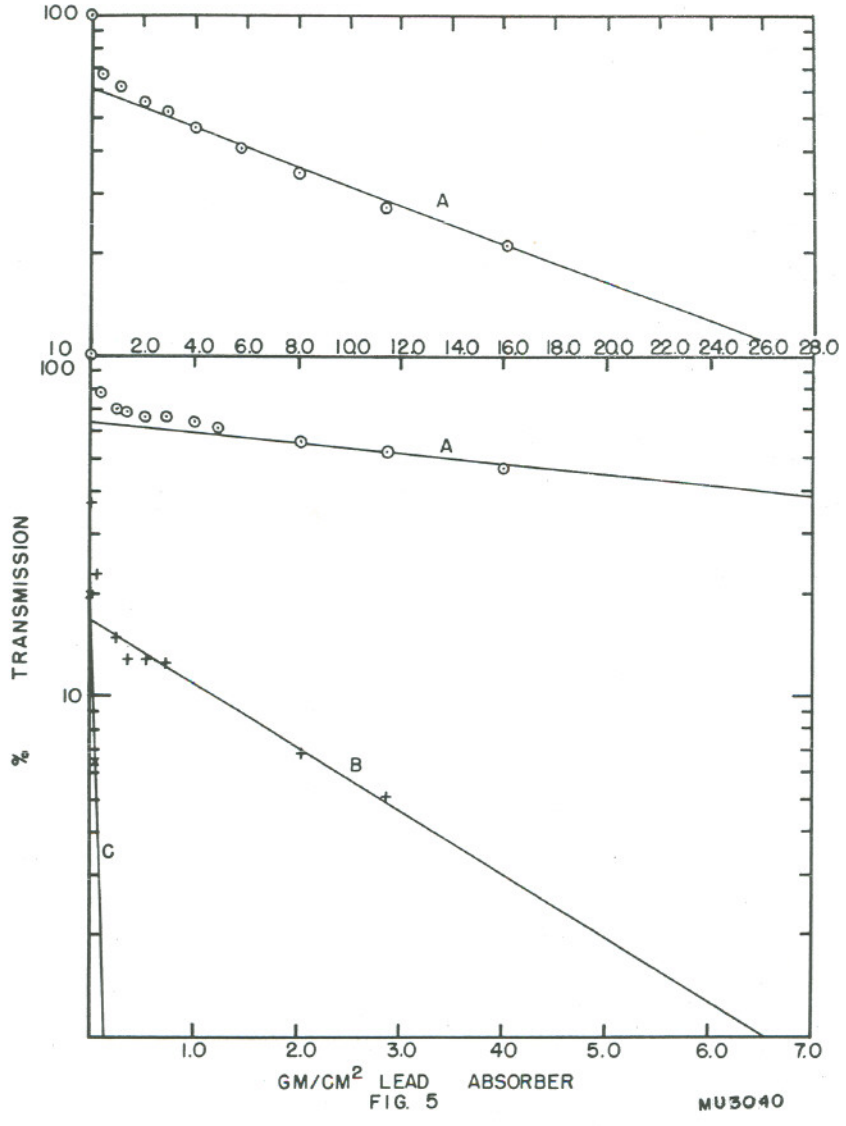


FIG. 5

Lead absorption data curve of the 161 minute activity from Nd^{150} + p bombardments. 1.48 Mev gamma ray (A), 0.34 Mev gamma ray (B), K-rays (C)

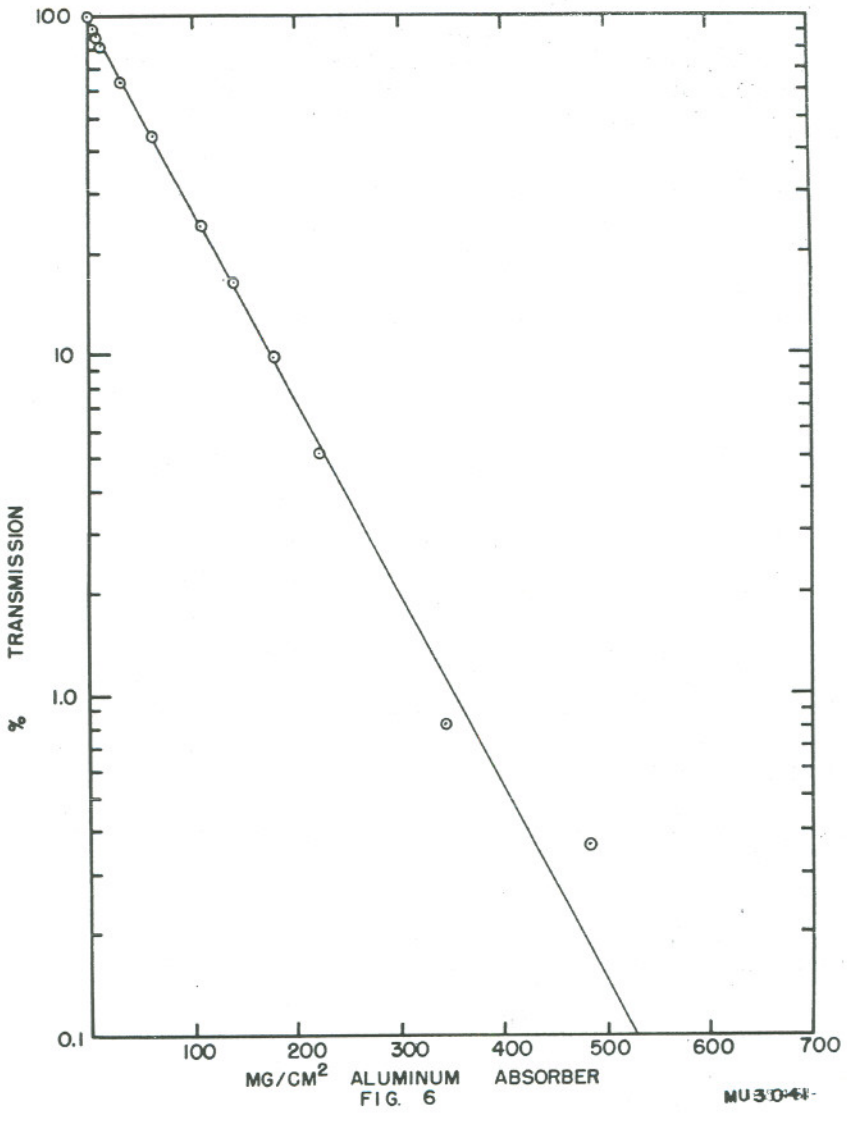


FIG. 6

Aluminum absorption data curve of the 54 hour activity from Nd^{150} + p bombardments. 1.03 Mev negatron

was observed. The half-lives of the activity at magnet currents corresponding to 1.5 Mev and 2.5 Mev were both 161 min.

The highly enriched neodymium sample made it possible to obtain sufficient activity to examine it on the high resolution beta spectrometer. The data shows clearly (Fig. 7) that a negatron component of energy greater than 2 Mev was present. Fermi-Kurie plots (Fig. 8) were made on the data from four examinations and components at 3.00 ± 0.01 Mev and 2.01 ± 0.03 Mev were found. The crude spectrometer decay measurement indicating that both these particles belong to a 161 min decay, was confirmed. The average of the ratios of the amounts of these components present in the four experiments is the following:

2 Mev negatron		3 Mev negatron
1.00	:	0.16 ± 0.03

In one case alone sufficient activity was present to permit determination of the shortest component. This yielded 1.05 ± 0.10 Mev.

Ratios were calculated from the absorptions with the assumption that all the ~0.3 Mev gamma observed came from the 161 min decay.

~0.9 Mev negatron		~2.1 Mev negatron		K x-rays: ~0.3 Mev gamma		~1.4 Mev gamma
0.08	:	0.26	:	1.0	:	0.15	:	0.21

The following ratio for the 54 hr activity was estimated on the basis of the aluminum absorption and the scintillation spectrometer result.

0.9 Mev negatron		K x-rays: ~0.3 Mev gamma
1.0	:	~1.0 : <1.0

The difference of the two ratios should give that for the 161 min activity. Combining this with the beta spectrometer ratio, the following was found.

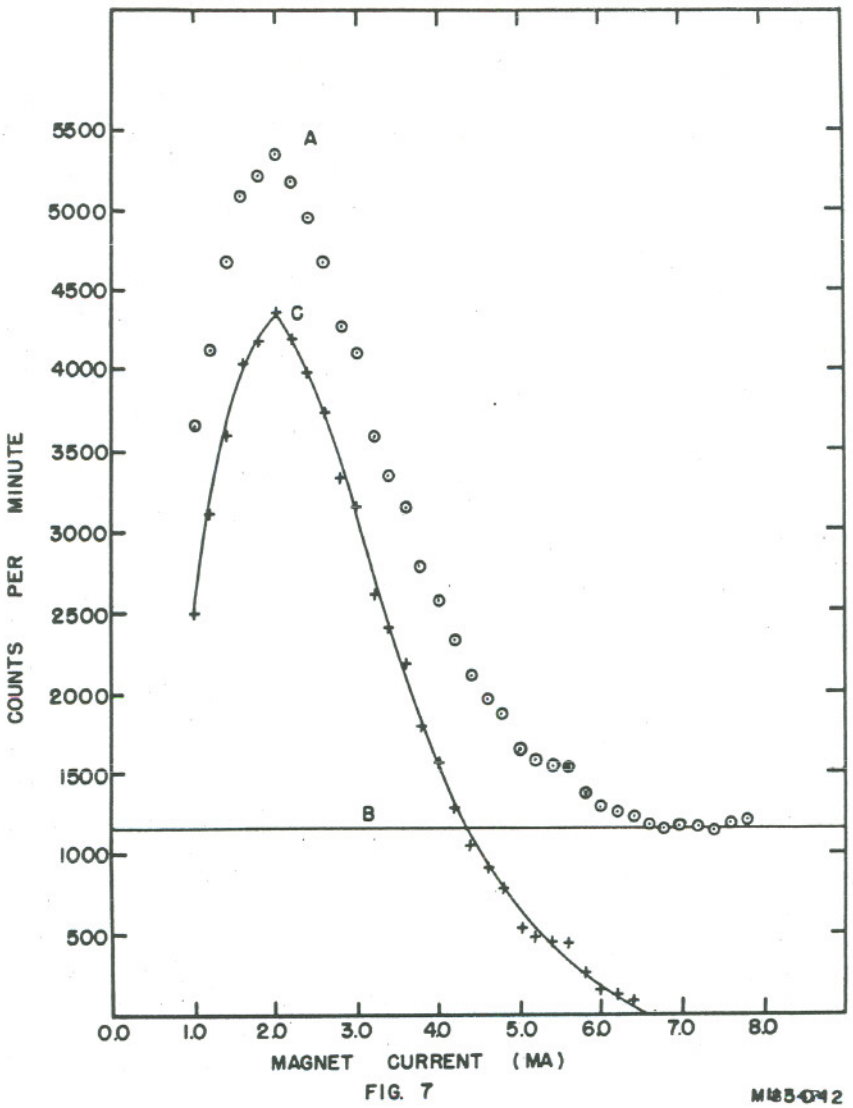
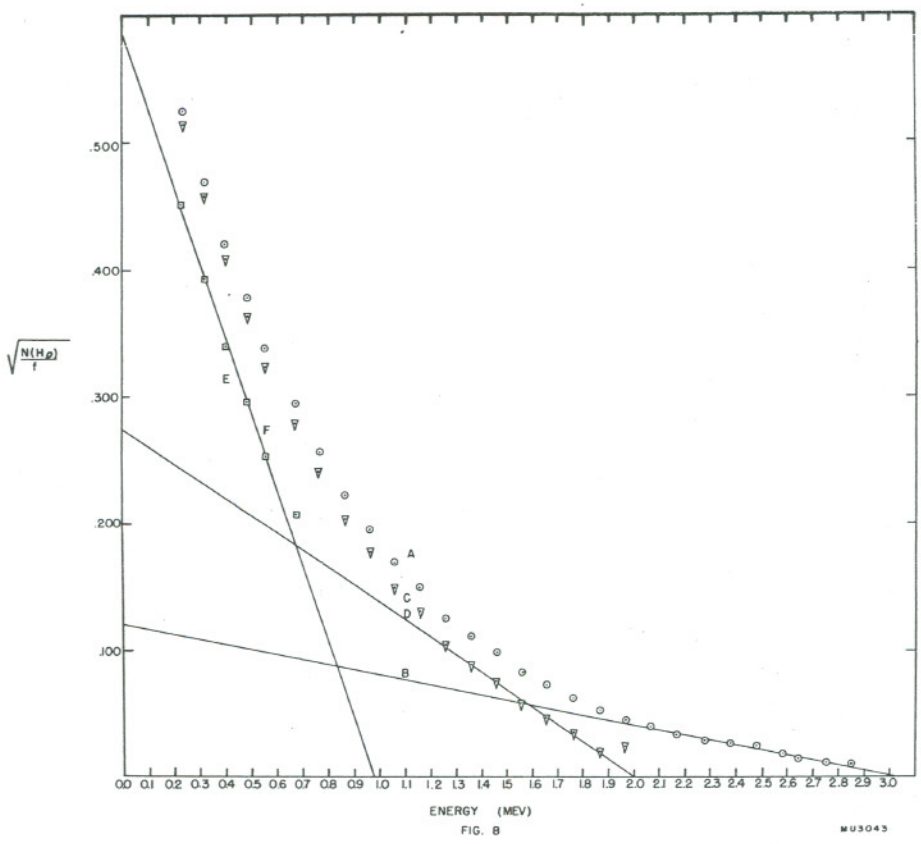


FIG. 7

Beta spectrometer data for the 161 minute and 54 hour activities from Nd^{150} + p bombardments. Readings uncorrected for decay (A), background (B), with background subtracted and corrected for 161 minute decay (C)



Fermi-Kurie plot of beta spectrometer data for the 161 minute and 54 hour activities from Nd150 + p bombardments. For data corrected for 160 minute decay (A), 3 Mev beta (B), (A) with 3 Mev beta contribution "subtracted" (C), 2 Mev beta (D), (C) with 2 Mev beta contribution "subtracted" and corrected for decay to 54 hour half-life (E), 0.97 Mev beta (F)

2 Mev negatron	3 Mev negatron	K x-rays	~0.3 Mev gamma	~1.4 Mev gamma
0.22	0.033	~1.0	~0.067	0.2

These data are consistent with a decay scheme in which the 161 min isotope emits a 3 Mev negatron followed by two 0.3 Mev gamma rays and a 2 Mev negatron followed by an ~1.4 Mev gamma ray. The 54 hr isotope emits a 1.0 Mev negatron and a low intensity 0.3 to 0.35 Mev gamma ray. K x-rays are associated with both activities.

Identification

A sample of the neodymium enriched in Nd^{150} and bombarded with protons was separated on a cation exchange column with a rapid drop rate so that all the activity was off in 8 hours. The decay of samples from the front, top, and back of the activity peak were compared with that of an unseparated portion of the sample. All four were identical (Fig. 9), containing both the 161 min and the 54 hr activities. The activity peak was found to occur immediately before the neodymium mass peak and thus identified as promethium.

The cross sections calculated for the formation of the 161 min and the 54 hr activities are 4 and 3 millibarns, respectively. These values are reasonable for an 8.9 Mev bombarding energy.

It is shown by the cross sections calculated for the formation of the isotopes with 161 min and 54 hr half-lives, from bombardments of neodymium samples enriched in other isotopes, that these activities are produced from Nd^{150} (see Table 7). Since both Sm^{149} and Sm^{150} are stable and in view of the column separation, the activities must be due to Pm^{149} or Pm^{150}.

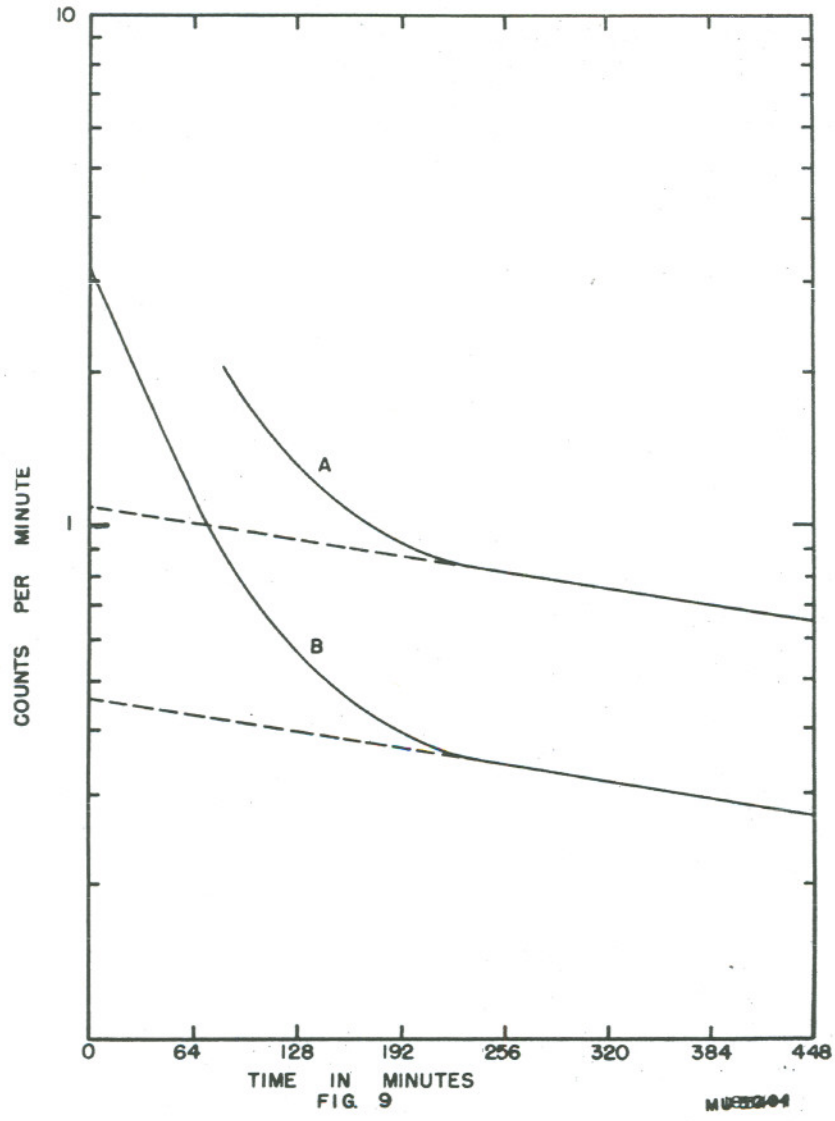


FIG. 9

Decay of the 161 minute and 54 hour activities from Nd^{150} + p bombardments. Sample from promethium peak of elution curve (A), unseparated sample (B)

Another sample enriched in Nd^{150} was bombarded with 9.8 Mev protons from the linear accelerator. The same two activities were observed and the ratio of their cross sections was calculated.

σ for 161 min	:	σ for 54 hr
0.8	:	1.0

Since the ratio at 8.9 Mev can be seen from above to be:

1.0	:	0.75

it was concluded that the 54 hr activity was Pm^{149} produced by a $Nd^{150}(p,2n)$ reaction and the 161 min activity, Pm^{150} from a $Nd^{150}(p,n)$ reaction.

