Radioactive Isotopes of the Rare Earth Elements
II. Neodymium Isotopes

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The techniques of study of radioactive isotopes of the rare earth elements described previously have been applied to neodymium. Using the 60-inch Crocker Laboratory cyclotron, bombardments have been made of praseodymium with 19- and 9-Mev deuterons and 10-Mev protons. The characteristics of the neodymium isotopes are given below.

Isotope	Type of radiation	Half-life	Energy of radiation in Mev Particles	γ-rays	Produced by
Nd^{140}	K	3.3±0.1 days		K x-rays ~1.2 (weak)	Pr-d-3n
Nd^{141}	K, β^+(2%), γ	145±3 min.	0.7(β^+)	K x-rays 1.05	Pr-d-2n Pr-p-n

THE isotope Nd^{141} has been reported[1] to have a 2.5-hour half-life, and to emit positrons of energy 0.78 Mev. The activity was produced by proton bombardment of praseodymium,[1] by $n,2n$[1–3] and γ,n[1,3] reactions on neodymium, and possibly by a d,H^3 reaction on neodymium.[1,2] Chemical separations were not made.

In the present work, chemical separations were made by ion exchange columns. The 2.5-hr. activity has been characterized in more detail and allocated to Nd^{141}, while a new isotope, Nd^{140}, has been observed by Pr-d-3n reaction. This isotope decays by orbital electron capture to form the positron emitter Pr^{140}. In bombardments, spectroscopically pure, column separated praseodymium oxide, Pr_6O_{11}, was used.[4] The techniques of bombardments, chemical separations, and measurements of radioactivity have been previously described.[5]

I. 145-MINUTE Nd^{141}

Measurements of the radiation characteristics of this isotope were made on unseparated praseodymium which had been bombarded with 10-Mev protons. The proof of the chemical identity by the standard ion-exchange resin column procedure used previously was almost impossible, since the neodymium fraction leaves the column only after about 2.5 days. A 4-cm×0.4-cm column was therefore used, with conditions of flow, eluting agent, etc., the same as before. Although the separation of neodymium and praseodymium is unsatisfactory from the chemical standpoint, these fractions leave the small column in about 12 hours, a time short enough to allow detection of the 145-minute activity. Such an experiment was made for a Pr+d bombardment. The ratio of the 145-minute activity to the 19.3-hr. Pr^{142} activity formed by d,p reaction was estimated, corrections for decay from a reference time being made for both activities. The ratio decreased rapidly in samples following the "break through" of active material showing that the 145-minute activity elutes before praseodymium. Further, spectroscopic analysis showed that the

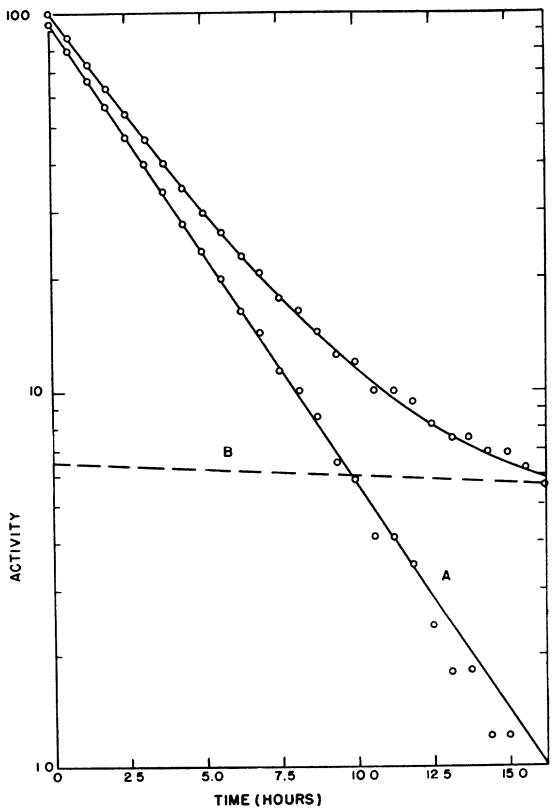

FIG. 1. Decay of 145-min. Nd^{141} positron from Pr+d bombardment followed on crude beta-ray spectrometer. A is the 145-min. activity and B the 3.3-day background.

