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UNIVERSITY OF CALIFORNIA
Radiation Laboratory

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UNCLASSIFIED

New Neutron Deficient Radioactive Isotopes
of Rare Earths and Osmium

Betsy M. Jones

April 19, 1950

Berkeley, California
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<td>Re</td>
<td>12.7 h</td>
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<tr>
<td>75</td>
<td>K, e, +</td>
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| 60Nd | 22 m | 5.50h | 3.3d | 2.42h | 27.13 |
| 59Pr | 120m | 4.50h | 3.5m | 100 | 19.3h |
| 57Ge | 6.30h | 72.0h | 22h | 0.193 | 360h | 0.250 | 140d | 68.48 | 26d | 11.07 |
| 60Lc | 4h | 6.5m | 19.5h | 10m | >400y | 0.089 | 39.91 | 40.4h | 3.7h |
| 133 | 134 | 135 | 136 | 137 | 138 | 139 | 140 | 141 | 142 |

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NEW NEUTRON DEFICIENT RADIOACTIVE ISOTOPES
OF RARE EARTHS AND OSMIUM

Betsy M. Jones
Radiation Laboratory and Department of Chemistry
University of California, Berkeley, California

ABSTRACT

The new neutron deficient isotopes of lanthanum, cerium, praeeseodymium, neodymium, and osmium are products of \((p,xn)\) reactions. Bombardments with protons of energy 40 Mev and greater were made in the 184-inch cyclotron, and energies of 10 to 32 Mev were obtained with the linear accelerator.

The characterization of each new activity consisted of the determination of atomic number and mass number, the identification of the radiations, and the determination of the energies and the relative abundances of the various radiations.

Table I. New isotopes.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Radiations</th>
<th>Half-life</th>
<th>Energies in Mev</th>
<th>Produced By</th>
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<tr>
<td>(^{57})La(^{134})</td>
<td>(\beta^+,K)</td>
<td>6.5±0.25 min.</td>
<td>2.7 no (\gamma)</td>
<td>Ce(^{134}) decay</td>
</tr>
<tr>
<td>(^{58})Ce(^{133})</td>
<td>(\beta^+,K,\gamma)</td>
<td>6.30±0.1 hrs.</td>
<td>1.3 1.8</td>
<td>La(p,7n)</td>
</tr>
<tr>
<td>(^{58})Ce(^{134})</td>
<td>(K)</td>
<td>72.0±0.5 hrs.</td>
<td>no (\gamma)</td>
<td>La(p,6n)</td>
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<tr>
<td>(^{59})Pr(^{138})</td>
<td>(e^-,\beta^+,K,\gamma)</td>
<td>120±5 min.</td>
<td>0.24(e(^-)),1.4((\beta^+)) 0.16, 1.3</td>
<td>Ce(p,3n)</td>
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<tr>
<td>(^{59})Pr(^{139})</td>
<td>(\beta^+,K,\gamma)</td>
<td>4.50±0.5 hrs.</td>
<td>1.0 1.0</td>
<td>Ce(p,2n)</td>
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<tr>
<td>(^{60})Nd(^{138})</td>
<td>(\beta^+,K)</td>
<td>22±2 min.</td>
<td>2.4</td>
<td>Pr(p,4n)</td>
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<tr>
<td>(^{60})Nd(^{139})</td>
<td>(e^-,\beta^+,K,\gamma)</td>
<td>5.50±0.5 hrs.</td>
<td>0.28(e(^-)),31((\beta^+)) 1.1</td>
<td>Pr(p,3n)</td>
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<tr>
<td>(^{76})Os(^{182})</td>
<td>(K)</td>
<td>24.0±1 hrs.</td>
<td></td>
<td>Re(p,4n)</td>
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<tr>
<td>(^{76})Os(^{183})</td>
<td>(e^-,K,\gamma)</td>
<td>12.0±0.5 hrs.</td>
<td>0.15(e(^-)),0.42((\beta^+)) 0.34,1.6</td>
<td>Re(p,3n)</td>
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Table II. Revised values.

| \(^{58}\)Ce\(^{135}\) | 22 hrs. | 0.80 \(\beta^+\) | La(p,5n) |
| \(^{58}\)Ce\(^{137}\) | | 0.24 e\(^-\) | La(p,3n) |
I. INTRODUCTION

A major part of the previous work of synthesis and characterization of neutron deficient radioactive isotopes in the rare earth region has been done in this laboratory using the 19-Mev deuterons and the 38-Mev helium ions of the 60-inch cyclotron. Thus the conversion of the 184-inch cyclotron to protons and the availability of the proton linear accelerator for bombardment work offers the possibility of producing new activities by (p,xn) reactions.