Two attempts were made to ascertain these assignments mass spectrographically,[64] but due to the small proportion of promethium in the sample and to the high loss factor in this region of the periodic table, no activity could be found on the plates.

B. Proton Bombardment of Neodymium Enriched in Nd^{148}

In the present work bombardment of neodymium enriched in Nd^{148} with 60-inch cyclotron protons yielded a 5.3 day, 42 day, and a long half-lived isotope in addition to the 161 min half-lived isotope previously assigned to Pm^{150}.

Pm^{148} has been reported by previous workers to have a 5.3 day half-life[10,18,43,65] and to emit 2.5 Mev negatrons[18,43] and 0.8 Mev gamma rays.[18,43] It was found among fission products and as the product of Nd(d,2n), Nd(p,n), and $Nd(n,\gamma)Nd^{148} \xrightarrow{\beta^-}$ reactions.[10,18,43] It was identified chemically as promethium and mass spectrographically as A = 148.[43]

Also assigned by ion exchange and mass spectrographic identification,[35] is the 3.7 yr isotope Pm^{147} found among fission products[13,14,18,27,28,35-37]

and from $Nd(n,\gamma)Nd^{147} \xrightarrow{\beta^-}$ reactions.[18,28,35] Various examinations of the negatron emitted have yielded slightly different energies. The value of 0.223 Mev found independently by Emmerich and Kurbatov[41] and by Price, Motz, and Langer[42] is probably the best. The latter investigators obtained the same value from both a 40 cm radius of curvature spectrometer and a small 180° focusing Helmholtz coil spectrometer.

Folger and Stevenson[48] found an isotope with 42 ± 2 day half-life among the products of the high energy fission of uranium. It emitted ~2.3 Mev and ~0.5 Mev negatrons and ~1 Mev gamma rays in the following ratio.

~0.5 Mev negatron	:	~2.3 Mev negatron	:	~1 Mev gamma ray
0.95	:	0.05	:	1.90

It was identified as promethium by column separation and tentatively assigned to Pm^{146}.

The 5.3 day, 42 day, and the long half-lived isotopes produced in the present bombardments were characterized and assigned as follows on the basis of ratios calculated from absorption data, cross sections, and previous work.

5.3 day	Pm^{148}
42 day	Pm^{148} or Pm^{147}
long	Pm^{147} (see Table 9)

Measurements

The decay of the activities from proton bombardment of a sample enriched in Nd^{148} was resolved into 161 ± 6 min, 5.3 ± 0.1 day, and 42 ± 1 day half-life components (Fig. 10). The presence of a longer component was noted, but the sample has not decayed sufficiently yet

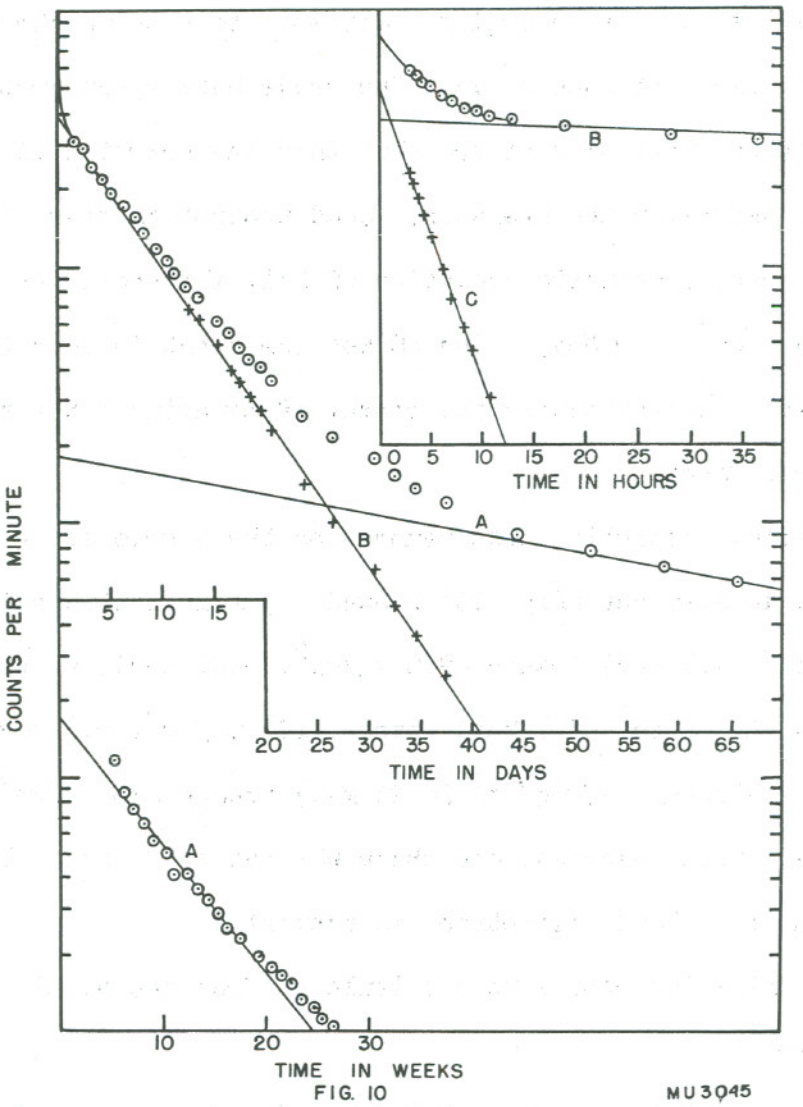


FIG. 10

Decay of activities formed by Nd^{148} + p bombard**ments.**
42 day activity (A), 5.3 day Pm^{148} (B), 2.7 hour Pm^{150} (C)

for a half-life determination.

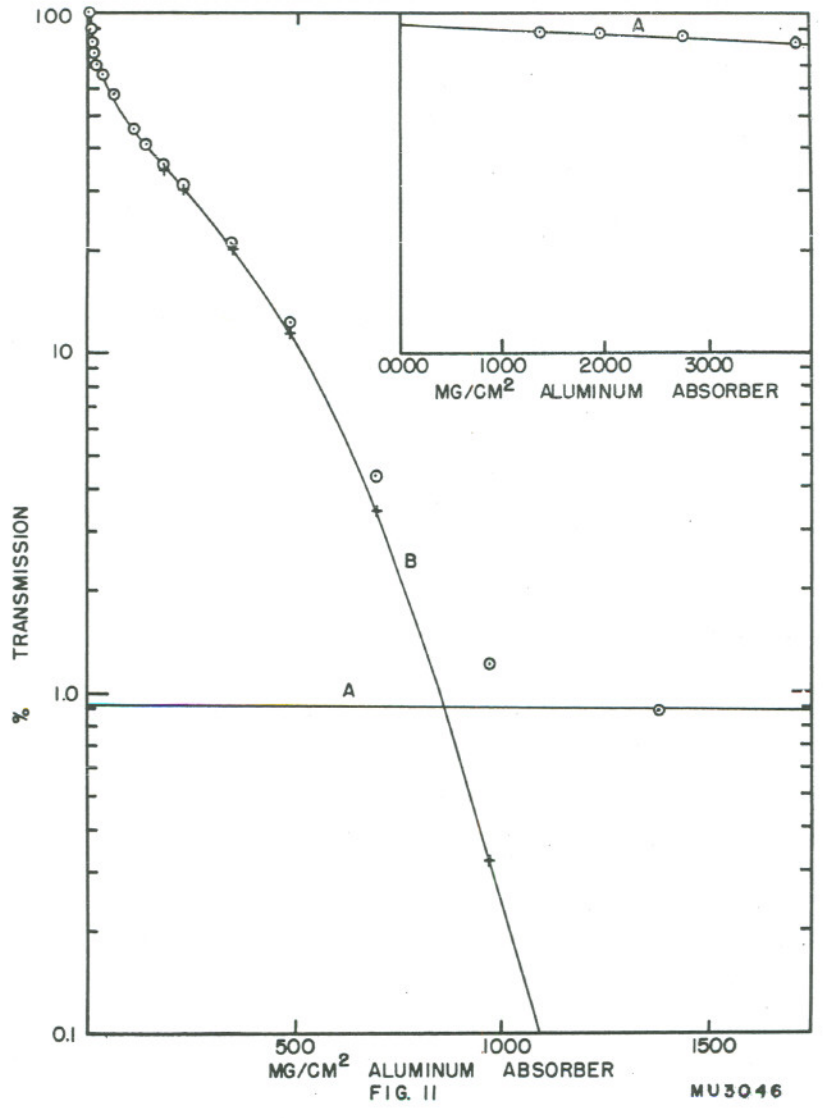
Aluminum absorption measurements after the 161 min activity had decayed (Fig. 11) showed a particle with range ~1.1 g/cm^2 (2.3 Mev), and particles at lower energies. Attempts to resolve the absorption data curve were not conclusive. The crude beta spectrometer showed that only negatrons were present and that they had energies in rough agreement with the absorption measurements. Lead absorption measurements (Fig. 12) indicated electromagnetic radiation of half-thickness ~48 mg/cm^2 (40 kev) and ~8.55 g/cm^2 (1.0 Mev). The 40 kev component is doubtful, but if present is at approximately the energy of samarium K x-rays. No L x-rays were found.

Aluminum absorption measurements on the sample after the 5.3 day Pm^{148} had decayed out (Fig. 13) showed a particle with half-thickness ~185 mg/cm^2 (2.7 Mev) (range ~970 mg/cm^2, 2.04 Mev), another with half-thickness ~25 mg/cm^2 (0.7 Mev) (range ~195 mg/cm^2, 0.6 Mev), and a third with half-thickness ~5 mg/cm^2 (0.25 Mev) (range ~35 mg/cm^2, 0.2 Mev). No L x-rays were observed, and there was not sufficient electromagnetic radiation for a lead absorption measurement.

The following ratios were calculated from the aluminum absorption data curves.

absorption	2.3 to 2.7 Mev negatron:	~0.7 Mev negatron:	~0.2 Mev negatron:	~1 Mev Y-ray and x-ray
1	1.00 :	:	:	0.9
2	0.062 :	1.00 :	1.76 :	3.44

It was noted qualitatively that whereas the 0.2 Mev negatron decayed with a half-life much greater than 42 days, the 2.5 Mev and the 0.7 Mev



Aluminum absorption data curve of the 5.3 day and 42 day activities from Nd148 + p bombardments. Gamma ray and K x-ray background (A), 2.3 Mev negatron (B)

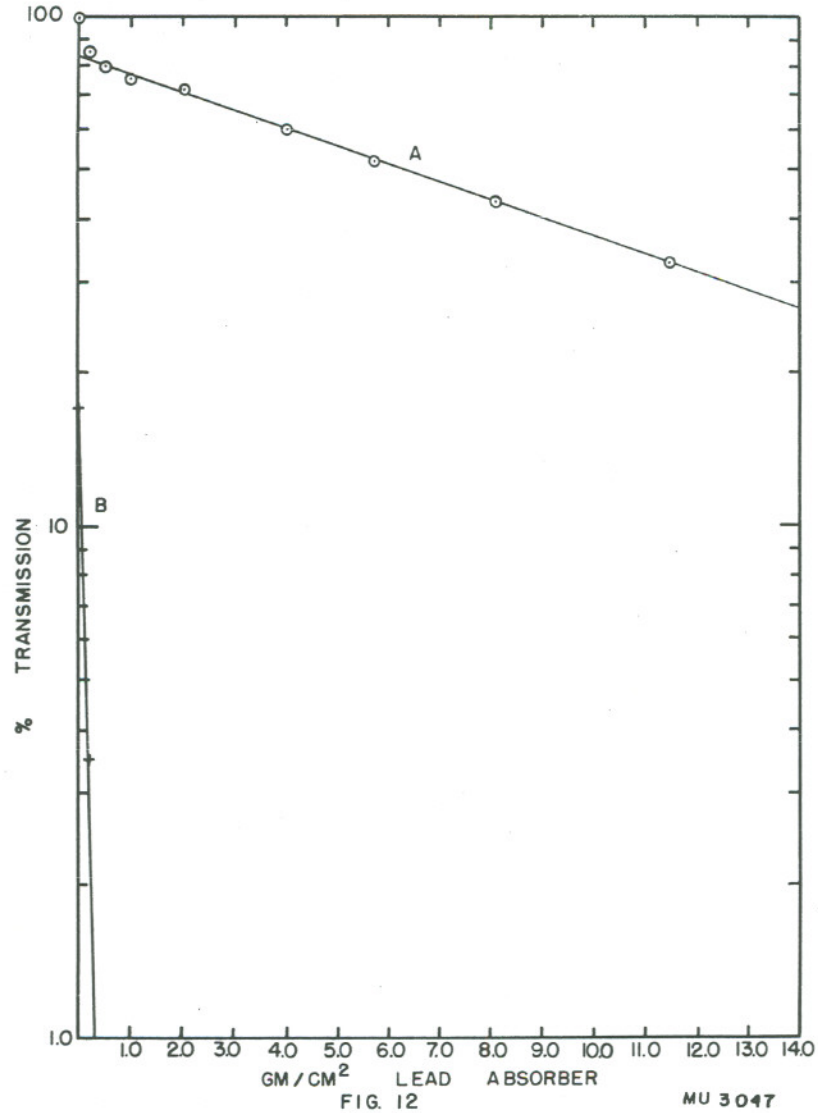
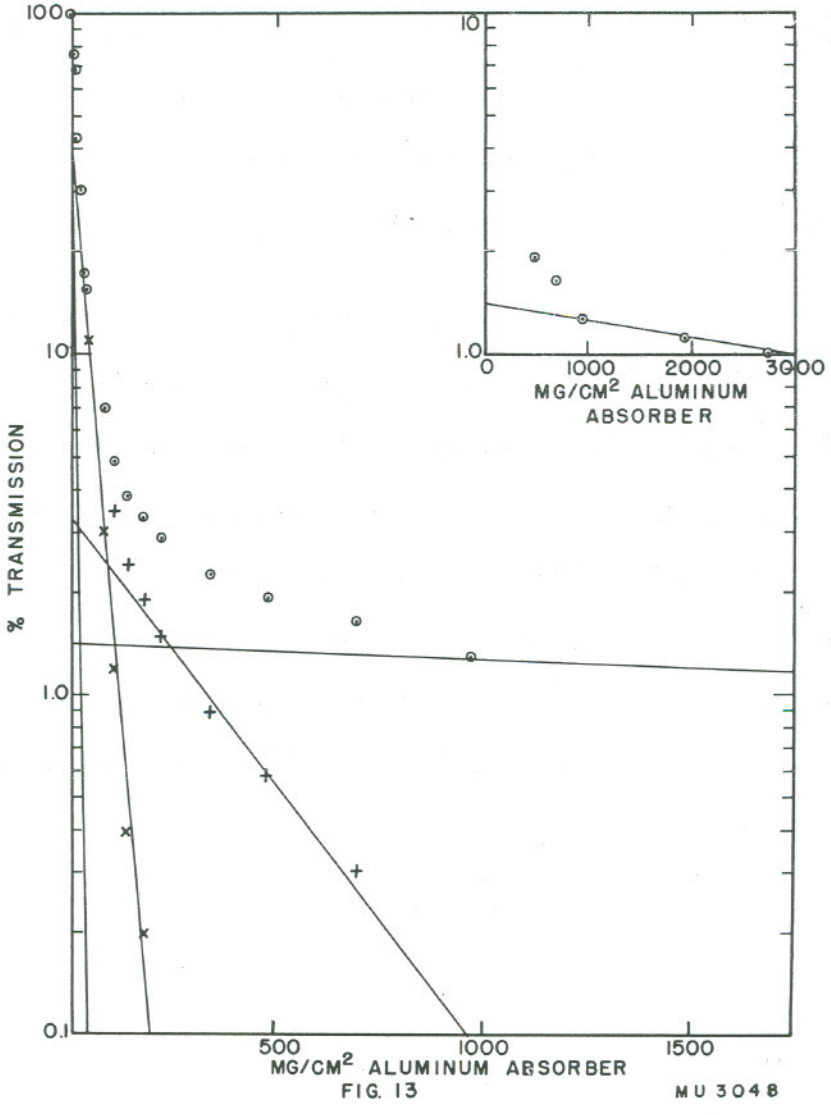


FIG. 12

Lead absorption data curve of 5.3 day and the 42 day activities from Nd^{148} + p bombardments.
1.06 Mev gamma ray (A), K x-ray (B)



Aluminum absorption data curve of the 42 day and long activities from Nd^{148} + p bombardments. K x-ray and gamma ray background (A), 2.7 Mev negatron (B), 0.7 Mev negatron (C), 0.25 Mev negatron (D)

negatrons and the gamma radiation appeared to decay at that rate.

Considering these results and previous work, the following decay schemes were postulated.

5.3 day	~2.3 Mev negatron followed by an ~1.0 Mev gamma ray
42 day	~2.7 Mev negatron and ~0.7 Mev negatron followed by two ~1.0 Mev gamma rays
long	~0.2 Mev negatron

Assignments

On the basis of the ratios calculated from the results of the absorption measurements, cross sections (Table 7) were calculated for the various activities found. It was assumed that the long-lived isotope decayed by emitting the 0.2 Mev beta particle and that it was the 3.7 yr Pm^{147} produced from the reaction $Nd^{148}(p,2n)Pm^{147}$. Dividing the cross sections of the 161 min activity by the percentage of Nd^{150} in the sample and the others by that of Nd^{148}, the following values were obtained.

161 min: 2.6 mb

5.3 day: 2.0 mb

42 day : 1.0 mb

3.7 yr : 2.0 mb

Thus the $Nd^{148}(p,n)Pm^{148}$ and the $Nd(p,2n)Pm^{147}$ reactions appear to go at approximately the same rate. The 42 day activity could belong to an isomer of either Pm^{147} or Pm^{148}. The assignment of the isotope with this half-life will be discussed further in Part IV.

C. Alpha Bombardments of Praseodymium

Wilkinson and Hicks[31,32] have bombarded praseodymium with helium ions at various energies. They first reported two activities, one about 350 days and the other 4.1 hr.[32] The long activity had been seen previously in similar bombardments[7-9] and also in deuteron bombardments of neodymium.[7-10] Hicks and Wilkinson reported a better value of 285 ± 3 days after observing the decay for the two half-lives.[31] When an ion exchange column separation was performed on the sample, the 4.1 hr activity could not be detected in the promethium peak.[66] Since the elution required about twenty hours, this did not constitute absolute proof that the 4.1 hr decay does not belong to a promethium isotope. The 285 day activity, however, was found in the promethium peak. Cross section calculations indicated that the 4 hr and the 285 day activities come from $Pr^{141}(\alpha,n)Pm^{144}$ and $Pr^{141}(\alpha,2n)Pm^{143}$ reactions, respectively. An activity corresponding to $Pr^{141}(\alpha,3n)Pm^{142}$ was not observed and an upper limit of 5 min was placed on the half-life of Pm^{142}.[31] The 285 day activity decayed by electron capture, emitting L and K x-rays of neodymium, an ~0.95 Mev gamma ray, and apparently a 0.7 Mev negatron.[32] The 4 hr activity emitted a 1.3 Mev positron and electromagnetic radiation.[32] The following absorption ratios were reported.[32]

	~0.7 Mev negatron	:	L x-rays	:	K x-rays	:	~0.95 Mev γ-ray
285 day isotope	0.005	:	0.2	:	1.0	:	0.3

	~1.3 Mev positron	:	γ-ray plus x-rays
4 hr isotope	1	:	1

The present study was undertaken to verify the chemical identity of the 4 hr activity and further characterize its decay. It was proved

that the activity did not belong to an isotope of promethium. Further bombardments were performed to verify the previous results for the 285 day activity and to discover the decay characteristics of Pm^{142} and Pm^{144}.