[1] Kurbatov, MacDonald, Pool, and Quill, Phys. Rev. **61**, 106 (1942).
[2] M. L. Pool and L. L. Quill, Phys. Rev. **53**, 437 (1938).
[3] Law, Pool, Kurbatov, and Quill, Phys. Rev. **59**, 936 (1941).
[4] We are indebted to Mr. R. C. Lilly of this laboratory for preparation of this material.
[5] G. Wilkinson and H. G. Hicks, Phys. Rev. **75**, 1370 (1949).

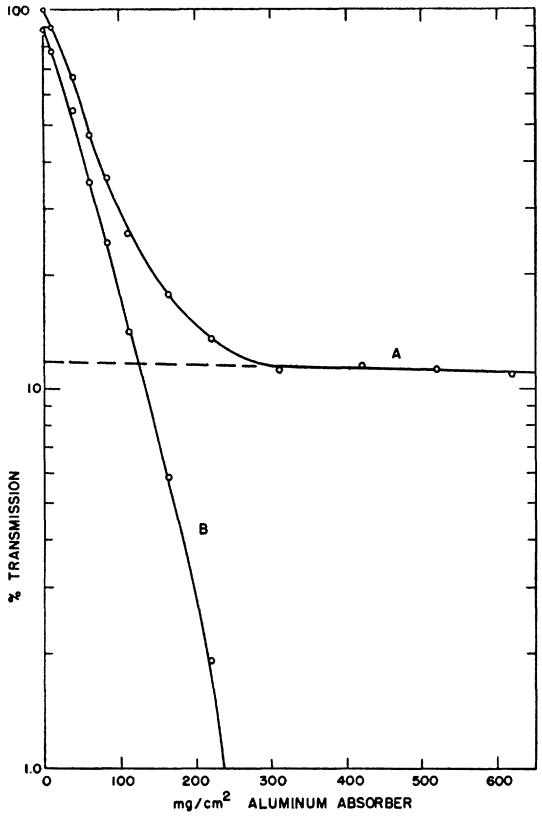

FIG. 2. Aluminum absorption of radiations of 145-min. Nd^{141} from $Pr+p$ bombardment. Electromagnetic radiation background (A), positron radiation range 245 mg/cm² (B).

19.3-hour activity followed the praseodymium, while in the first active sample where the 145-minute decay was observed, praseodymium was below the limits of detection. The chemical identity of the 145-minute activity as neodymium is, therefore, fairly certain.

The gross decay of the activity was followed through eight half-lives, the hard γ-radiation, through six half-lives, and the positron decay (Fig. 1), followed on a crude beta-ray spectrometer, also through six half-lives. The half-life is 145 ± 3 minutes.

The radiations consist of positrons, x-rays and γ-rays. No negative electrons were observed. The aluminum absorption curve of the 145-minute activity from $Pr+p$ bombardment, corrected for decay during the time of measurement, is shown in Fig. 2. The range of the electron, 245 ± 5 mg/cm² (0.7 Mev), agrees with the value of the positron energy measured on the crude beta-ray spectrometer. The lead absorption (Fig. 3) shows electromagnetic radiations of half-thicknesses 39 ± 2 mg/cm² (38 kev), ~4.5 g/cm² (0.5 Mev) and 11.5 g/cm² lead (1.2 Mev). The soft component agrees well with the K x-radiation of praseodymium, while the low abundant 0.5-Mev γ-ray is almost certainly annihilation radiation.

From the measurements, the following ratios were calculated; counting efficiencies of 0.5 percent for the K x-rays and 0.5-Mev γ-rays, and 1.2 percent for the 1.2-Mev γ-radiation were assumed, together with a fluorescence yield of 0.8 for the K x-radiation.

$$\beta^+ : K \text{ x-rays} : 0.5\text{-Mev } \gamma : 1.2\text{-Mev } \gamma = 0.02 : 1 : 0.02 : 0.02.$$

It is thus clear that the isotope decays by orbital electron capture with approximately 2 percent positron branching. The hard γ-radiation probably arises from an excited or metastable state of the daughter nucleus following electron capture.

