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RADIOACTIVE ISOTOPES OF THE RARE EARTH ELEMENTS

II. NEODYMIUM ISOTOPES

Geoffrey Wilkinson and Harry G. Hicks

January 20, 1949

Berkeley, California
Radioactive Isotopes of the Rare Earth Elements

II. Neodymium Isotopes

Geoffrey Wilkinson and Harry G. Hicks

Radiation Laboratory and Department of Chemistry
University of California, Berkeley, California
January 20, 1949

Abstract

The techniques of study of radioactive isotopes of the rare earth elements described previously (5) 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 in Table I.

Table 1

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Type of Radiation</th>
<th>Half-Life</th>
<th>Energy of Radiation in Mev</th>
<th>Produced by</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ndl40 K</td>
<td>K</td>
<td>3.3±0.1</td>
<td>K x-rays 1.2</td>
<td>Pr-d-3n</td>
</tr>
<tr>
<td>Ndl41 K, 8⁺(2⁺), γ</td>
<td>145±3 min</td>
<td>0.7(8⁺)</td>
<td>K x-rays 1.05</td>
<td>Pr-d-2n</td>
</tr>
</tbody>
</table>

Pr-p-n
Radioactive Isotopes of the Rare Earth Elements

II. Neodymium Isotopes

Geoffrey Wilkinson and Harry G. Hicks

Radiation Laboratory and Department of Chemistry
University of California, Berkeley, California
January 20, 1949

The isotope Nd141 has been reported 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, by n,2n1,2,3 and


Y,γ1,3 reactions on neodymium, and possibly by a d,H3 reaction on neodymium1,2. 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 Nd141, while a new isotope, Nd140, has been observed by Pr-d-3n reaction. This isotope decays by orbital electron capture to form the positron emitter Pr140. In bombardments, spectroscopically pure, column separated praseodymium oxide, Pr6011, was used. The techniques of bombardments, chemical separations,

(4) We are indebted to Mr. R. C. Lilly of this laboratory for preparation of this material.
and measurements of radioactivity have been previously described.


I. 145-Minute Nd

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. x 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+ reaction bombardment. The ratio of the 145-minute activity to the 19.3-hr. Pr activity formed by the 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 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 $\gamma$-radiation through six half-lives, and the positron decay (Fig. I), followed on a "magnetic counter", also through six half-lives. The half-life is $145 \pm 3$ minutes.

The radiations consist of positrons, x-rays and $\gamma$-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. II. The range of the electron, $245 \pm 5$ mg/cm$^2$ (0.7 Mev), agrees with the value of the positron energy measured on the magnetic counter. The lead absorption (Fig. III) shows electromagnetic radiations of half-thicknesses 39 $\pm$ 2 mg/cm$^2$ (33 Kev), $\sim$4.5 g/cm$^2$ (0.5 Mev) and 11.5 g/cm$^2$ lead (1.2 Mev). The soft component agrees well with the K x-radiation of praseodymium, while the low abundant 0.5 Mev $\gamma$-ray is almost certainly annihilation radiation.

From the measurements, the following ratios were calculated: counting efficiencies of 0.5% for the K x-rays and 0.5-Mev $\gamma$-rays, and 1.2% for the 1.2 Mev $\gamma$-radiation were assumed, together with a fluorescence yield of 0.8 for the K x-radiation.

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

It is thus clear that the isotope decays by orbital electron capture with approximately 2% positron branching. The hard $\gamma$-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 \(3 \times 10^{-2}\) barns was obtained. The deuteron cross sections are given in Table II below.

II. 3.3-Day Nd\(^{140}\)

In the column separated neodymium fraction from Pr\(_d\) bombardments a new long-lived activity was observed. The half-life, measured through seven periods, is \(3.3 \pm 0.1\) days. The aluminum absorption (Fig. IV) shows a hard electron, range \(1150\pm 50\) mg/cm\(^2\) (2.4 Mev) with K x-ray and \(\gamma\)-ray background. The lead absorption (Fig. V) shows K x-radiation, \(40\) mg/cm\(^2\) (38 Kev) half-thickness, together with harder \(\gamma\)-radiation. If a small percentage of hard \(\gamma\)-radiation \(\sim 12\) g/cm\(^2\) lead (1.2 Mev) is assumed to be present, the bulk of the \(\gamma\)-radiation has a half-thickness of \(4.6 \pm 0.1\) g/cm\(^2\) lead (0.51 Mev).

The ratio of electrons to the various electromagnetic radiations obtained is:

Electrons: K x-rays: 0.51 Mev\(\gamma\) : \(\sim 1.2\) Mev\(\gamma\)  
\[0.2: 1 : 0.2 : 0.01\]

Study of the electron radiations on the magnetic counter 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\(^{140}\). 

The observed radiations of the 3.3-day activity are consistent with the isotope, Nd$^{140}$, decaying by orbital electron capture, in equilibrium with its positron emitting Pr$^{140}$ daughter. The observed 0.5 Mev $\gamma$-radiation is undoubtedly annihilation radiation, the harder $\gamma$-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 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 activity appears to be about 1.5 minutes, somewhat shorter than the value of 3.5 minutes reported\(^8\) for Pr$^{140}$.

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(7) N. E. Ballou, private communication.

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 II 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 $\beta^-$ 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.

**Table II**

**Cross Sections in Barns for Deuteron Reactions on Praseodymium**

<table>
<thead>
<tr>
<th>Half-Life</th>
<th>Deuteron Energy</th>
<th>Reaction</th>
<th>Isotope</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>19 Mev</td>
<td>9 Mev</td>
<td></td>
</tr>
<tr>
<td>19.3 hours</td>
<td>0.06</td>
<td>0.1</td>
<td>d,p</td>
</tr>
<tr>
<td>145 minutes</td>
<td>0.3</td>
<td>0.9</td>
<td>d,3n</td>
</tr>
<tr>
<td>3.3 days</td>
<td>0.08</td>
<td>---</td>
<td>d,3n</td>
</tr>
</tbody>
</table>

**Acknowledgements**

We would like to thank Professor J. G. Hamilton, Mr. T. Putnam, Mr. B. Rossi and the crew of the 60-inch Crocker Laboratory cyclotron for their cooperation and assistance in bombardments; we are also indebted to Professors G. T. Seaborg, I. Perlman and B. B. Cunningham for their continued interest and advice.

This work was done under the auspices of the Atomic Energy Commission.
Legends for Figures

Fig. I  Decay of 145-min. Nd$^{141}$ positron from Pr + d bombardment followed on "magnetic counter". A is the 145-min. activity and B the 3.3-day background.

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

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

Fig. IV  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/cm$^2$ (B).

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