Measurements

All the alpha bombardments of praseodymium at the various energies used (Table 6) gave a long-lived activity. It was assumed to be the 285 day activity previously observed. A 4.0 ± 0.1 hr activity was observed from all bombardments except those at 35.5 Mev (see Fig. 14 for the cross sections found for the production of the isotopes with these half-lives). A 2.4 ± 0.2 hr decay was observed with 0.0008 barn cross section at 35.5 Mev and with 0.001 barn cross section at 32.7 Mev. The only other activity produced decayed with a 20 ± 5 hr half-life and was formed with 0.0002 barn cross section solely at 35.5 Mev.

Aluminum absorption measurements (Fig. 15) on the 4 hr activity indicated a particle with half-thickness 64 mg/cm^2 (1.3 Mev) (range 630 mg/cm^2, 1.4 Mev). Measurement with the crude beta spectrometer showed this to be a positron and agreed with the energy found by absorption. Lead absorption measurements (Fig. 16) indicated a single type of electromagnetic radiation with half-thickness ~5.92 g/cm^2 (0.76 Mev). The following ratio was calculated from the results of the absorption measurements.

~1.2 Mev positron: ~0.8 Mev γ-ray

1 : 1

Aluminum, lead, and copper absorption measurements were performed on the long half-life activity formed at various bombardment energies.

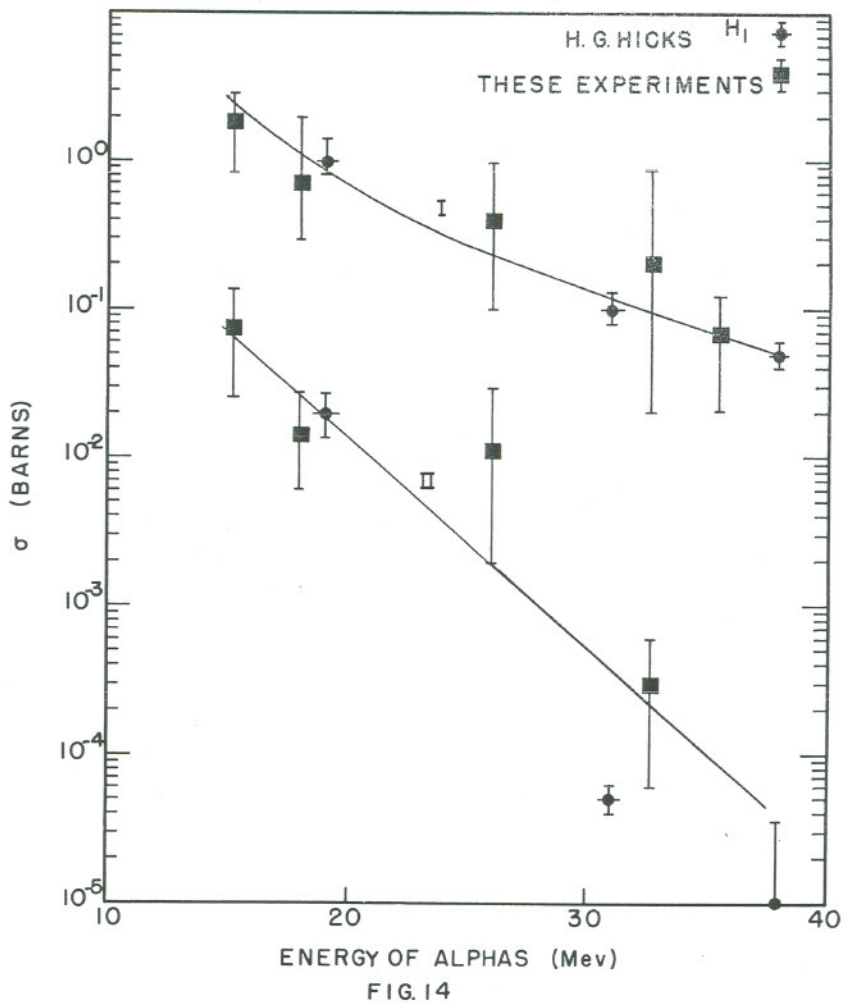


FIG. 14

MU 3049

Cross sections of activities produced in Pr + α bombardments. Long half-life (I), 4 hour (II)

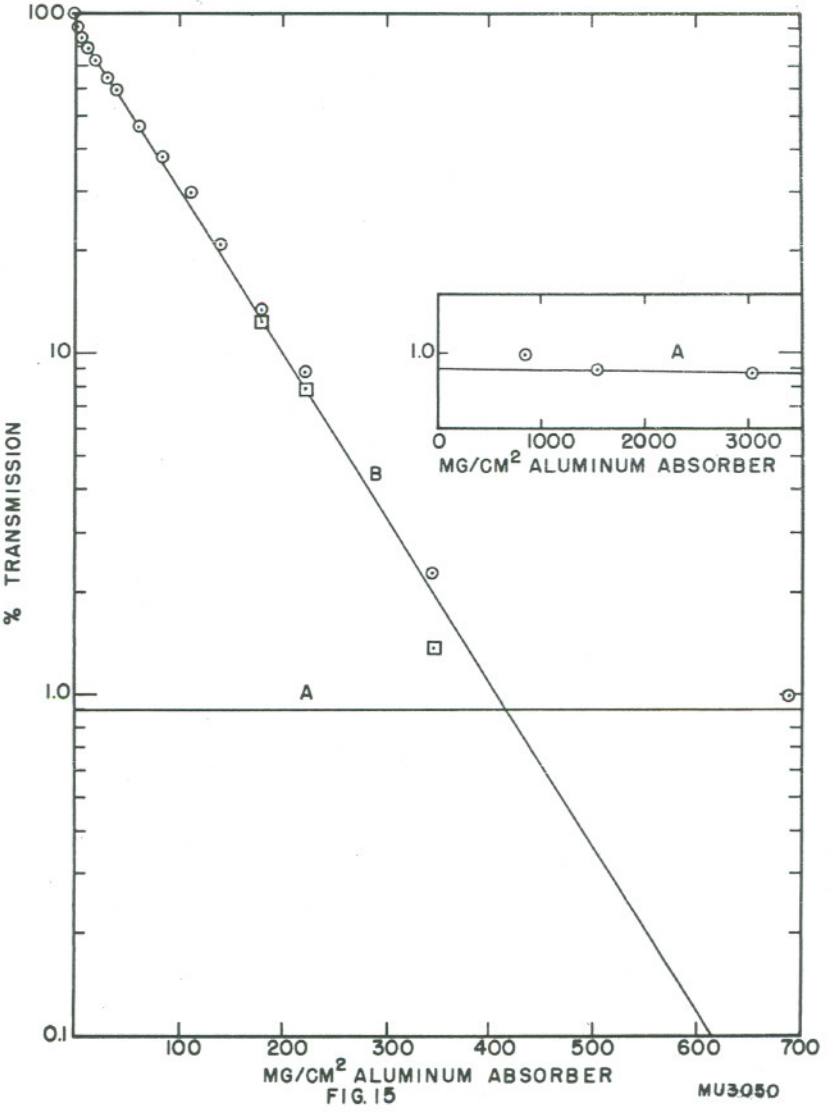


FIG. 15

Aluminum absorption data curve of the 4 hour activity from Pr + α bombardments. Gamma ray background (A), 1.2 Mev positron (B)

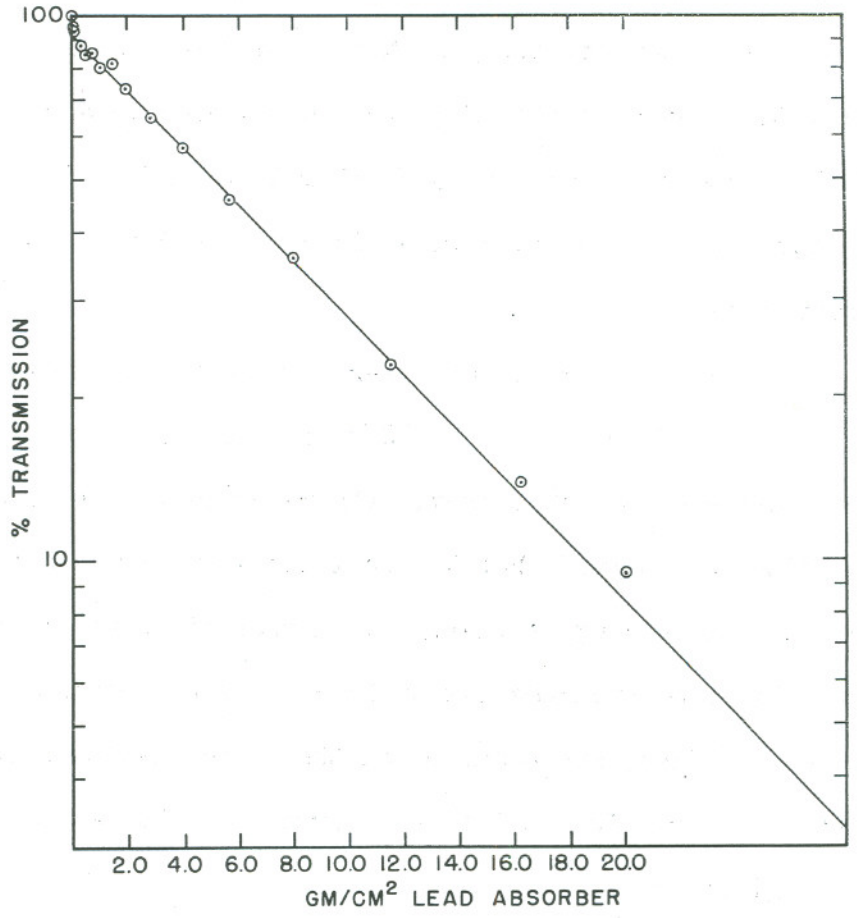


FIG. 16

MU3051

Lead absorption data curve of the 4 hour activity from Pr + α bombardments. 0.76 Mev gamma ray

In each case the result was approximately as follows. Aluminum absorption measurements (Fig. 17) showed a particle with range ~220 mg/cm^2 (0.65 Mev). Copper absorption measurements (Fig. 18) showed electromagnetic radiation with half-thickness ~5.0 mg/cm^2 (6 kev) and ~105 mg/cm^2 (37.5 kev) corresponding to the L and K x-rays of neodymium. Lead absorption measurements (Fig. 19) showed electromagnetic radiation with half-thickness ~40 mg/cm^2 (38 kev) and ~7.4 g/cm^2 (0.93 Mev). The following ratios were calculated from the results of these absorption measurement data.

0.6 Mev beta particle:	L x-ray:	K x-ray:	0.9 Mev γ-ray
0.006	0.8	1.0	0.2

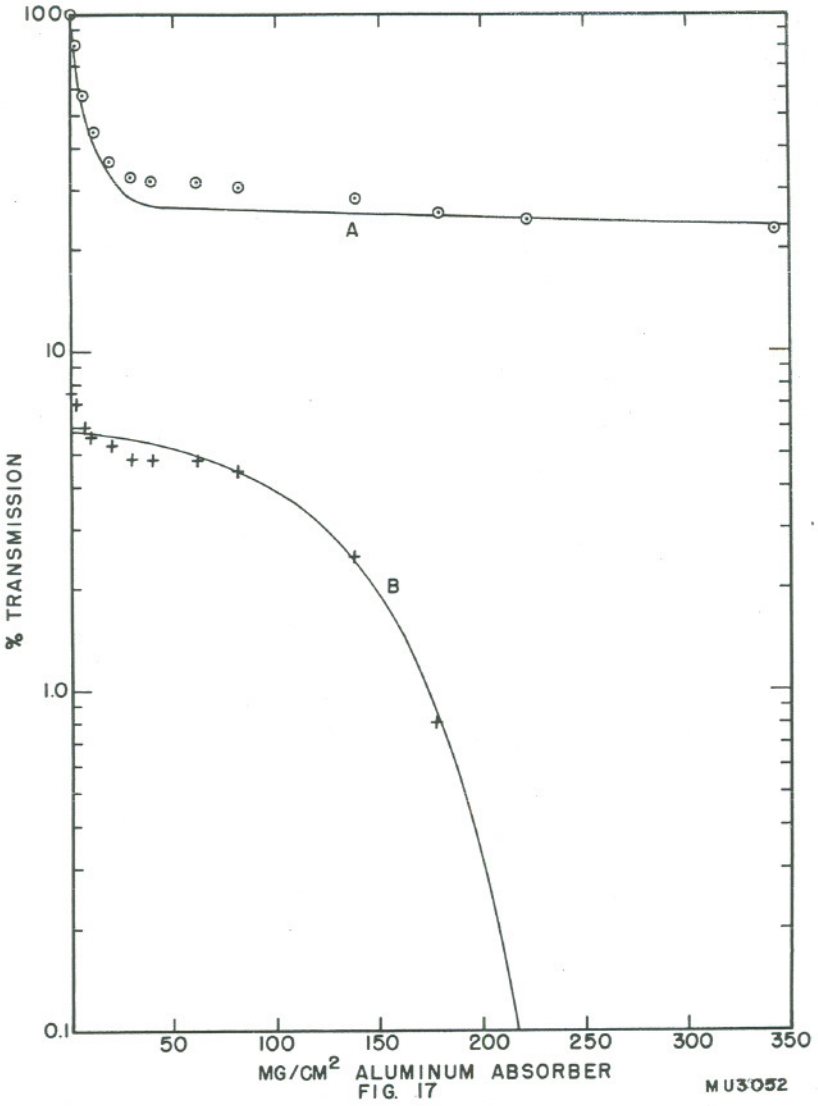
This ratio differs from that previously reported in the greater amount of L x-radiation observed, but the activity again is seen to consist of electron capture leading partially to an excited state of the daughter.

A sample from bombardment with 15.5 Mev alpha particles which had been column purified, was examined on the pulse discriminator type gamma spectrometer. The radiation observed and the ratio of the components is given below.

Nd K x-rays: 0.16 $\pm$ 0.02 Mev	γ-ray: 0.45 $\pm$ 0.02 Mev	γ-ray: 0.62 $\pm$ 0.02 Mev	γ-ray
1.0	0.6	0.2	0.3

This illustrates the uncertainty of absorption measurements, but does not change the picture as to the type of the decay being observed.

Ten minute bombardments were performed at 26 Mev and 18 Mev and the samples examined for short half-life decay. No chemical separations were done. In both cases a 2 to 3 min activity in very low abundance and a 2 to 4 hr activity in moderate abundance were observed.



Aluminum absorption data curve of the long activity from Pr + α bombardments. L x-ray plus K x-ray and gamma ray background (A), 0.78 Mev beta particle (?) (B)

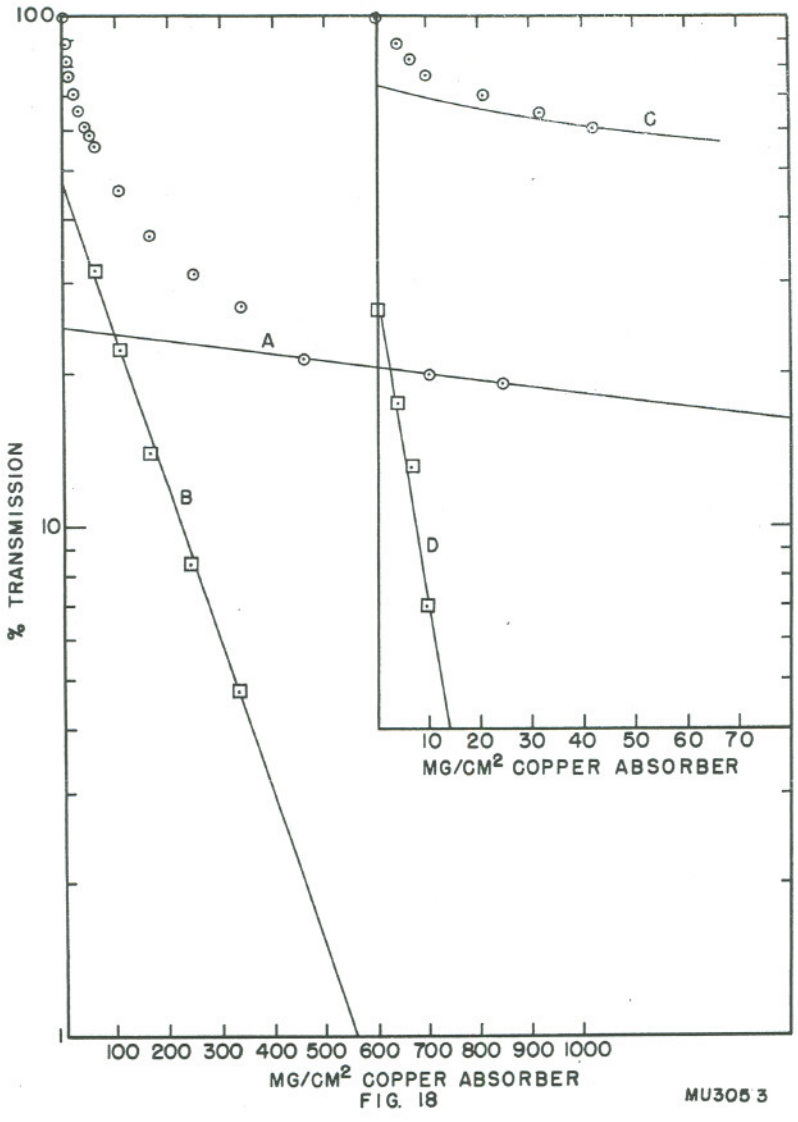


FIG. 18

Copper absorption data curve of the long activity from Fr + α bombardments. Gamma ray background (A), K x-ray (B), K x-ray + gamma ray (C), and L x-ray (D)

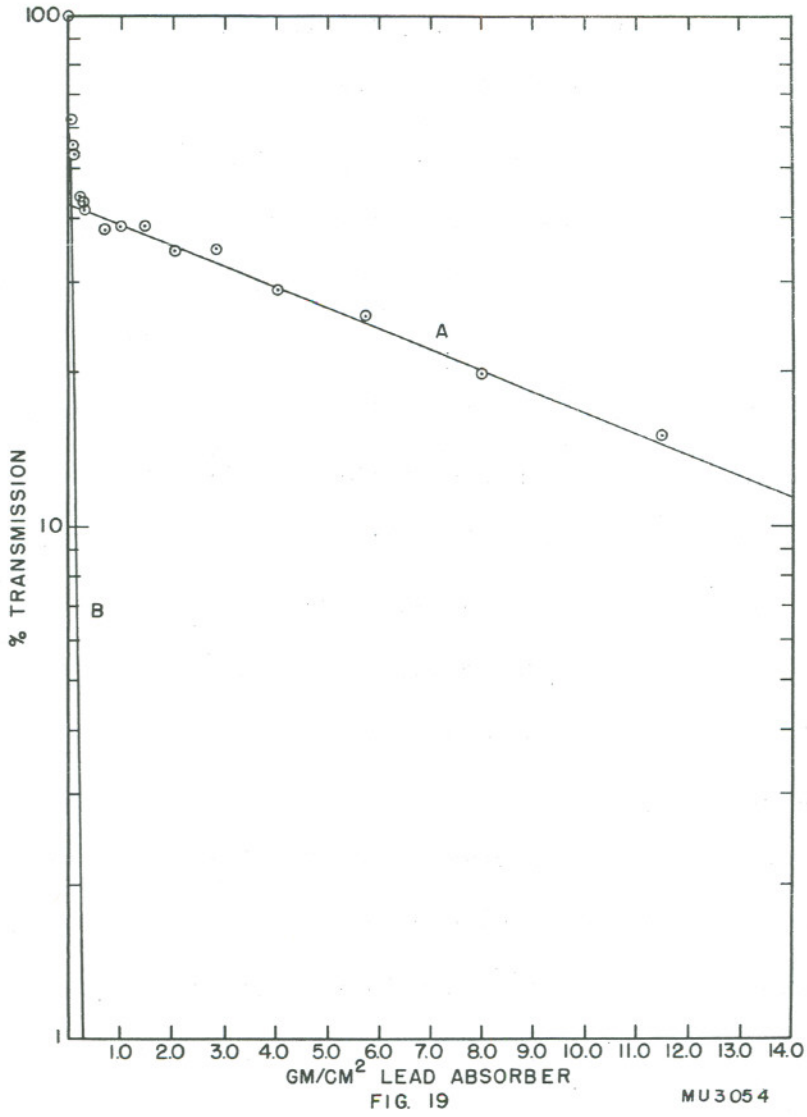


FIG. 19

Lead absorption data curve of the long activity from Pr + α bombardments. 0.93 Mev gamma ray (A), K x-rays (B)

Assignments

A sample prepared by bombarding praseodymium with 15.5 Mev alpha particles was eluted from an ion exchange resin column within 12 hours. Only one activity peak was observed after the initial "breakthrough" activity and this occurred immediately before the neodymium mass peak. It was found to decay with a very long half-life alone, while an unseparated sample still showed the 4 hr half-life (Fig. 20). No further activity was observed on the column. Thus the 4 hr activity does not belong to promethium, although the variation of the cross section for its energy (Fig. 14) would seem to indicate a $Pr(\alpha,n)Pm^{144}$ reaction. The column run indicates further that the 4 hr isotope is not a rare earth in the vicinity of promethium, unless it is a samarium isotope. This may be the case since it has been observed in preliminary studies that frequently samarium is not eluted in the proper sequence. Its adsorption on the column in a +2 oxidation state would account for this phenomenon.