From the measured K x-ray intensities, together with data on chemical yields and bombardment, the cross section for production of the 145-minute activity has been calculated. For 10-Mev protons a value of 3×10^{-2} barn was obtained. The deuteron cross sections are given in Table I below.

II. 3.3-DAY Nd^{140}

In the column separated neodymium fraction from $Pr+d$ bombardments a new long-lived ac-

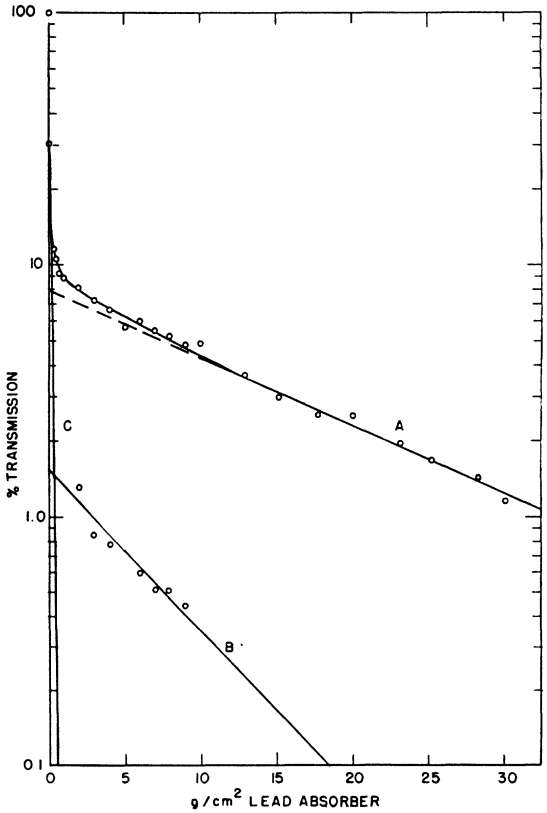

FIG. 3. Lead absorption of electromagnetic radiation of 145-min. Nd^{141} from $Pr+p$ bombardment. Hard γ-radiation (A), 0.5-Mev γ-ray (B) and K x-rays (C).

tivity was observed. The half-life, measured through seven periods, is 3.3±0.1 days. The aluminum absorption (Fig. 4) shows a hard electron, range 1150±50 mg/cm² (2.4 Mev) with K x-ray and γ-ray background. The lead absorption (Fig. 5) shows K x-radiation, 40 mg/cm² (38 kev) half-thickness, together with harder γ-radiation. If a small percentage of hard γ-radiation ~12 g/cm² lead (1.2 Mev) is assumed to be present, the bulk of the γ-radiation has a half-thickness of 4.6±0.1 g/cm² lead (0.51 Mev). The ratio of electrons to the various electromagnetic radiations obtained is:

Positrons: K x-rays: 0.51-Mev γ: ~1.2-Mev γ
= 0.2 : 1 : 0.2 : 0.01

Study of the electron radiations on the crude beta-ray spectrometer showed that positrons of energy 2.4 to 2.5 Mev, were present; no negative electrons were observed. The positron energy agrees well with that reported for the 3.5-minute Pr^{140}.[6]

The observed radiations of the 3.3-day activity are consistent with the isotope, Nd^{140}, decaying by