The recent development of rapid methods for the separation of rare earths, i.e., ion exchange columns, also made exploration of this region of the periodic table more feasible.

The characterization of radioactive isotopes represents a necessary step in the study of the mode of nuclear reaction and of nuclear structure. The following material represents a desirable addition to the knowledge of nuclear species.

II. GENERAL EXPERIMENTAL

A. Bombardment Methods

1. Targets for the 184-inch cyclotron.-- All the materials bombarded in the 184-inch cyclotron were powders, rare earth oxides, and rhenium metal which is not fusible. The most satisfactory method found for mounting the target material consisted of placing the powder in an envelope of 0.5 mil aluminum foil. The foil was then clamped in a hinged copper target holder designed to fit the probe of the cyclotron. The primary advantage of this method is the speed with which the target holder, which is always quite active, can be opened and then removed from the laboratory.

There was no evidence in any of the bombardments for recoil activities from the aluminum foil. The 112-minute F$^{18}$ formed by $^{18}$O(p,n)F$^{18}$ was not detected in any of the bombardments of oxides, since protons of energies 40 Mev
to 80 Mev give a very low yield of (p,n) products even in heavier elements. Other (p,xn) reactions of oxygen would give isotopes of too short a half-life to be detected.

The energy of the proton beam of the 184-inch cyclotron is accurate only at energies close to the maximum energy of 350 Mev. At the comparatively low energies for these experiments straggling is appreciable and the spread of the proton energy 10 to 20 Mev. For this reason the calculation of cross sections and absolute yields at various energies would be somewhat meaningless. Consideration of relative yields and determination of approximate threshold energies in series of excitation function type bombardments have been found to be much more valuable as a method for making mass assignments.

2. Targets for the proton linear accelerator. -- Target material for the linear accelerator bombardments consisted of powders of the same type as used in the 184-inch cyclotron bombardments and of 10 mil cerium metal.

The low intensity of the linear accelerator beam minimizes heat effects and simplifies the mounting of target material. The powders were placed in a 0.5 mil aluminum foil envelope which was scotch-taped to an aluminum plate with a hole in the center of approximately the same diameter as the beam, one-half inch. The foil was centered in this opening and backed up with another aluminum plate to strengthen the target. The cerium metal was wrapped tightly with aluminum foil to minimize oxidation and mounted similarly.

The 32-Mev proton beam was degraded to energies of 10, 20, and 25 Mev by means of calibrated aluminum absorbers. The energy of the degraded beam was known to ±1 Mev; the effect of the aluminum foil is not included. Thus series of excitation function type bombardments could be done with the linear accelerator.
No recoil activities were detected from the aluminum foil. However, at energies of 10 and 20 Mev, the reaction $^{18}_{\text{O}}(\text{p},\text{n})^{18}_{\text{F}}$ was detected. This difficulty was resolved by using cerium metal which had a minimum of oxide coating.

B. Counting and Radiation Characterization Methods

All samples were mounted on thin glass microscope cover slides to minimize back-scattering except samples from column runs which were counted on 10 mil steel discs of 1-inch diameter. This was done to facilitate the handling of a large number of samples.

End-on type Geiger-Muller counters were used throughout. Argon tubes, filled with 10 cm. argon and 1 cm. ethyl alcohol, which have mica windows of approximately 3 mg/cm$^2$ thickness, were mounted in a lead housing and connected to a scaling circuit of 64. A coincidence correction of 1.2 percent per thousand counts was used. Most of the decay data and all of the absorption data were obtained with argon counters.

To measure some of the short half-lives, tubes filled with chlorine and argon were used with a scaling circuit of 256. The chlorine tube is much more stable and can be used with higher counting samples than the argon tube. Since the counting efficiencies for argon tubes are better known, they were used exclusively for absorption measurements. In the lead absorptions of hard gamma radiation, the amount of lead shielding around the tube was reduced to minimize scattering effects.

A UX$_2$ standard served to detect counter fluctuations. Series of counts extending over days or weeks were corrected to an arbitrary value of the standard.

Particles were assumed to have a counting efficiency of one. For x-rays and gamma-rays of energy 0.2 to 0.5 Mev a value of 0.5 percent is used, and for gammas of higher energy, 1 percent per Mev. The counting efficiency for
L x-rays is determined by their absorption in the gas.

The energies of light rare earth L x-rays are too low to permit detection by these methods, and the K x-rays are appreciably absorbed by aluminum. The ranges of the particles of the light rare earth isotopes were best determined by beryllium absorptions, while those of osmium isotopes were determined by aluminum absorptions coupled with a beryllium point to determine the amount of L x-radiation.