The cross section versus energy plot for the long-lived activity (Fig. 14) is consistent with a $Pr^{141}(\alpha,2n)Pm^{143}$ reaction and thus the assignment to Pm^{143} was accepted. Hicks[32] postulated that the 0.7 Mev electron may arise from a long-lived isomer of Pm^{143} which decays by both electron capture and beta emission. Since it is observed in absorption on samples bombarded at different energies, this did not at first seem probable. This will be discussed more fully in the next section.

The 2.4 hr or the 20 hr half-lived isotopes could be formed by a $Pr^{141}(\alpha,3n)Pm^{142}$ reaction, but both these half-lives are not of the magnitude which would be expected for this isotope. This point will

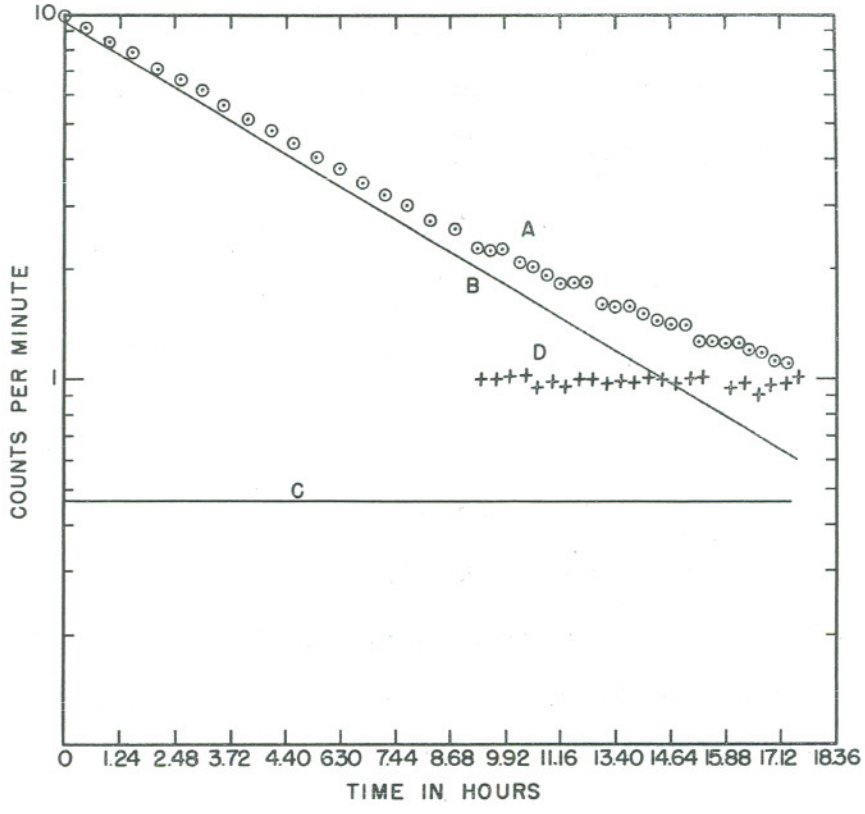


FIG. 20

MU3055

Decay of 4 hour and long half-life activities produced by Pr + α bombardments. Decay of sample before column separation (A), 4 hour component (B), long component (C), decay of promethium peak of elution curve (D)

be discussed later. It is more probable that they are the 2.42 hr Nd^{141} which emits a 0.78 Mev positron[4] and the 19.3 hr Pr^{142} which emits a 2.14 Mev negatron and a 1.9 Mev gamma.[4] These would come from $Pr^{141}(a,4n)Pm^{141} \xrightarrow{\beta^+}$ and $Pr^{141}(a,dp)Pr^{142}$ reactions, respectively.

The 2 to 3 min activity found in short bombardments could be Pm^{142}. If it decays by electron capture rather than positron emission, the counting rate observed for this half-life would be reasonable. However, as will be discussed later, electron capture is not likely for this isotope. Thus the 2 to 3 min activity is probably the 2.1 min positron emitter from $C^{12}(a,n)O^{15}$.[4] The accompanying longer activity is probably a mixture of several decays.

D. Proton Bombardments of Neodymium Enriched in Nd^{146}, Nd^{145}, Nd^{144}, Nd^{143}, and Nd^{142}

In the present series of experiments samples of neodymium enriched separately in Nd^{146}, Nd^{145}, Nd^{144}, Nd^{143}, or Nd^{142} were bombarded with protons from the 60-inch cyclotron. In addition to some of the activities previously assigned to heavier promethium isotopes only a nuclide or mixture of nuclides decaying with a half-life between 200 days and 5 years was observed.

The assignment of a promethium isotope decaying by electron capture with half-life 285 days to A = 143 has already been discussed. In the region from A = 142 to 146 the only other previous assignment which must be considered was made by Butement.[34] He bombarded a sample of very pure Sm_2O_3 with slow neutrons, and, after allowing the short lived activities to die out, he cleaned the sample of all rare earths other than samarium by several ion exchange column cycles. A 410 day activity

emitting some very low energy beta particles and the K and L x-rays of promethium remained in the samarium peak. The daughter of this isotope decayed with the emission of some low energy beta particles and the K and L x-rays of neodymium. The beta particles found in the two activities were assumed to belong to Sm^{151} and Pm^{147}, respectively. The 410 day activity and its daughter were then assigned to the decay, by the orbital electron capture, of Sm^{145} and Pm^{145}. The daughter half-life was calculated to be ~30 years from its specific activity.

An unidentified 12.5 hr beta particle emitter observed in deuteron bombardment of neodymium,[6] may also be a promethium isotope in the region from A = 142 to A = 146.

Rasmussen[67] has established the existence of alpha emitters with observable half-life among the rare earth elements. Since proximity to a closed nucleon shell is a requirement for alpha instability at low mass numbers, it was thought that some of the promethium isotopes might be alpha emitters with half-lives observable to the techniques of these experiments. The isotopes that are most probable to decay in this manner are ${}_{81}Pm_{85}^{146}$ and ${}_{61}Pm_{84}^{145}$ although even these cases would be expected to have half-lives much too long for observation.

The bombardments performed in the present work were made in order to verify the previous assignment of Pm^{143} and to discover the activities belonging to the other mass numbers. The length of bombardment was not sufficient for the 30 yr activity of Pm^{145} to be formed in large enough amounts to be observed, but indirect confirmation in the form of absence of another predominant activity was obtained. Wherever there was a sufficiently high counting rate to permit it, decay data

was supplemented by absorption measurements and observations on the crude beta spectrometer. Pm^{146} was found to be a long half-life negatron emitter from these measurements and cross section consideration, and Pm^{143} and Pm^{144} were both shown to be decaying by orbital electron capture with half-lives between 200 and 400 days. No alpha counts were observed (Table 9).

Measurements

The decay curves (Fig. 21 through 25) of the five samples previously described did not lend themselves to obvious resolution, especially since in no case was the longest component observed for a sufficiently extended period to make its half-life definite. Each curve was found to be resolvable into several sets of half-lives and the set which gave values corresponding most closely to the known 161 min, 54 hr, and 5.3 day decays was chosen. It cannot be emphasized too strongly that the following results are subject to change on a better knowledge of the long half-lives.

A summary of the half-lives obtained in all Nd + p bombardments is given in Table 7. The cross sections given in this table are not divided by the fraction of the parent isotope in the original sample. Those given for the short and medium-short decays assume the ratios found for the 161 min Pm^{150} and the 54 hr Pm^{149} (page 41), whereas those for medium-long half-lives assume the ratios for the 5.3 day Pm^{148} and the 42 day $Pm^{147-148}$ (page 49). The cross sections in the long half-life column were calculated as follows. σ_{EC} was calculated as if all the long activity arose from orbital electron capture with the ratios observed for Pm^{143} (page 59), while σ_{β} assumed that the activity consisted of ~0.8 Mev beta particles alone.

Table 7

Half-Lives and Cross Sections (Barns) Uncorrected for Sample Composition from Nd + p Bombardments

Sample Enriched in	Short		Medium short		Medium long		Long		
	~$T_{1/2}$	~σ	~$T_{1/2}$	~σ	~$T_{1/2}$	~σ	~$T_{1/2}$	~σ	~σ
142	161 min	2×10^{-6}	42 hr	2×10^{-6}	5.3 d	7×10^{-6}	266 d	5×10^{-3}	4×10^{-5}
143	"	1×10^{-5}	15 hr	1×10^{-6}	6 d	6×10^{-6}	288 d	1×10^{-2}	1×10^{-4}
144	"	8×10^{-6}	13 hr	4×10^{-6}	5.3 d	3×10^{-6}	288 d	9×10^{-3}	7×10^{-5}
145	"	1×10^{-5}	21 hr	8×10^{-6}	6 d	4×10^{-5}	285 d and longer	2×10^{-2}	1×10^{-4}
146	"	5×10^{-6}	60 hr	1×10^{-5}	5.3 d	4×10^{-5}	"	4×10^{-2}	4×10^{-4}
148	"	1×10^{-4}			5.3 d 42 d	4×10^{-3} 2×10^{-3}	3.7 yr		4×10^{-3}
150	"	4×10^{-3}	54 hr	3×10^{-3}					

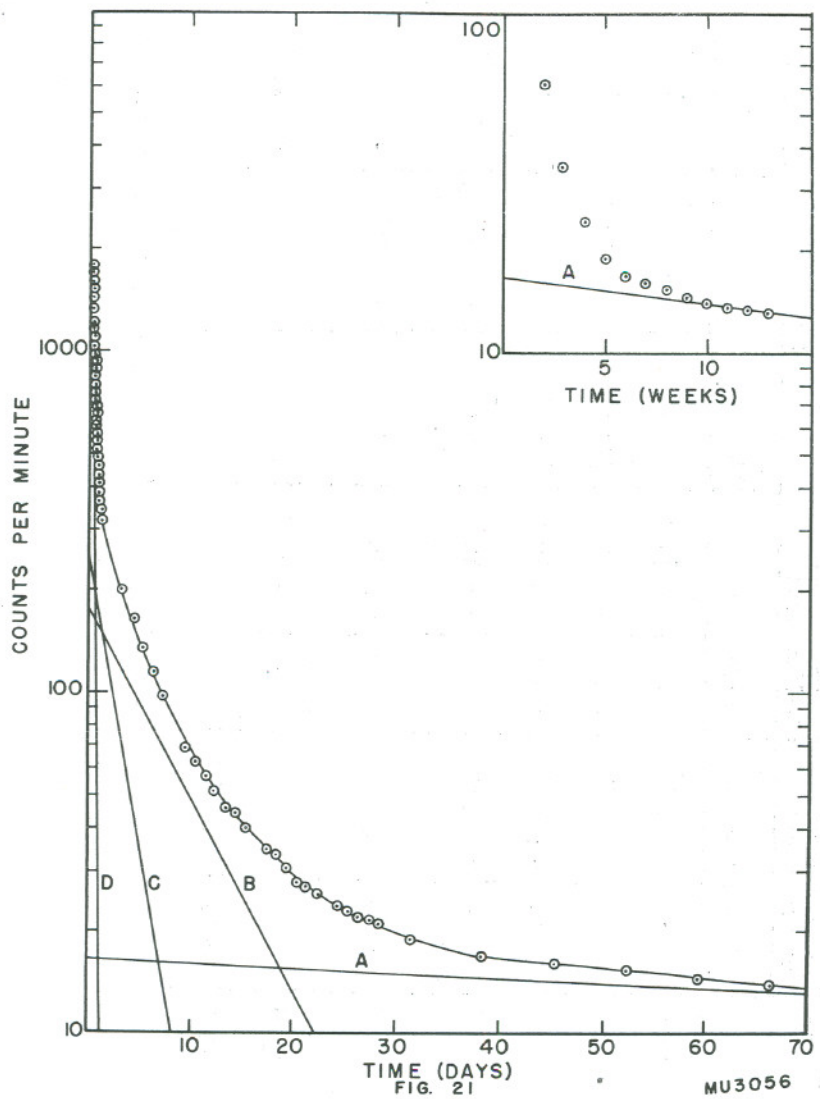


FIG. 21

Decay of activities produced by Nd^{142} + p bombardments. (A) ~266 day half-life, (B) ~5.3 day half-life, (C) ~42 hour half-life, (D) ~2.7 hour half-life

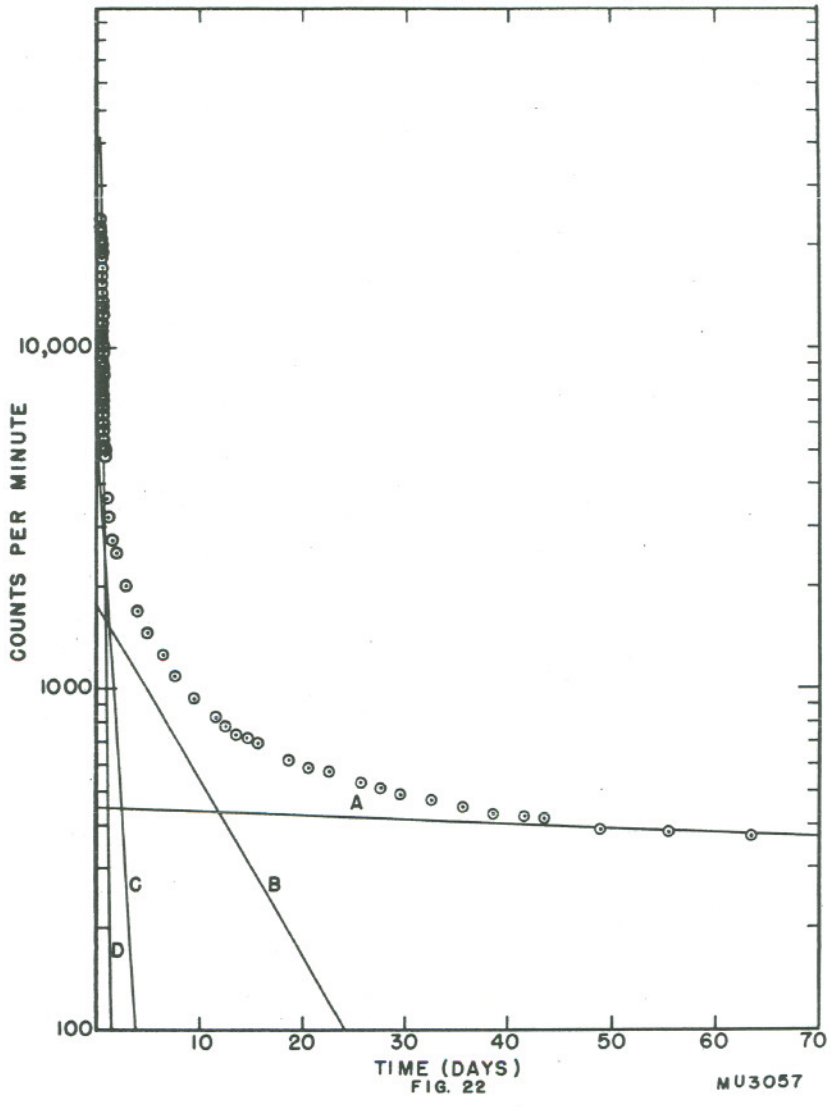


FIG. 22

ıy of activities produced by Nd^{143} + p bombardments.
280 days (A), 6 days (B), 15 hours (C), 2.7 hours (D)