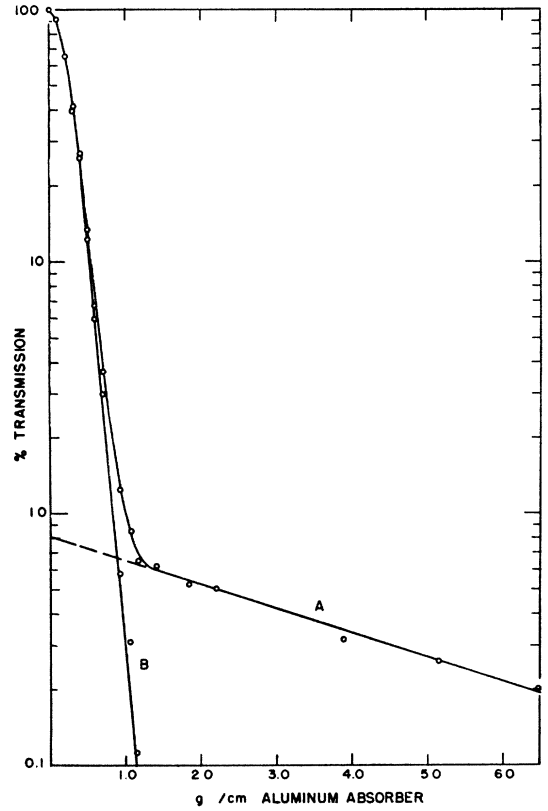

FIG. 4. Aluminum absorption of radiations of column separated 3.3-day Nd^{140} from Pr+d bombardment. Electromagnetic radiation background (A) positrons range 1150±50 mg/cm² (B).

[6] Huber, Lienhard, Scherrer, and Wäffler, Helv. Phys. Acta 18, 22 (1945); DeWire, Pool, and Kurbatov, Phys. Rev. 61, 544, 564 (1942); Weimer, Pool, and Kurbatov, Phys. Rev. 63, 67 (1943).

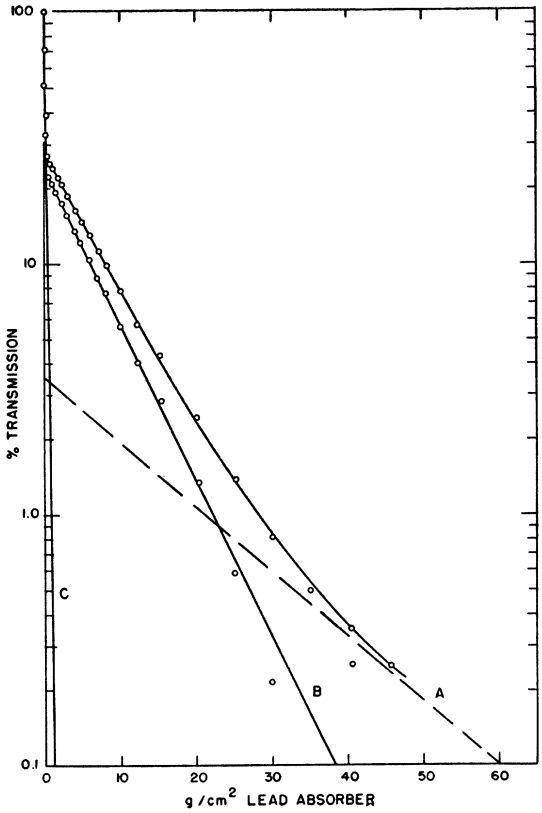

FIG. 5. Lead absorption of radiations of column separated 3.3-day Nd^{140} from Pr+d bombardment. Estimated hard γ-ray contribution (A), 0.51-Mev γ-ray (B) and K x-rays (C).

orbital electron capture, in equilibrium with its positron emitting Pr^{140} daughter. The observed 0.5-Mev γ-radiation is undoubtedly annihilation radiation; the harder γ-ray may be associated with either Pr^{140} or Nd^{140}. From the ratio of positrons to K x-radiation, it appears that the daughter Pr^{140} activity is not a pure positron emitter, but decays also by orbital electron capture, about two-thirds of the Pr^{140} disintegrations proceeding by this process.

A rapid chemical separation[7] of praseodymium and neodymium was attempted in order to show the chemical identity of the daughter of the 3.3-day activity. The mixed rare earth chlorides were fused with potassium hydroxide at 450°C for periods from 5 to 15 minutes. After extracting the melt with water, the mixed oxides were leached with 1N acetic acid. The neodymium oxide which dissolves preferentially was recovered as hydroxide for counting. The praseodymium remains largely in the residue insoluble in acetic acid. Although the chemical separation achieved is unsatisfactory, the activity of the neodymium fraction has been observed to grow, showing the formation of a daughter activity. The half-life of the daughter

[7] N. E. Ballou, private communication.