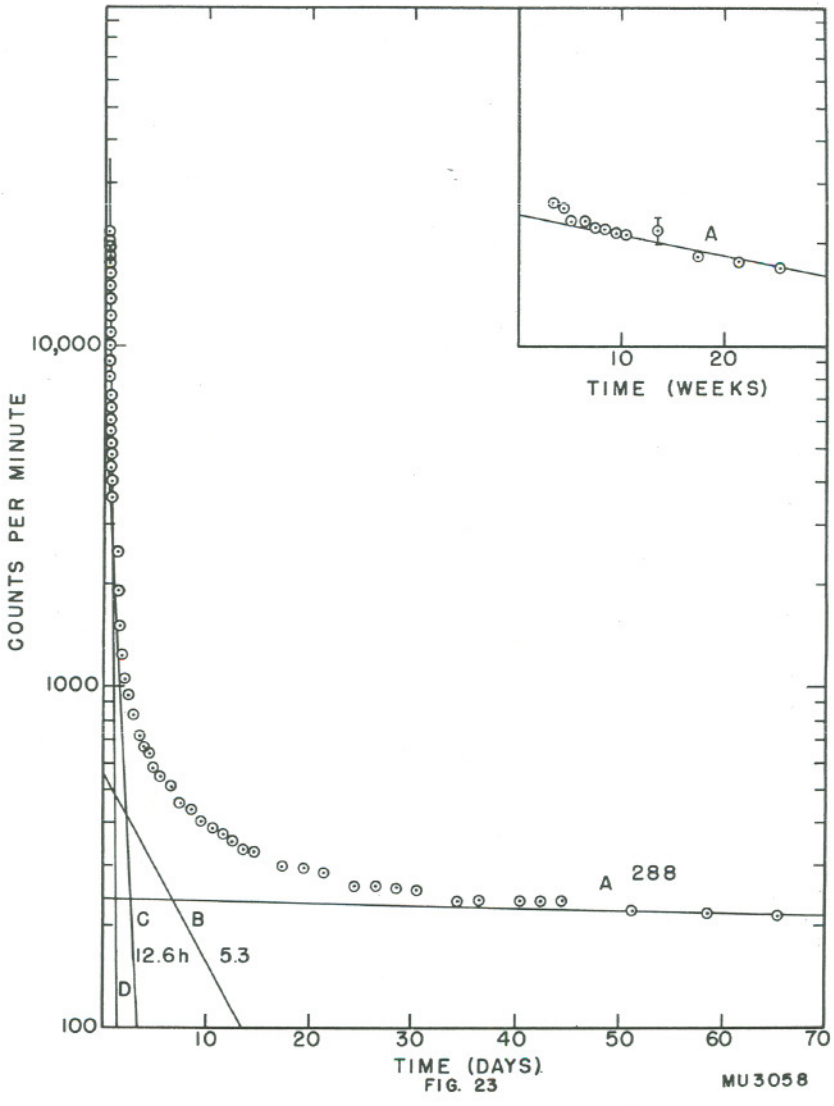


FIG. 23

Decay of activities formed by Nd^{144} + p bombardments.
288 days (A), 5.3 days (B), 12.6 hours (C), 2.7 hours (D)

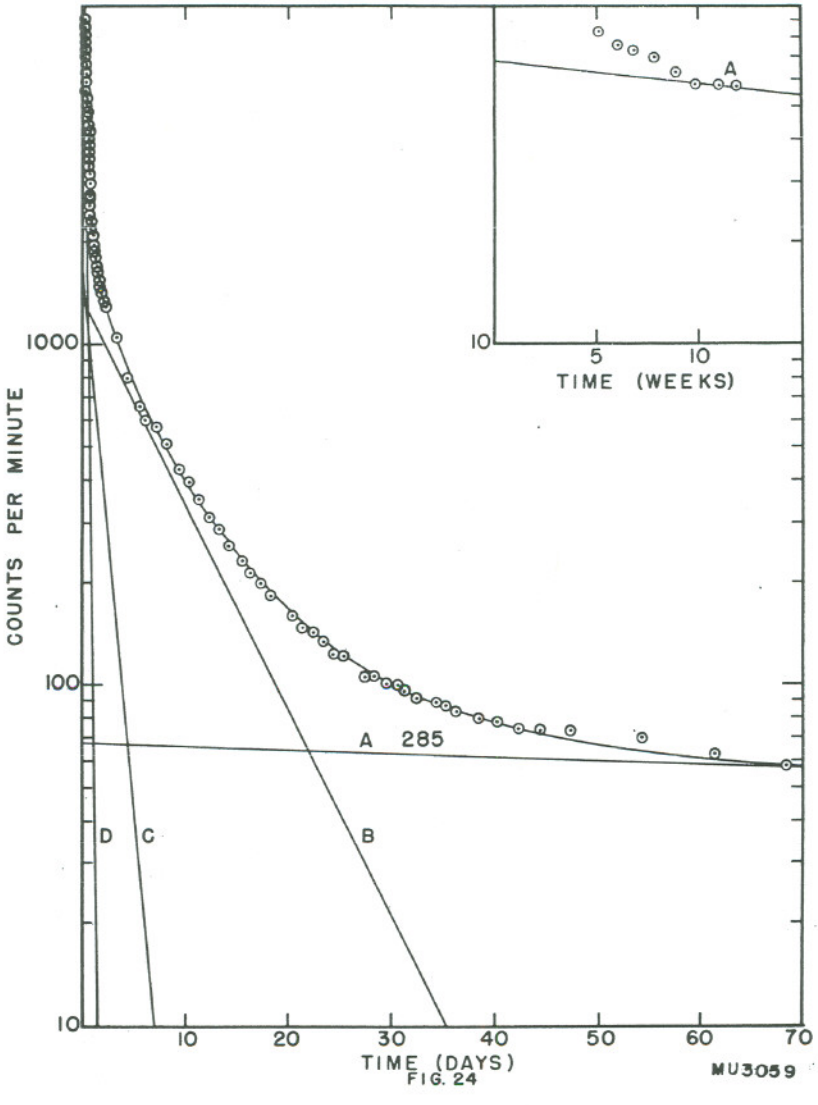


FIG. 24

Decay of activities from Nd^{145} + p bombardments.
285 days (A), 6 days (B), 21 hours (C), 2.7 hours (D)

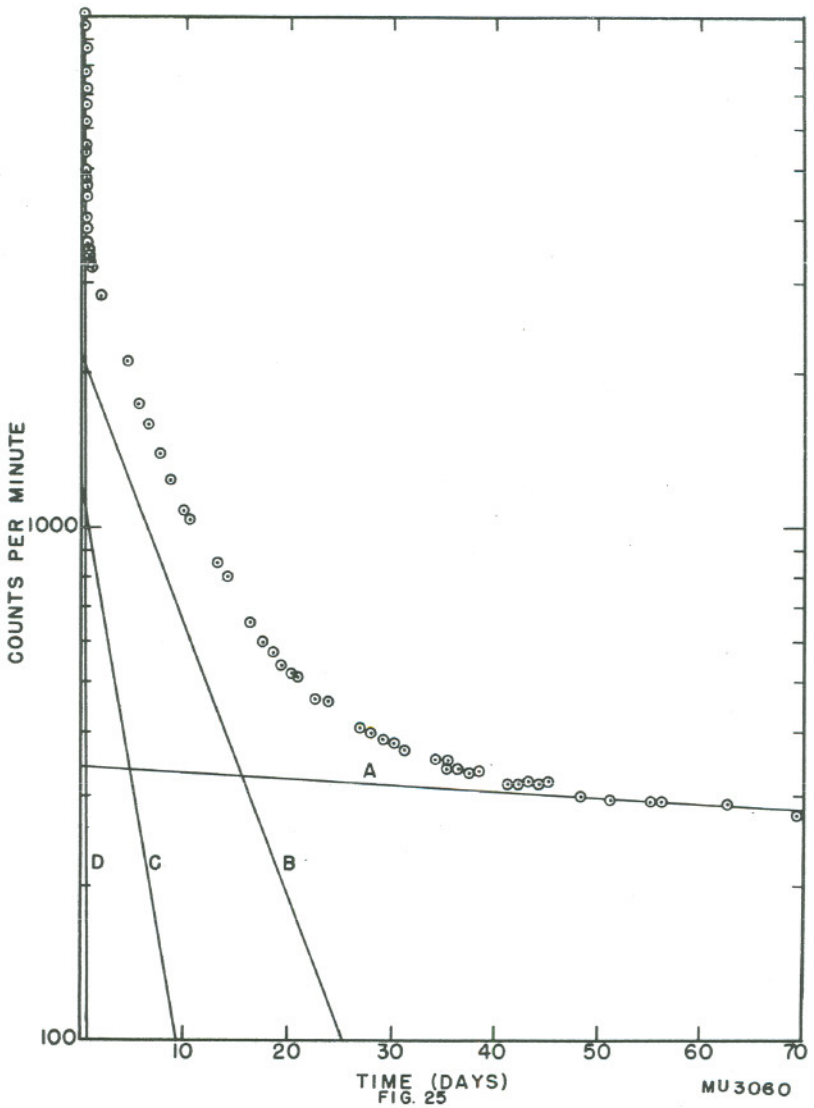


FIG. 25

Decay of activities formed by Nd^{146} + p bombardments.
285 days (A), 5.3 days (B), 60 hours (C), 2.7 hours (D)

Aluminum absorption measurements on the activities of intermediate and long half-lives from Nd^{146} + p bombardments indicated a particle with half-thickness ~154 mg/cm^2 (2.35 Mev) (range ~1300 mg/cm^2, 2.36 Mev) and another with half-thickness ~20 mg/cm^2 (0.6 Mev) (range ~88 mg/cm^2, 0.3 Mev). This supports the assumption that intermediate activity is the 5.3 day Pm^{148} (Fig. 11). The crude beta ray spectrometer indicated the sample emitted negative particles only and confirmed the absorption energies. Examination on the scintillation spectrometer[61] showed a weak 0.8 to 0.9 Mev gamma and strong components at 0.5 Mev and at 0.67 Mev. The 0.8 to 0.9 Mev value agrees with the ~1 Mev found for Pm^{148} and with the 0.8 Mev previously assigned to it. The lower energy components may be annihilation radiation, Compton radiation for the 0.9 Mev gamma or lower energy gamma rays. K x-rays were also observed.

Aluminum absorption measurements on the long component of the sample prepared from Nd^{146} + p bombardments (Fig. 26) yielded a particle with half-thickness ~30 mg/cm^2 (0.76 Mev) (range 250 mg/cm^2, 0.72 Mev). L x-rays were observed but there was not sufficient activity for a lead absorption measurement or a crude beta spectrometer inspection. Assuming a 1 percent counting efficiency for the K x-ray and gamma background, the following ratios were calculated.

~0.8 Mev beta particle: L x-rays: K x-rays and gamma rays

0.2 : 0.3 : 1.0

The bombardment of neodymium enriched in Nd^{145} gave such a low level of activity that measurements other than decay were not quantitatively performed. Qualitatively, a marked resemblance to the results for Nd^{146} was observed. Aluminum absorption measurements on a sample

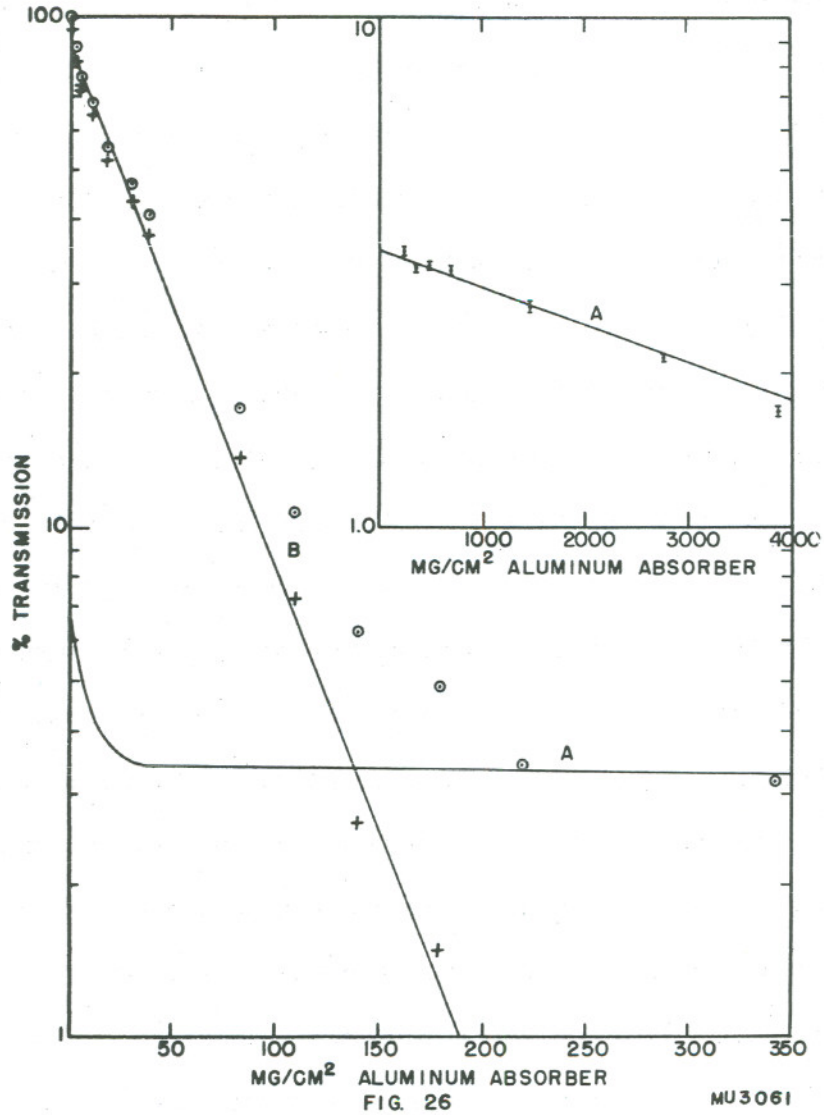


FIG. 26

Aluminum absorption data curve of the long half-life activities **from** Nd^{146} + p bombardments. Electromagnetic background (A), **0.76** Mev negatron (B)

from Nd^{145} + p bombardments immediately after bombardment with the data corrected for 161 min decay (Fig. 27) indicated a particle with half-thickness ~132 mg/cm^2 (2.0 Mev) (range ~1.00 g/cm^2, 2.1 Mev). The difference between this line and the experimental points could be interpreted to be a particle present in low yield with range ~5.5 mg/cm^2 (0.24 Mev). This, however, is dubious.

The short and medium half-lives produced from Nd^{144} + p, Nd^{143} + p, and Nd^{142} + p bombardments are similar to those of Nd^{145} + p.

Aluminum absorption measurements of the long lived activities in both Nd^{144} + p (Fig. 28) and Nd^{143} + p (Fig. 29) yielded beta particles with range ~270 mg/cm^2 (0.74 Mev). L x-rays were also observed, but there was not sufficient activity for lead absorption measurements. Neither could the crude beta spectrometer be used. Assuming a 1 percent counting efficiency for the K x-ray and gamma background, the following ratios were calculated.

Sample	~0.7 Mev beta	:	L x-rays	:	K x-rays and gamma rays
144	0.008	:	0.4	:	1.0
143	0.004	:	0.5	:	1.0

Samples from the Nd^{143} + p, Nd^{144} + p, Nd^{145} + p, and the Nd^{146} + p bombardments were examined on the pulse discriminator type gamma spectrometer with the following results.

Nd^{143} + p	Nd K x-rays:	0.17 ± 0.02 Mev gamma ray:		0.45 ± 0.02 Mev gamma ray:		0.69 ± 0.02 Mev gamma ray		
	1.0	:	0.1	:	0.08	:	0.08	
Nd^{144} + p	Nd K x-rays:	0.18 ± 0.02 Mev gamma ray:		0.43 ± 0.02 Mev gamma ray:		0.61 ± 0.02 Mev gamma ray		
	1.0	:	0.6	:	0.2	:	0.3	

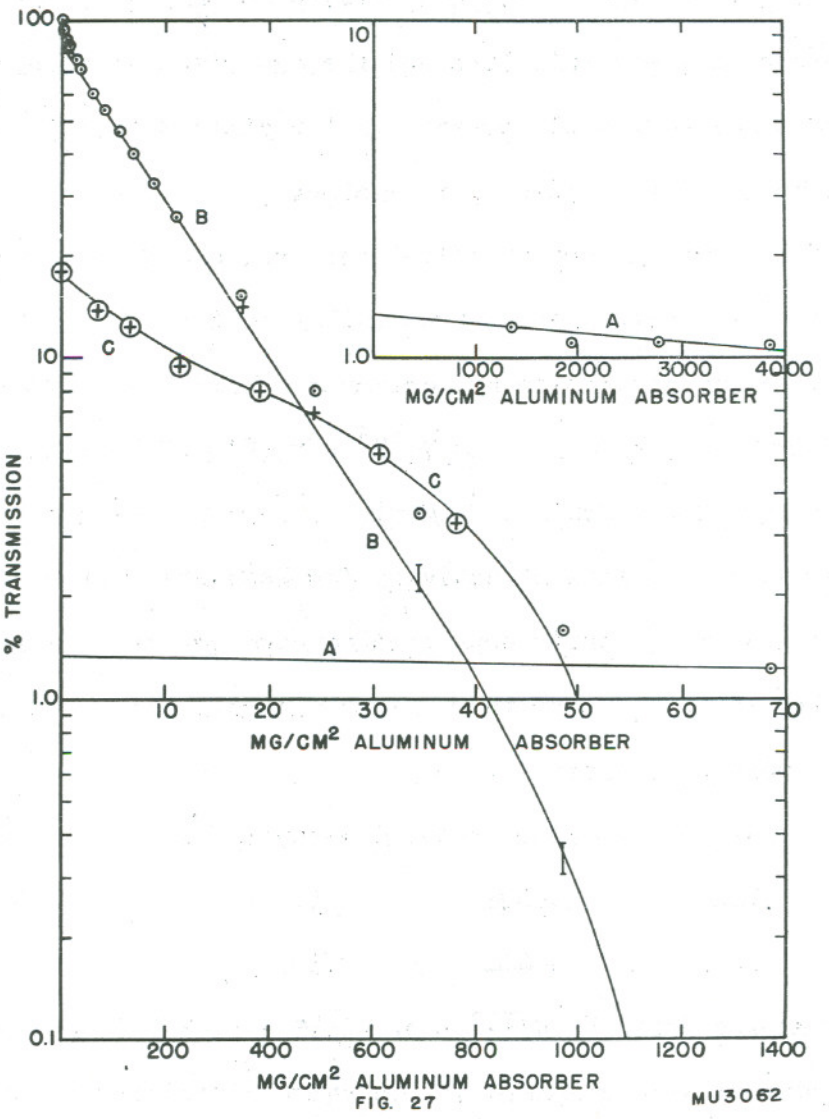
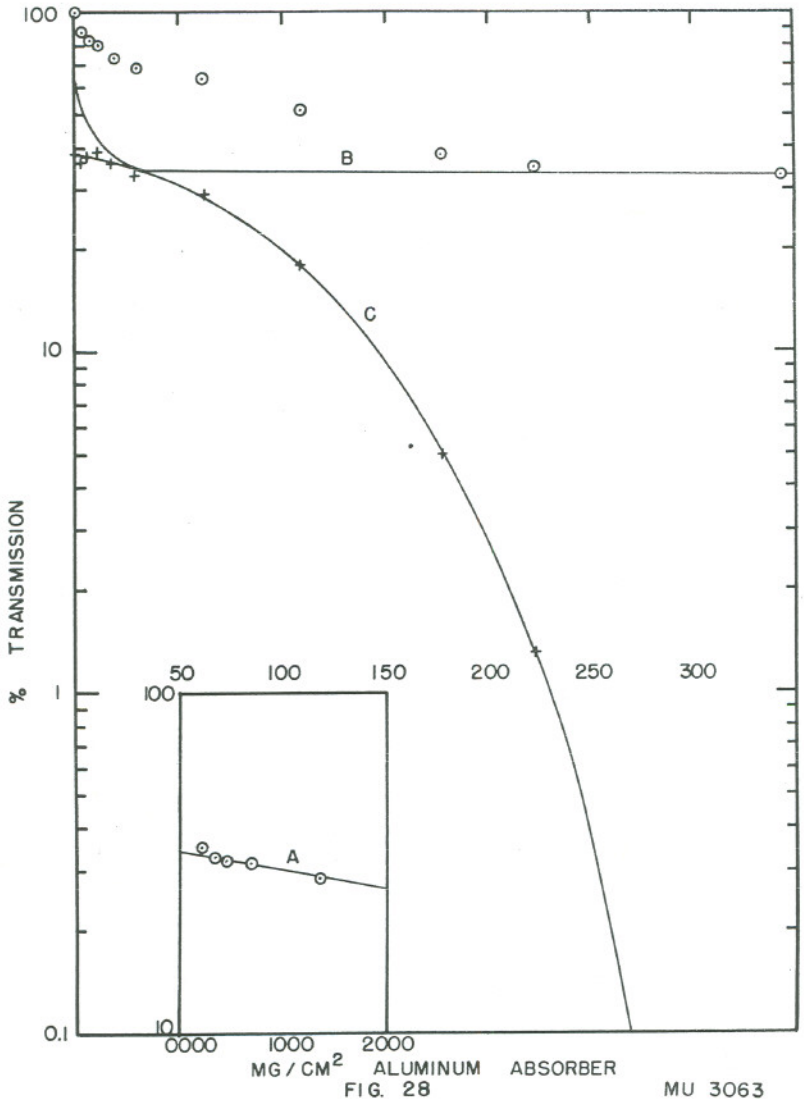