TABLE I. Cross sections in barns for deuteron reactions on praseodymium.

Half-life	Deuteron energy 19 Mev	9 Mev	Reaction	Isotope
19.3 hours	0.06	0.1	d,p	Pr^{142}
145 minutes	0.3	0.9	$d,2n$	Nd^{141}
3.3 days	0.08	—	$d,3n$	Nd^{140}

activity appears to be about 1.5 minutes, somewhat shorter than the value of 3.5 minutes reported[2] for Pr^{140}.

Additional evidence for the allocation of the 3.3-day activity to Nd^{140}, and its production by Pr-d-3n reaction has been obtained from yields in the deuteron bombardment of praseodymium. It was assumed that 0.6 of the measured K x-radiation of the 3.3-day activity represent one disintegration of Nd^{140}. In Table I are given the cross sections for production of the isotopes Pr^{142}, Nd^{141}, and Nd^{140}. The yields of the Pr^{142} were calculated from measured beta-particle activity. The absolute values of the yields may be in error in view of the uncertainties in counting efficiencies, etc., but the relative yields of the three reactions at the two bombarding energies should be more reliable.

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Columbium 96

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Cb^{96} has a half-life of 23.35 hours. It decays with the emission of negative beta-particles of maximum energy 0.67-Mev and 1.03-Mev gamma-rays. No x-rays are emitted. The assignment is made by using enriched isotopes of Zr.

INTRODUCTION

THE proton bombardment of zirconium was reported[1] to have produced Cb^{96} which has a half-life of 4 days. Later workers[2] reported its production by $(d,2n)$ and (d,α) reactions from zirconium and molybdenum, respectively, and changed the value of the half-life to 3 days. Recently[3] by the bombardment of enriched Mo^{98} with deuterons, the half-life has been further changed to 2.8 days and the decay characteristics given as negative beta-rays of 1.8 Mev by absorption in aluminum and gamma-rays of 1 Mev by coincidence absorption in lead.

The data to be presented in this paper are in complete disagreement with the above and will be briefly described.

TABLE I. Percent isotopic constitution of bombarded target samples of Zr.

Sample	Mass numbers 90	91	92	94	96
Natural Zr	51.46	11.23	17.11	17.40	2.80
Zr enriched in 90	91.7	3.5	2.2	1.8	0.8
Zr enriched in 91	6.2	86.6	5.9	1.3	0.1
Zr enriched in 92	2.4	2.2	92.7	2.3	0.4
Zr enriched in 96	8.7	2.2	4.3	10.3	74.6

RESULTS

Isotopes of zirconium separately enriched* in Zr^{90}, Zr^{91}, Zr^{92} and Zr^{96} were bombarded with 5-Mev protons and 10-Mev deuterons. The isotopic composition of the samples are shown in Table I.

A rotating target arrangement[4] was employed by which two isotopes could be bombarded simultaneously under the same beam and thus any activity produced could be associated with one or the other or none of the rotated isotopes. The degree of enrichment is such that the difference in activity between two samples will be a factor between 14 and 870, depending upon the selection made, if the particular activity be produced from either of the rotated isotopes. Deuteron bombardments produce

[1] L. A. DuBridge, private communication quoted by G. T. Seaborg and I. Perlman, Rev. Mod. Phys. **20**, 585 (1948).
[2] L. Jacobson and R. Overstreet, Plutonium Project Report CC-2345 (December 1944).
[3] G. E. Boyd, private communication (October 1948), quoted by G. T. Seaborg and I. Perlman, Rev. Mod. Phys. **20**, 585 (1948).

* Kindly supplied by the Y-12 Plant, Carbide and Carbon Chemicals Corporation, through the Isotope Division, US-AEC, Oak Ridge, Tennessee.
[4] D. N. Kundu and M. L. Pool, Phys. Rev. **74**, 1574 (1948).