FIG. 27

Aluminum absorption data curve of short half-life activities from Nd^{145} + p bombardments. Electromagnetic background (A), 2 Mev negatron of Pm^{150} (B), ~0.2 Mev particle (?) (C)



Aluminum absorption data curve of long-lived activities from Nd^{144} + p bombardments. K x-rays and gamma ray background (A), (A) plus L x-rays (B), 0.84 Mev beta particle (?) (C)

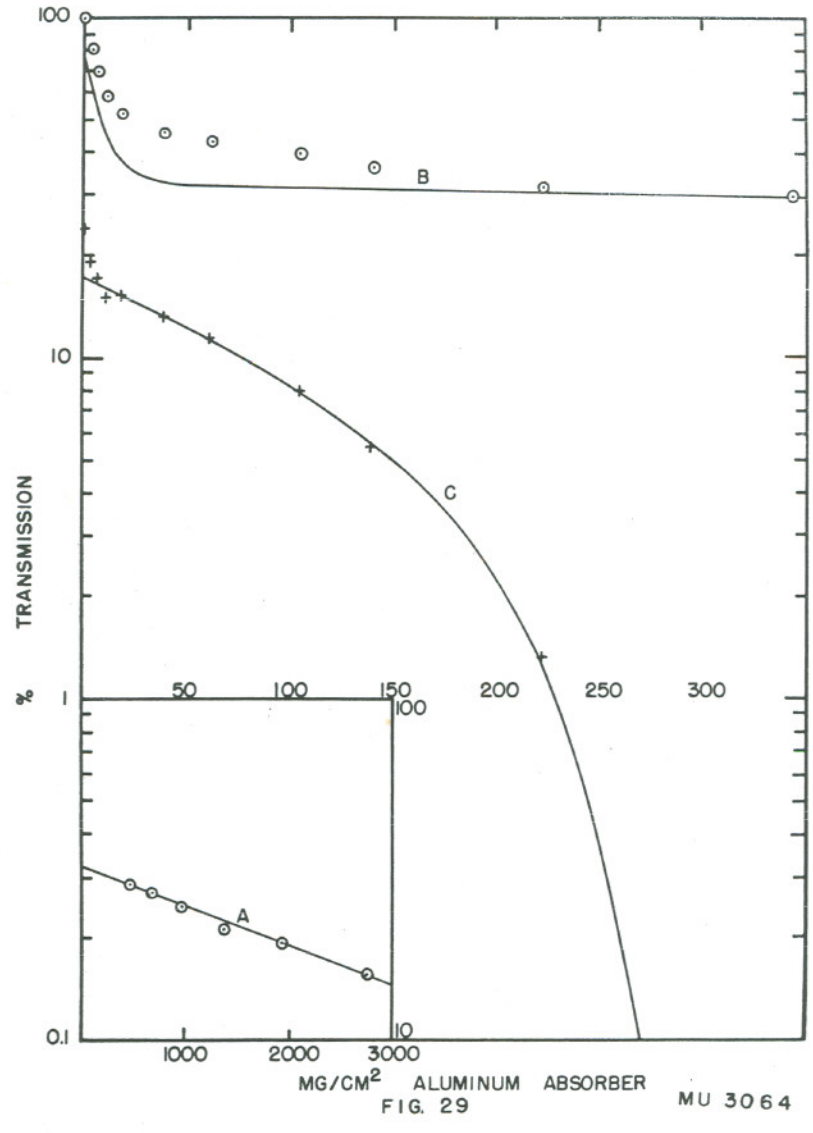


FIG. 29

Aluminum absorption data curve of long-lived activities from Nd^{143} + p bombardments. K x-rays and gamma rays (A), K and L x-rays and gamma rays (B), 0.74 Mev beta particle (?) (C)

	Nd K x-rays:	0.18 ± 0.02 Mev gamma ray:		0.42 ± 0.02 Mev gamma ray:		0.61 ± 0.02 Mev gamma ray	
$Nd^{145} + p$	1.0	:	0.2	:	0.09	:	0.1
	Nd K x-rays:	0.16 ± 0.02 Mev gamma ray:		0.44 ± 0.02 Mev gamma ray:		0.74 ± 0.02 Mev gamma ray	
$Nd^{146} + p$	1.0	:	0.7	:	0.4	:	0.2

For comparison's sake the result of a similar examination of a sample from $Pr^{141} + \alpha$ bombardment given in the preceding section is repeated.

	Nd K x-rays:	0.16 ± 0.02 Mev gamma ray:		0.45 ± 0.02 Mev gamma ray:		0.62 ± 0.02 Mev gamma ray	
$Pr^{141} + \alpha$	1.0	:	0.6	:	0.2	:	0.3

Samples from the bombardments of Nd^{145}, Nd^{146}, and Nd^{148} were examined for alpha emission, but none was observed.

Assignments

The cross sections given in Table 7 were corrected for the percentage in the total sample of the isotope for which they were calculated. To facilitate comparison, the resulting values for each sample were then divided by the cross section for the production of the 161 min activity from it. The previous assignments of the 161 min, 5.3 day, 42 day, and 3.7 yr activities were used to calculate the first four columns of Table 8. The results are in agreement with the previous assignments. Their variation is to be expected in view of the uncertainties of the bombardments. The cross sections for the reactions producing activities with 10 to 60 hr half-lives were calculated using the half-life observed in each case but corrected as if all these activities (see Table 7)

Table 8

Corrected and Normalized Cross Sections for Nd + p Bombardments

Sample Enriched in	σ For Production of Activity								
	161 min Pm^{150}	5.3 d Pm^{148}	42 d $Pm^{148-147}$	3.7 yr Pm^{147}	10 to 60 hr Pm^{149}(?)	long Pm^{146}	long L x-rays	long K x-rays + γ rays	long L and K x-rays + γ rays
142	1.0	2.8			1.0				54.0
143	1.0	0.4			0.1	0.2	0.3	1.0	1.3
144	1.0	0.2			0.5	0.1	11.0	64.0	70.0
145	1.0	1.4			0.8				33.0
146	1.0	1.1			2.0	0.1	26.0	185.0	156.0
148	1.0	1.8	0.9	1.8					
150	1.0				0.8				

were the 54 hr decay previously assigned to Pm^{149}. The values thus obtained lead to the conclusion that in spite of the variation in half-life observed, these decays are due to the reaction $Nd^{150}(p,2n)Pm^{149}$ in each case. It was assumed that the 12 to 20 hr activities observed for samples enriched in Nd^{143} through Nd^{145} were due to a 12 hr positron decay of Pm^{143}, Pm^{144}, or Pm^{145}. This gave cross sections varying by factors of over one hundred. Thus it was concluded that the variation in the half-life was due to the great uncertainties of the resolutions of the decay curves and that all decay with half-life between 10 and 60 hours was due to Pm^{149}. The cross sections support this assignment since they agree well considering all the sources of error present. The cross sections in the column headed "long, Pm^{146}" were those calculated for the 0.7 Mev beta particle observed in the long half-life decay using the ratios found in absorption and assuming a 285 day half-life. The values were corrected on the assumption that this activity was formed from $Nd^{146}(p,n)Pm^{146}$ reaction. This choice, based on the values of σ_β in Table 7 and on comparison of the absorption ratios, is justified by the consistency of the results. The factor of 10 between the cross sections for the formation of Pm^{150} and those for the formation of Pm^{146}, serve as a strong indication that the half-life of Pm^{146} is longer than 285 days. A particle of similar energy was observed in low yield in Pr^{141} + α bombardments. If it is assumed to arise from the decay of Pm^{143}, it explains the fact that the calculated formation cross section of Pm^{146} from the sample enriched in Nd^{143} is 0.2 millibarn instead of 0.1 millibarn.

The last three columns were calculated on the assumption that all the electromagnetic radiation was emitted with a 285 day half-life by

Pm^{143}. The L x-ray and K x-ray plus gamma ray cross sections were calculated from the ratios observed in the absorptions. The last column was obtained by assuming an average counting efficiency of 1 percent for the total electromagnetic radiation. In the cases of Nd^{142} + p and Nd^{145} + p bombardments where absorption measurements were not possible, the formation of Pm^{146} with a 0.2 millibarn cross section was assumed. The contribution of beta decay calculated from this was subtracted from the total decay and the cross section for the production of Pm^{143} with the net decay calculated as before. The result of these maneuverings is far from conclusive since the cross sections for Pm^{143} vary with factors of over one hundred. Even if a portion of the electromagnetic radiation is assigned to Pm^{146}, there are still large discrepancies, and indeed, it was found that the only assumption which brought uniform results was that the long lived electromagnetic radiation belonged to all isotopes from Pm^{142} to Pm^{146}. It need not be pointed out that these results are very puzzling.

The results of the gamma spectrometer investigation of the samples shed some light on the problem. The great similarity of the energies in all five samples makes interpretation difficult, but this much is obvious. Pm^{143} emits principally K x-rays, little or no 0.17 Mev gamma rays, probably some 0.45 Mev and some 0.69 Mev gamma rays. Pm^{144} emits K x-rays, 0.17 Mev, 0.43 Mev, and 0.61 Mev gamma rays. The electromagnetic radiation of the Nd^{145} + p sample is a mixture of that of Pm^{143} and Pm^{144}. That of the Nd^{146} + p sample seems to contain not only a similar mixture, but 0.16 Mev and 0.44 Mev gamma rays belonging to Pm^{146}. The sample from the Pr^{141} + a bombardment is seen to be Pm^{144} at least

predominantly. Thus the empirical excitation curve must be reinterpreted as being the sum of the $Pr^{141}(\alpha,n)Pm^{144}$ and the $Pr^{141}(\alpha,2n)Pm^{143}$ excitation functions. The 285 day half-life observed by Hicks and Wilkinson[28] is probably the sum of the half-lives of Pm^{143} and Pm^{144} which are of that order of magnitude and too similar to be resolved. These hypothesis will be checked by periodic repetitions of the gamma spectrometer investigations (for summary, see Table 9).

E. Short Proton Bombardments of Neodymium Enriched in Nd^{142}

Wilkinson and Hicks[28] have set an upper limit of 5 minutes on the half-life of Nd^{142} from the results of the Pr + α bombardments they performed.

Pr + α bombardments in the present work (Part III-C) yielded only a doubtful 2 to 3 min activity. When Nd^{142} + p bombardments of one or more hours duration (Part III-D) did not result in an activity assignable to Pm^{142}, calculations were performed (Part IV) which indicated that this isotope is a positron emitter and that its half-life is of the order of magnitude of minutes. Thus it was attempted to observe such an activity by 10 min Nd^{142} + p bombardments on the 60-inch cyclotron. The lack of results prompted a transfer to the linear accelerator and an 11 Mev bombardment energy. A 10 min positron emission was found, and attempts were made to characterize it. In the process of these, however, chemical separations proved that the activity did not belong to a rare earth isotope. Thus it was concluded that it belongs to N^{13}.[4] Subsequently, bombardments at energies of 32 Mev and 20 Mev were found to produce a 20 min positron emitter assigned to Pm^{141}.

Measurements

A 10.0 ± 0.3 min activity alone was observed on four occasions from 10 min bombardments of Nd^{142} by 9.6 to 10.6 Mev protons from the linear accelerator. The crude beta spectrometer indicated a positron decaying with a half-life of that order of magnitude. On a fifth bombardment careful chemical separations involving hydroxide, fluoride, and oxalate precipitations proved conclusively that the isotope was not a rare earth.

A 20.0 ± 1.0 min activity (Fig. 30) was observed from 10 min bombardments of Nd^{142} by 32 Mev and 20 Mev protons from the linear accelerator. This activity was proved chemically to belong to a rare earth. Examination on the crude beta spectrometer showed a positron with energy of 2.4-2.8 Mev (Fig. 31) decaying with a 20 ± 5 min half-life.

Assignments

Since the 20 min positron emitter has a production threshold greater than 12 Mev, and since it is observed in substantial yield at 32 Mev, it is almost certainly not produced by a Nd(p,n) reaction. Tentative assignment to Pm^{141} from a Nd^{142}(p,2n) reaction was made. This is confirmed by the non-observal of this isotope in the Pr + α bombardments.

Since no activity assignable to Pm^{142} has been observed, an upper limit of 2 minutes can be set on its half-life if it is short. If it is long, the lower limit is 10^2 years. These probabilities will be discussed in Part IV (see Table 9 for summary).

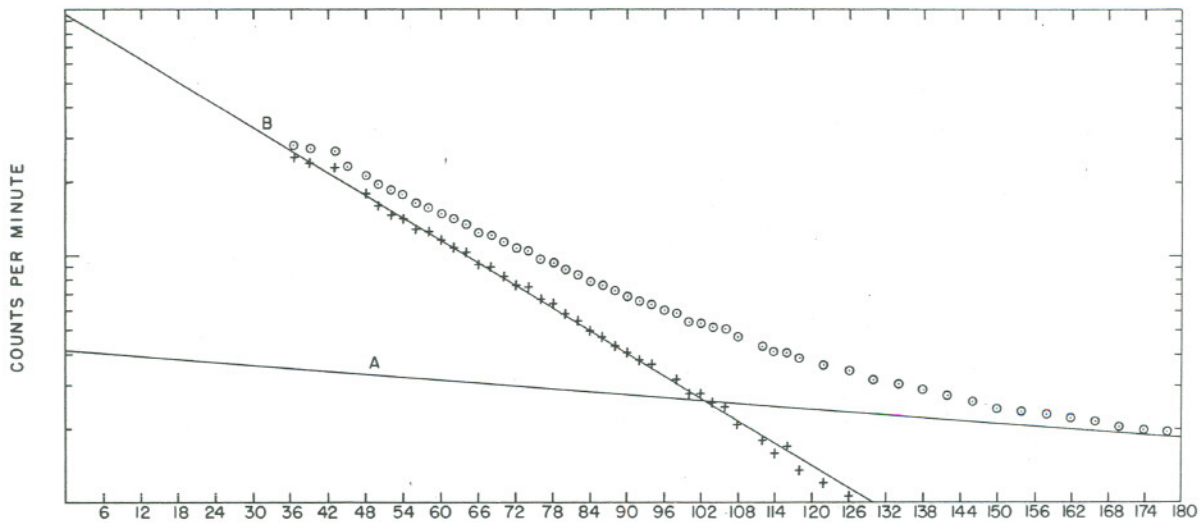


FIG. 30

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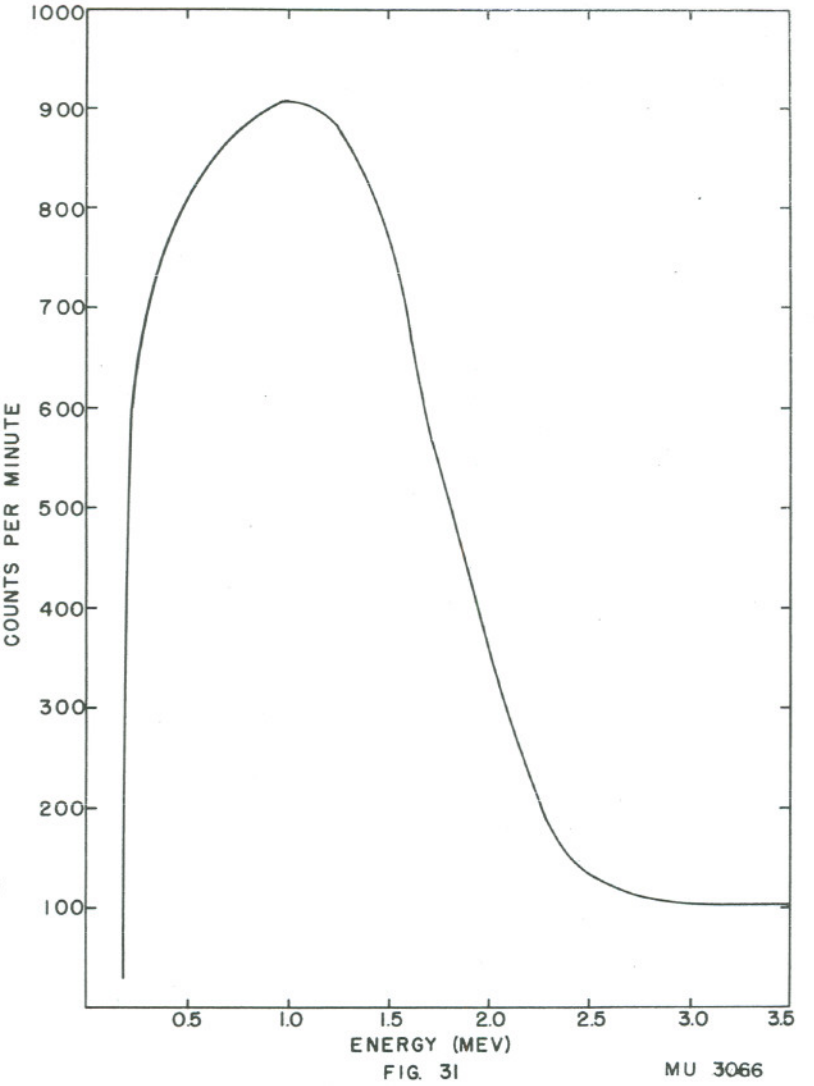
Decay of 20 minute activity produced by 10 minute Nd^{142} + p bombardments at ~20 Mev and ~32 Mev



Data from the crude beta spectrometer for the 20 minute activity from 10 minute Nd^{142} + p bombardments at ~20 Mev and ~32 Mev

IV. CONCLUDING REMARKS

There are many ways to verify a theory such as that of nuclear shell structure and not the least of these is that assumption of the theory gives a coherent picture from a set of data.

The experimental work in this paper was not uniformly conclusive, but it was found that Maria Mayer's theory offered reasonable explanation for some of its uncertainties. Conversely, the reliable data was found to agree with the theory.

This section deals with the attempts made to connect theory and experiment. The combined results from both are summarized in Table 10.

A. Pm^{141}

A calculation using a modified form of the Weiszäcker equation[5] gave 2.9 Mev as the energy difference between $_{60}Nd^{141}_{81}$ and $_{61}Pm^{141}_{80}$. In the case of positron emission, 1.02 Mev must be taken from this for the two electrons lost. The observed positron energy of 2.4 to 2.8 Mev may be explained by an increased binding energy for the 81st neutron which would add to the ordinary calculated value.

Since $_{59}Pr^{141}_{82}$, $_{63}Eu^{151}_{88}$, and $_{63}Eu^{153}_{90}$ all have their odd proton in the $2d_{5/2}$ state,[21,22] it is probable that the odd proton of the promethium isotopes will also be in this state. Thus the configuration of $_{61}Pm^{141}_{80}$ will probably be $D_{5/2}$ since paired nucleons do not contribute. Analogously to $_{56}Ba^{137}_{81}$,[21,22] $_{60}Nd^{141}_{81}$ should have its odd neutron in the $2d_{3/2}$ state and its configuration will then be $D_{3/2}$. The decay of $_{61}Pm^{141}_{80}$ to $_{60}Nd^{141}_{81}$ thus is allowed, involving a spin change of one and no parity charge ($\Delta I = 1$, $\Delta L = 0$). For a promethium isotope emitting a 2.8 Mev positron with a 20 min half-life, log ft = 5.3. Since

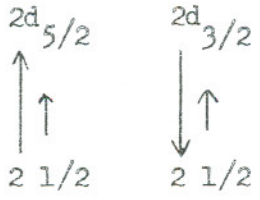
log ft = 4.8 to 5.5 for an allowed transition, theory and experiment are seen to agree.

B. Pm^{142}

When no activity belonging to Pm^{142} was observed from 2 hr bombardments, the following calculations were made to estimate its half-life. A calculation using a modified form of the Weiszäcker equation[5] gave the energy difference between $_{60}Nd^{142}_{82}$ and $_{61}Pm^{142}_{81}$ as 3.1 Mev. Since $_{60}Nd^{142}_{82}$ has a closed neutron shell and thus probably greater binding energy, the energy difference if anything else will be greater. With so much energy to be lost in decay, positron emission would be greatly favored over orbital electron capture.

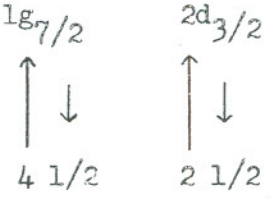
The energy of the positron emitted by $_{61}Pm^{142}_{81}$, the total decay energy minus 1.02 Mev for the two electrons lost, was assumed to be 2.1 Mev and the transition was chosen to be allowed to calculate a minimum half-life. $Log_{10}f = 2.4$ was obtained from the graphs of Feenberg and Trigg[68] and since $log_{10}ft = 4.8$ to 5.5 for an allowed transition, a minimum half-life of 4.2 to 21.0 minutes was found for the decay of $_{61}Pm^{142}_{81}$.

Nordheim[69] has given a set of rules which make it possible to discover the coupling of the total spins of an odd proton and an odd neutron in a nucleus. For instance, $_{61}Pm^{142}_{81}$ has an odd neutron which, by analogy to the odd neutron of $_{56}Ba^{137}_{81}$, should be in a $2d_{3/2}$ state.[21,22] As discussed in Part IV-A, the odd proton will be in a $2d_{5/2}$ state. Since Nordheim postulates that the total spins couple with the s vectors in the same direction, the situation is as indicated in the following diagram. The resultant total spin is $|5/2 - 3/2| = 1$ and the resultant



angular momentum $|2 - 2| = 0$. Thus the ground state of Pm^{142} should be an S_1 state which would decay by an allowed transition ($\Delta L = 0$, $\Delta I = 1$) to the S_0 ground state of $_{60}Nd^{142}_{82}$.

If, however, we consider another odd-odd isotope with 81 neutrons, $_{57}La^{138}_{81}$, another possibility becomes obvious. $_{57}La^{138}_{81}$ decays both by negatron emission and K-capture or positron emission with a half-life $\leq 1.2 \times 10^{11}$ years[70] but $\geq 3 \times 10^{9}$ years since it is observed in nature. According to Mayer,[22] the 57th proton should be in a $2d_{5/2}$ state, but is observed to have nuclear spin 7/2. Assuming that the 61st proton of $_{61}Pm^{142}_{81}$ is analogously in a $1g_{7/2}$ state, coupling with the $2d_{3/2}$ state of the 81st neutron will occur as indicated. The resultant total spin is



$|7/2 + 3/2| = 5$ and the resultant angular momentum $|4 + 2| = 6$. Thus an I_5 nuclear state could be postulated for $_{61}Pm^{142}_{81}$ and a highly forbidden ($\Delta L = 6$, $\Delta I = 5$) transition to the S_0 state of $_{60}Nd^{142}_{82}$ results. Since promethium has not been observed in nature, the upper limit for its half-life is 3×10^{9} years.

Two possibilities become evident for the decay of Pm^{142} from the experimental limits and the above considerations. Either Pm^{142} decays by positron emission with half-life less than 2 minutes (in which case the energy of the positron must be greater than 2.6 Mev), or it decays by positron emission or electron capture with a half-life between 10^2 and 3×10^9 years.

The description of the reasoning in the preceding paragraphs is detailed to serve as an example. In the following sections much of the detail will be omitted.

C. Pm^{143}

Since the decay

$$_{61}Pm^{143}_{82} \xrightarrow[285\ d]{\text{oec},\gamma} {}_{60}Nd^{143}_{83}$$

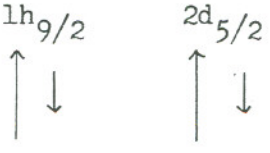
involves the addition of a single neutron to a closed shell, the 2 Mev decrease in binding energy for the single neutron given by Suess (Part I-B) must be taken from the 0.86 Mev of decay energy calculated from the Weiszäcker equation. Either the above calculation is badly at fault or the observed decay is from an excited level of the $_{61}Pm^{143}_{82}$. Since the first excited level would be at a much greater energy than the ground state, it would be expected to decay instantaneously by gamma emission. Thus in this case, observation and calculation cannot be reconciled.

In order to see if the 0.6 Mev electron observed in the Pm^{143} decay could be due to internal conversion, the K conversion coefficient for a ~0.7 Mev gamma ray was calculated from the relation $a_K \propto Z^3$ and the curve for Z = 84 given by Fermi.[71] A value of three electrons per

thousand gamma rays was obtained whereas 17 electrons per thousand gamma rays were seen (Part III-C). Considering the approximate nature of the calculation this constitutes an agreement. Thus the electron is postulated to be an ~0.64 Mev K conversion electron for a 0.69 Mev gamma ray.

D. Pm^{144}

The mass difference between $_{60}Nd^{144}_{84}$ and $_{61}Pm^{144}_{83}$ was found to be 2.05 Mev which would be decreased by 0.6 Mev due to the change from two to one open shell neutrons. This may be calculated from the values given by Suess, e.g., the binding energy of the two neutrons above the closed 82 shell in $_{60}Nd^{144}_{84}$ is decreased by 1.3 Mev each while that of the last neutron in $_{61}Pm^{144}_{83}$ is decreased by 2.0 Mev, yielding a 2.6 - 2.0 = 0.6 Mev difference. Similarly, the energy difference between $_{62}Sm^{144}_{82}$ and $_{61}Pm^{144}_{83}$ was calculated to be 0.0 Mev from the Weiszäcker equation plus 2.0 Mev for the decrease in binding energy of the 83rd neutron of the promethium. Thus it would seem that $_{61}Pm^{144}_{83}$ should decay by both electron capture and negatron emission. On the basis of the various bombardments, the half-life of this isotope for electron capture should be not less than 200 days, and for negatron emission, not less than 10^2 yr if it occurs at all. This indicates a high degree of forbiddenness. If it is assumed that the 83rd neutron is in the first configuration following the closed shell ($1h_{9/2}$), the coupling will be as indicated and $_{61}Pm^{144}_{83}$ will be in an F_2 state. Since both



$_{60}Nd^{144}_{84}$ and $_{62}Sm^{144}_{82}$ are in the S_0 state, lowest level to lowest level transitions would be second forbidden ($\Delta L = 3$, $\Delta I = 2$). Actual occurrence of this is very unlikely since gamma rays are almost certain to be emitted, but the possibility of a highly forbidden decay is indicated.

E. Pm^{145}

The mass difference between $_{60}Nd^{145}_{85}$ and $_{61}Pm^{145}_{84}$ is -0.1 Mev but since $_{60}Nd^{145}_{85}$ has one more open shell neutron another 2 Mev must be taken from this. Decay from a higher level of $_{61}Pm^{145}_{84}$ must be postulated, and even so it is not surprising that no gamma emission is observed with the orbital electron capture. It is surprising, however, that there is not an isomeric transition to the ground state.

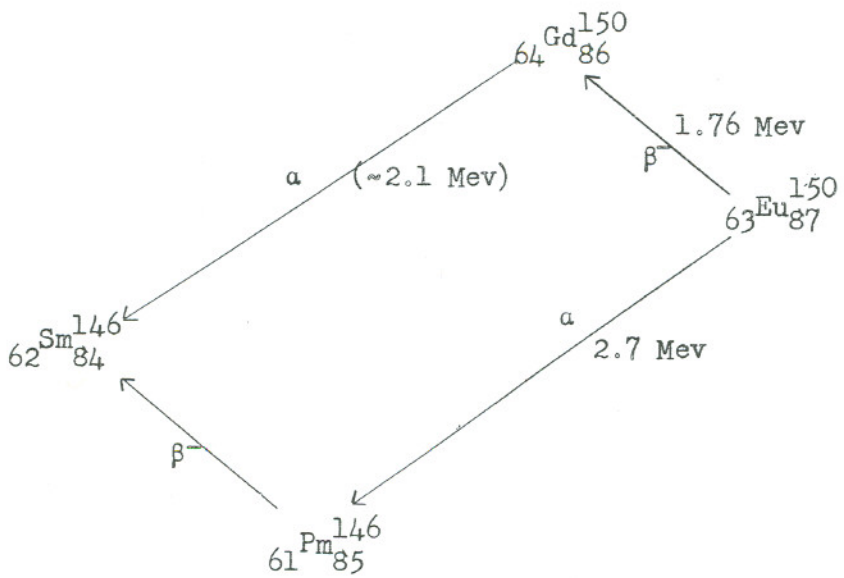
No choice will be made for the state of $_{61}Pm^{145}_{84}$, but the ground state of $_{60}Nd^{145}_{85}$ is chosen to be $H_{9/2}$ (Fig. 32a).

F. Pm^{146}

The charge of the particle emitted by $_{61}Pm^{146}_{85}$ was not established experimentally. Thus it could decay to $_{60}Nd^{146}_{86}$, a naturally occurring isotope, or $_{62}Sm^{146}_{84}$, which has not been proved beta unstable. Its absence in nature is probably explained in that it is an alpha emitter with half-life less than 3×10^9 yr. This has yet to be proved. In any case, $_{61}Pm^{146}_{85}$ could decay to either of its neighbors.

Weiszäcker calculations combined with the open shell neutron criterion gave 1.12 - 0.6 = 0.52 Mev for the energy difference of $_{60}Nd^{146}_{86}$ and $_{61}Pm^{146}_{85}$, and 0.93 + 2.0 = 2.93 Mev for that of $_{62}Sm^{146}_{84}$ and $_{61}Pm^{146}_{85}$. Thus if decay to $_{60}Nd^{146}_{86}$ occurs, it is by orbital electron capture, and it may be concluded that the particle observed was a negatron.

It is possible to show that the particle observed can be a negatron by a "closed cycle" type calculation. Rasmussen[67] has found that $_{64}Gd^{150}_{86}$ decays to $_{62}Sm^{146}_{84}$ by emitting a 2.7 Mev alpha particle. $_{63}Eu^{147}_{84}$ emits a 2.88 Mev alpha particle,[67] and since it is a first approximation rule in this region that the decay energy of a given isotope is roughly 0.8 Mev less than that of an isotope three mass numbers higher, $_{63}Eu^{150}_{87}$ should decay to $_{61}Pm^{146}_{85}$ by emitting a 2.1 Mev alpha particle. Concurrently with the experiments discussed in this paper, a sample of $_{63}Eu^{150}_{87}$ was examined for negatron decay. The maximum decay energy of this isotope as determined from a Fermi-Kurie plot on beta spectrometer data, was 1.76 Mev. It thus becomes possible to calculate the total negatron decay energy of $_{61}Pm^{146}_{85}$ by closing the following cycle.



This yields a total decay energy of 2.3 Mev for $_{61}Pm^{146}_{85}$, and thus not only the 0.7 Mev negatron observed but also 1.6 Mev of gamma rays are possible.

Assuming that the 85th neutron has a $1h_{9/2}$ state, $_{61}Pm^{146}_{85}$ like $_{61}Pm^{144}_{83}$ has a lowest state F_2. Negatron emission from this would not go to the ground state of $_{62}Sm^{146}_{84}$ but to a level 2.2 Mev higher (Fig. 32b). Since $\log_{10}ft = 8.6$ for $E_{\beta-} = 0.7$ Mev and t = 285 days, the transition is second forbidden.

G. Pm^{147}

The mass equation together with the open-shell neutron criterion give -0.24 + 0.6 = 0.36 Mev for the energy difference between $_{62}Sm^{147}_{85}$ and $_{61}Pm^{147}_{86}$. This is in remarkably close agreement with the 0.223 Mev energy of the negatron solely emitted. It is tempting to say that the effect of the open-shell neutrons is 0.14 Mev weaker for the 86th neutron than for the 84th, but this is putting too much weight on the theoretical values.

$Log_{10}ft = 7.6$ and so the transition is either first or second forbidden. If the 85th neutron is again assumed to be in a $1h_{9/2}$ state, then $_{62}Sm^{147}_{85}$ is in an $H_{9/2}$ halves state. $_{61}Pm^{147}_{86}$ is in a $D_{5/2}$ state and the decay is theoretically second forbidden ($\Delta L = 3$, $\Delta I = 2$) (Fig. 32c), agreeing with the observations.

H. Pm^{148}

The energy difference between $_{62}Sm^{148}_{86}$ and $_{61}Pm^{148}_{87}$ was calculated to be 1.1 + 2.0 = 3.1 Mev in the usual manner. Since $_{61}Pm^{148}_{87}$ is known to emit a 2.3 Mev negatron and a 0.8 Mev gamma ray, a decay from its lowest state is indicated.

$Log_{10}ft = 9.86$ (t = 5.3 days and $\log_{10}f = 4.20$) and thus the transition is a higher forbidden one ($\Delta L = 0,1$; $\Delta I = 3,4$, etc.). Since

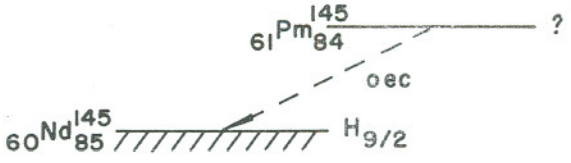


FIG. 32a

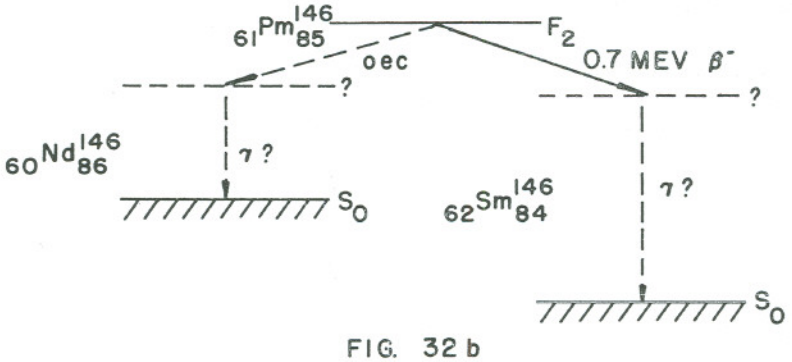


FIG. 32b

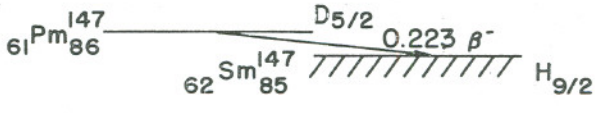


FIG. 32c

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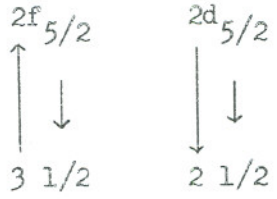
Possible simplest decay schemes for (a) $_{61}Pm^{145}_{84}$; (b) $_{61}Pm^{146}_{85}$; (c) $_{61}Pm^{147}_{86}$, 3.7 year isomer

the assumption that the 87th neutron was in a $2f_{5/2}$ state gave good results in the case of $_{62}Sm^{149}_{87}$ (see Part IV-I)it was also used here. This coupled with the $2d_{5/2}$ state of the proton as indicated to give a P_0 state. The ground state of $_{62}Sm^{148}_{86}$ is of course S_0 (Fig. 33a).



I. The 42 Day $Pm^{147-148}$

The total decay energy for the 42 day isotope (Part III-B) was 2.7 Mev. At first this activity was thought to belong to an isomer of $_{61}Pm^{148}_{87}$ since an odd-odd isotope in this region might very easily have two low-lying states differing little in energy but greatly in spin. This would cause a gamma transition between them to be very highly forbidden, and thus favor beta decay over isomeric transition for the isomer at the higher energy. The total decay energy of the 5.3 day isomer of $_{61}Pm^{148}_{87}$ is 3.1 Mev, and thus if the 42 day activity also belongs to a $_{61}Pm^{148}_{87}$ isomer, the levels are 0.4 Mev apart. Since the measurements are not very accurate, this may even be considerably less, increasing the plausibility of the assignment.

A tentative assignment of the activity to an isomer of $_{61}Pm^{147}_{86}$ is indicated only by a purely empirical consideration. The 3.7 yr $_{61}Pm^{147}_{86}$ has frequently been observed without the 42 day activity, but only as the result of the beta decay of $_{60}Nd^{147}_{87}$ (Table 3). The 5.3 day activity, however, has been observed without the 42 day activity from

bombardments that produced $_{61}Pm^{148}_{87}$ directly (Table 3). The two occasions on which the 42 day activity were observed were both bombardments in which the isotope could have been formed directly (high energy fission, Nd + p) and in which it could have been given considerable excitation energy. Thus it was concluded that the 42 day half-life could not belong to an isomer of $_{61}Pm^{148}_{87}$ or it would have been observed previously. As an isomer of $_{61}Pm^{147}_{86}$, there never before had been enough bombardment energy to produce it in company with the 3.7 yr isomer.

The assumption that the 42 day activity belongs to an isomer of $_{61}Pm^{147}_{86}$ may, however, be shown to lead to internal inconsistency. $Log_{10}ft$ was found to be 10.94 for the 2.7 Mev negatron corresponding to a highly forbidden transition (ΔL = 0,1, etc.; ΔI = 3,4, etc.). The 0.7 Mev negatron had $log_{10}ft$ = 8.34 and thus a second forbidden transition (ΔL = 1,3,5; ΔI = 2). The ground state of $_{62}Sm^{147}_{85}$ has previously been shown to be $H_{9/2}$. An $S_{1/2}$ state for the excited $_{61}Pm^{147}_{86}$ isomer was found to be the best of several possible choices (the states available for an odd proton $50<Z<82$ are $1g_{7/2}$, $2d_{5/2}$, $3d_{3/2}$, $3S_{1/2}$, and $1h_{11/2}$),[21,22] giving a highly forbidden (ΔL = 5, ΔI = 4) 2.7 Mev transition to the ground state. The 0.7 Mev negatron emission was assumed to take the nucleus to a $F_{5/2}$ level of $_{62}Sm^{147}_{85}$, a second forbidden transition (ΔL = 3, ΔI = 2). If one quantum of 1 Mev magnetic dipole radiation ($|I' - I| = 1, |I' + I| > 1$, ΔL = 0) is emitted, a $F_{7/2}$ level would be reached, and by emission of a second ($|I' - I| = 1, |I' + I| > 1$, ΔL = 2), the $H_{9/2}$ ground state (Fig. 33b). (The states available for an odd neutron $82<N<126$ are $1h_{9/2}$, $2f_{7/2}$, $2f_{5/2}$, $3p_{3/2}$, $3p_{1/2}$, $1i_{13/2}$.)[21,22] The percentages observed of each of the two decay paths agree with its

relative forbiddenness (92.6 percent for the second forbidden and 7.4 percent for the higher forbidden). However, this decay scheme implies that an isomeric transition between the two levels of $_{61}Pm^{147}_{86}$ has a half-life long compared to 42 days. The energy of the transition is ~2.5 Mev and the spin change is two with no parity change according to the level assignments given above. Using the half-life formula given by Bethe,[72] this transition should have a 7×10^{-11} second half-life. In order for a 2.5 Mev gamma emission to have a half-life $\geq$42 days, $\Delta I \geq 7$, and this is not possible for an odd-even isotope.

If the 42 day activity belongs to an isomer of $_{61}Pm^{148}_{87}$, then the decay scheme is as given in Fig. 33a. No level assignments can be attempted without further knowledge. As previously discussed, there would only be a 0.4 Mev energy difference between the isomeric levels. It was calculated, therefore, that there must be a difference in the nuclear spins of the two levels. This is entirely possible.

J. Pm^{149}

The energy difference between $_{62}Sm^{149}_{87}$ and $_{61}Pm^{149}_{88}$ was calculated to be 0.7 + 0.6 = 1.3 Mev while the radiation emitted by $_{61}Pm^{149}_{88}$ was observed to be a ~1 Mev negatron and a 0.3 to 0.35 Mev gamma ray. The close agreement indicated a transition from the lowest state of $_{61}Pm^{149}_{88}$ ($D_{5/2}$) to the ground state of $_{62}Sm^{149}_{87}$. The 87th neutron of the latter was assigned to the $2f_{5/2}$ state, giving a $F_{5/2}$ ground state. Since $\log_{10}ft$ = 6.94 (t = 54 hr) indicated a first forbidden (ΔL = 1, ΔI = 0,1) negatron transition, the level in which the emission of the negatron left the nucleus was assumed to be a $P_{3/2}$ state (ΔL = 1, ΔI = 1). One quantum of 0.3 Mev magnetic dipole radiation ($|I' - I|$ = 1,

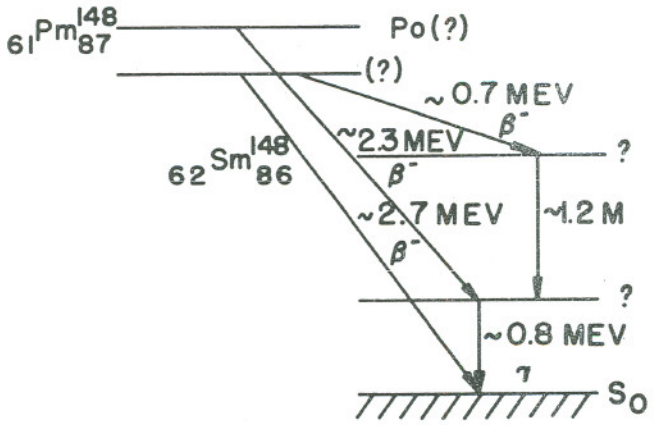


FIG. 33a

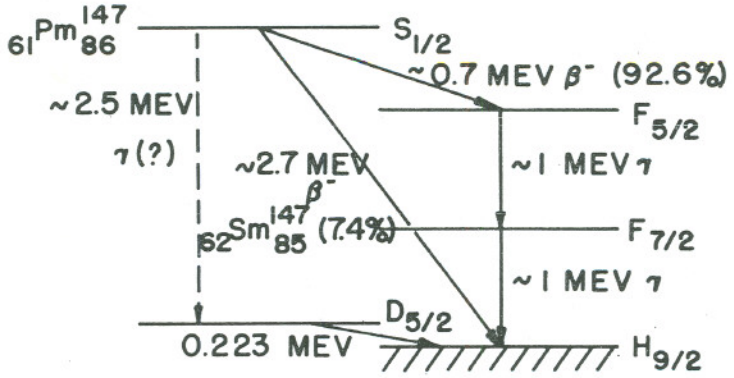


FIG. 33b

MU 3068

Decay schemes for (a) $_{61}Pm^{148}_{87}$, 5.3 day and 42 day isomers (?);

(b) $_{61}Pm^{147}_{86}$, 42 day isomer (?)

$|I' + I| > 1$, $\Delta L = 2$) could be assumed to be emitted finally taking the nucleus to the ground state (Fig. 34a).

K. Pm^{150}

The energy difference between $_{62}Sm^{150}_{88}$ and $_{61}Pm^{150}_{89}$ was calculated to be 2.70 + 2.00 = 4.7 Mev. The observed maximum decay energy was ~3.5 Mev. Thus it must either be assumed that the effect of the open shell neutrons has been greatly decreased or that the assignment of this activity is at fault. In view of the approximations involved in the theoretical consideration and the presence of seven open-shell neutrons, the former assumption was chosen.

Calculation of $\log_{10}ft$ (t = 161 min) yielded 6.78 for the 2 Mev and 7.51 for the 3 Mev negatron emitted. Thus the former transition is first forbidden ($\Delta L = 1,3$; $\Delta I = 0,1$) and the latter either first or second ($\Delta L = 1,3$; $\Delta I = 2$). Since this is the case of an odd-odd nucleus decaying to an even-even, assignment of the higher levels of the daughter is very difficult. The ground state of course is S_0. Assuming the 89th neutron to be in the same state as the 87th, $_{61}Pm^{150}_{89}$ was chosen to be in the P_0 state (Part IV-I) (Fig. 34b).

L. Pm^{151}

The energy difference between $_{62}Sm^{151}_{89}$ and $_{61}Pm^{151}_{90}$ was calculated to be 1.57 + 0.6 = 2.17 Mev. Rutledge and co-workers[30] found a 27.5 hr activity consisting of a 1.1 Mev negatron, a weak 0.615 Mev gamma ray and a large number of lower energy gamma rays. Assuming the complete transition energy to be the sum of 1.1 and 0.615 Mev, it is again seen that the effect of the open-shell neutrons must nearly disappear for the 7th and 8th neutrons above the closed shell.

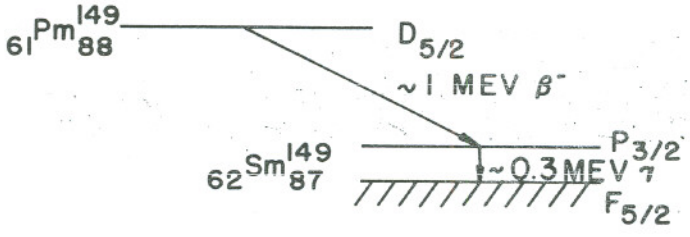


FIG. 34a

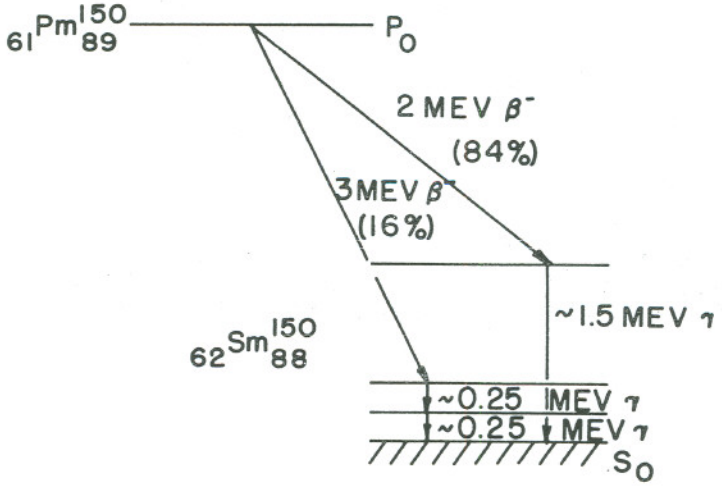


FIG. 34b

MU 3069

Possible decay schemes for (a) $_{61}Pm^{149}_{88}$; (b) $_{61}Pm^{150}_{89}$

$Log_{10}ft = 7.78$ for $t = 27.5$ and thus the negatron emission is a second forbidden transition. The lowest state of $_{61}Pm^{151}_{90}$ is the $D_{5/2}$ state while the ground state of $_{62}Sm^{151}_{89}$ is $F_{5/2}$ since the 89th neutron has been seen to be $2f_{5/2}$ (Part IV-J). If the negatron emission is assumed to lead to a $P_{1/2}$ state, it undergoes a second forbidden transition ($\Delta L = 1$, $\Delta I = 2$). The $P_{1/2}$ state can then decay by the emission of a 0.615 Mev quantum of electric quadruple radiation ($\Delta L = 2 \left| I' - I \right| = 2 \left| I' + I \right| > 2$) or by a succession of lower energy gamma rays.

M. Theoretical Considerations

The experimental and theoretical conclusions on the radioactive characteristics of the isotopes of promethium are summarized in Table 9. The notation is the same as that used in Tables 3 and 4. All values were found in the present experimental work with the exception of those with footnote indication. The designation (calc) indicates that the given value was arrived at on theoretical considerations. Table 10 summarizes the discussion of the results in connection with nuclear shell theory. $\Delta E_{(obs)}$ is the experimentally determined decay energy, t is the half-life and $log_{10}ft$ is calculated from them. $\Delta E_{(calc)}$ is the theoretical energy difference. The configurations and transition types are those values found in the preceding sections.

It has been shown that nuclear shell theory and especially the rules for open-shell neutrons, make it possible to explain much that is puzzling about promethium isotopes. The decay of Pm^{145} and Pm^{143} to what theoretically are heavier isotopes, is a possible exception, but even this can be accounted for by applying Kowarski's theory of the inequality of n-n and p-p binding. Conversely, the success of the theory is its vindication.

Table 9

Summary of Results: Radioactive Characteristics of Promethium Isotopes

A	Class	Type of Radiation	Half-Life	Energy of Radiation: Particles	Energy of Radiation: Electromagnetic	Produced by
141	b	β^+	20.0 ± 1.0 min	2.4 - 2.8 Mev		$Nd^{142}(p,2n)$
142		β^+	<2 min or 10^2 to 3 x 10^9 yr			
143	b	K,L x-ray γ,e^-	200-400 d	0.6 Mev (abs) 0.64 Mev (calc)	0.9 Mev (abs) 0.69 (calc) L and K x-rays 0.45,0.69 Mev (spec)	Pr(α,2n) $Nd^{143}(p,n)$
144	b	K,L x-ray,γ	200-400 d		0.18,0.43,0.61 Mev(spec) L and K x-rays	Pr(α,n) $Nd^{144}(p,n)$
145	a^{31}	K,L x-ray[31] e^-(?)	~30 yr[31]	low energy[31]	K and L x-rays[31]	$Sm(n,\gamma)Sm^{145}$ [31] EC
146	a	β^-	≥285 d	0.7 Mev (abs)		$Nd^{146}(p,n)$
146	a_{ms}[32]	β^-	long 3.7 yr[15,18,32]	0.223 Mev (spec)[38,39] 0.2 Mev		$Nd^{148}(p,2n)$
147 or 148	c	β^-,γ K x-rays(?)	42 ± 1 d	2.7 Mev (abs)(7.4%) 0.7 Mev (abs)(92.6%)	~1 Mev (abs) ~0.9 Mev (abs)[45] K x-rays(?)	$Nd^{148}(p,2n)$
148	a_{ms}[40]	β^-,γ K x-rays(?)	5.3 ± 0.1 d	2.3 Mev (abs) 2.5 Mev (abs)[18,40]	~1 Mev (abs) ~0.8 Mev (abs)[18,40] K x-rays(?)	$Nd^{148}(p,n)$
149	a_{ms}[41]	β^-,γ K x-rays(?)	54.4 ± 1.1 hr	1.0 Mev (abs + spec)	0.3 to 0.35 Mev (spec) K x-rays(?)	$Nd^{150}(p,2n)$
150	b	β^-,γ K x-rays(?)	161 ± 1 min	2.01 ± 0.03 Mev (84%)(spec) 3.00 ± 0.01 Mev (16%)(spec)	~1.4 Mev (abs) ~0.3 Mev(abs),K x-rays(?)	$Nd^{150}(p,n)$
151	a^{27}	β^-,γ [27]	27.5 hr[27]	1.1 Mev (spec)[27]	0.615 kev(abs,weak)[27] (see Table 1)	$Nd^{150}(n,\gamma)^{27}$ Nd^{151} β^-

Table 10

Application of Nuclear Shell Theory to the Decay of Promethium Isotopes - Summary

Decay	$\Delta E_{(obs)}$	t	$\log_{10} ft$	$\Delta E_{(calc)}$	Configuration Pm p	Configuration Pm n	Pm Nucleus	Configuration Daughter p	Configuration Daughter n	Daughter Nucleus	Transition
$_{61}Pm^{141}_{80} \xrightarrow{\beta^+} {}_{60}Nd^{141}_{81}$	2.4-2.8 Mev	20 min	5.3	2.9	$2d_{5/2}$	-	$D_{5/2}$	-	$2d_{3/2}$	$D_{3/2}$	allowed $\Delta L = 0$, $\Delta I = 1$
$_{61}Pm^{142}_{81} \xrightarrow{\beta^+} {}_{60}Nd^{142}_{82}$		<2 min or		3.1 Mev	$2d_{5/2}$	$2d_{3/2}$	S_1	-	-	S_0	allowed
		10^2 to 3 x 10^9 yr			$1g_{7/2}$	$2d_{3/2}$	I_5	-	-	S_0	highly forbidden
$_{61}Pm^{143}_{82} \xrightarrow{EC} {}_{60}Nd^{143}_{83}$	0.69 Mev γ	200-400 d		-1.15 Mev				-	$1h_{9/2}$	$H_{9/2}$	forbidden
$_{61}Pm^{144}_{83} \xrightarrow{\beta^-} {}_{62}Sm^{144}_{82}$				2.0 Mev	$2d_{5/2}$	$1h_{9/2}$	F_2	-	-	S_0	2nd forbidden $\Delta L = 3$, $\Delta I = 2$
$\xrightarrow[\gamma]{EC} {}_{60}Nd^{144}_{84}$	0.61 Mev γ	200-400 d		1.45 Mev	$2d_{5/2}$	$1h_{9/2}$	F_2	-	-	S_0	2nd forbidden $\Delta L = 3$, $\Delta I = 2$
$_{61}Pm^{145}_{84} \xrightarrow{EC} {}_{60}Nd^{145}_{85}$		30 yr		-2.1 Mev				-	$1h_{9/2}$	$H_{9/2}$	forbidden
$_{61}Pm^{146}_{85} \xrightarrow[\gamma]{EC} {}_{60}Nd^{146}_{86}$				1.72 Mev	$2d_{5/2}$	$1h_{9/2}$	F_2	-	-	S_0	$\Delta L = 3$, $\Delta I = 2$
$\xrightarrow{\beta^-} {}_{62}Sm^{146}_{84}$	0.7 Mev β^-	$\geq$285 d	8.6	2.93 Mev	$2d_{5/2}$	$1h_{9/2}$	F_2	-	-	S_0	2nd forbidden $\Delta L = 3$, $\Delta I = 2$

Table 10 (Cont.)

Decay	$\Delta E_{(obs)}$	t	$\log_{10} ft$	$\Delta E_{(calc)}$	Configuration Pm p	Configuration Pm n	Pm Nucleus	Configuration Daughter p	Configuration Daughter n	Daughter Nucleus	Transition
$_{61}Pm^{147} \xrightarrow{\beta^-} {}_{62}Sm^{147}_{85}$	2.7 Mev β^- 0.7 Mev β^- 2(1.0 Mev γ)	42 d	10.94 8.34		$3s_{1/2}$	-	$S_{1/2}$	-	$1h_{9/2}$	$H_{9/2}$	higher forbidden $\Delta L = 5$, $\Delta I = 4$ 2nd forbidden $\Delta L = 3$, $\Delta I = 2$
	0.223 Mev β^-	3.7 yr	7.6	0.36 Mev	$2d_{5/2}$	-	$D_{5/2}$	-	$1h_{9/2}$	$H_{9/2}$	2nd forbidden $\Delta L = 3$, $\Delta I = 2$
$_{61}Pm^{148}_{87} \xrightarrow{\beta^-} {}_{62}Sm^{148}_{86}$	2.3 Mev β^- 0.8 Mev γ	5.3 d	9.86	3.1 Mev	$2d_{5/2}$(?)	$2f_{5/2}$(?)	P_0(?)	-	-	S_0	higher forbidden
	2.7 Mev β^- 0.7 Mev β^- 2(1.0 Mev γ)	42 d	10.94 8.34					-	-	S_0	higher forbidden $\Delta L = 5$, $\Delta I = 4$ 2nd forbidden $\Delta L = 3$, $\Delta I = 2$
$_{61}Pm^{149}_{88} \xrightarrow{\beta^-} {}_{62}Sm^{149}_{87}$	1.0 Mev β^- 0.3 Mev γ	54 hr	6.94	1.3 Mev	$2d_{5/2}$	-	$D_{5/2}$	-	$2f_{5/2}$	$F_{5/2}$	1st forbidden $\Delta L = 1$, $\Delta I = 1$
$_{61}Pm^{150}_{89} \xrightarrow{\beta^-} {}_{62}Sm^{150}_{88}$	2 Mev β^- 3 Mev β^- ~1.4 Mev γ 2(~0.3 Mev γ)	161 min	6.78 7.51	4.7 Mev	$2d_{5/2}$	$2f_{5/2}$	P_0	-	-	S_0	1st forbidden 1st or 2nd forbidden
$_{61}Pm^{151}_{90} \xrightarrow{\beta^-} {}_{62}Sm^{151}_{89}$	1.1 Mev β^- ~0.6 Mev γ lower energy gamma rays	27.5 hr	7.78	2.17	$2d_{5/2}$	-	$D_{5/2}$	-	$2f_{5/2}$	$F_{5/2}$	2nd forbidden $\Delta L = 1$, $\Delta I = 2$

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$$\tau = 5.2 \times 10^{-21} (\ell!)^2 (20/\hbar\omega)^{2\ell+1} \text{ seconds}$$

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