The maximum energies were obtained from the well known formulae, \( E_m = 1.85R + 0.245 \) for \( E_m > 0.8 \) Mev and \( E_m = 1.92 R^{0.725} \) for \( E_m < 0.8 \) Mev. The visual endpoints were compared with the Feather Range. In the case of conversion electrons the effects of counter window, 3 mg/cm\(^2\), and air gap, 1 mg/cm\(^2\) per cm. were added to the observed ranges.

The K x-rays and gamma-rays were analyzed by lead absorptions in which sufficient beryllium to remove all particles from the sample was placed below the lead, and an amount sufficient to absorb all the secondary electrons was placed above. Both L and K x-radiations were corrected for the Auger effect.

The use of a magnetic counter made it possible to determine the signs of the particles and to make an approximate determination of the energies to substantiate the absorption data. Decay of a positron or an electron could also be followed.

C. Chemical Analysis

The spectrographic analyses to determine the purity of target materials and the validity of chemical separations were made by J. Conway and M. Moore of this laboratory.
III. LANTHANUM AND CERIUM ISOTOPES

Lanthanum oxide prepared by Dr. F. H. Spedding served as target material. Spectrographic analysis revealed no other rare earths within the limits of detection, i.e., less than 0.1 percent.

The bombarded material was dissolved in nitric acid and cerous carrier and barium hold-back carrier were added. The material was then subjected to fluoride hydroxide cycles. This consists of precipitating the fluoride from warm nitric acid solution, dissolving the fluoride with 8N nitric acid saturated with boric acid, precipitating the hydroxide with ammonium hydroxide, dissolving in nitric acid, and repeating to obtain sufficient purity. The chemistry is specific for rare earths and yttrium.

To the purified lanthanum and cerium in nitric acid cooled to 0°C. were added first potassium bromate to oxidize the cerium and then iodic acid to precipitate ceric iodate. The coprecipitation of lanthanum is less than 2 percent by spectrographic analysis, and two reprecipitations of the ceric iodate were sufficient to remove those lanthanum activities which did not grow in rapidly.

Separation of the lanthanum daughter activities was affected by precipitation of the fluoride from the ceric solution. Additional purification was obtained by methods similar to the above.

Proton energies of 50, 60, 70, and 80 Mev were used in the lanthanum (La$^{139}$, 99.911 percent abundance) bombardments. The 36-hour Ce$^{137}$ observed by Chubbuck$^1$ was produced in all bombardments, as was Ce$^{135}$, of estimated 16-hour half-life,$^1$ which has been shown to be a positron emitter and the parent of the 19.5-hour La$^{135}$. A 72-hour activity appeared at 60 Mev and in larger amounts at 70 and 80 Mev. At 70 and 80 Mev a 6.3-hour activity was formed. The 72-hour and 6.3-hour periods have been assigned to Ce$^{134}$ and Ce$^{133}$, respectively.
Since this isotope was formed in all bombardments, and since its conversion electron is of comparatively high abundance, it was necessary to verify Chubbuck's measurements. The only discrepancy was in the energy of the electron. Magnetic counter measurements (Fig. 1) and Be absorption data (Fig. 4) both give 0.24 Mev. Decay of the electron through about eight half-lives showed it to be the correct one. A re-examination of Chubbuck's data revealed similar results with an interpretive error on his part.

Ce$^{137}$

This isotope had been identified as a positron emitter and shown to be the parent of the 19.5-hour La$^{135}$. A half-life estimate of 16 hour was made by considering the areas under the curves from positron sweeps on the magnetic counter. An unusually large cross section for formation of Ce$^{137}$ was offered as explanation for the fact that no 16-hour period could be resolved from decay curves of Ce$^{135}$ and Ce$^{137}$. However, a more satisfactory explanation is the very low $\beta^+$/electron-capture branching ratio for Ce$^{135}$ and the fact that the growth of La$^{135}$ gives an apparent slope of about 36 hours for Ce$^{135}$.

The cerium fraction from a lanthanum plus 50-Mev proton bombardment, which contained only Ce$^{135}$ and Ce$^{137}$, was analyzed on the magnetic counter and found to have a 0.81 Mev (HP = 4060 gauss-cm.) positron which decayed with a 22-hour half-life (Figs. 2, 3).

A beryllium absorption gave a range of 300 mg. Be/cm$^2$ by Feather analysis for the Ce$^{135}$ positron, and 60 mg/cm$^2$ = 0.24 Mev for the Ce$^{137}$ conversion electron (Fig. 4). Since an unambiguous resolution of decay curves could not be done, no quantitative analysis of Ce$^{135}$ radiations can be offered except that the positron branching is less than 1 percent.
FIG. I  
\( \text{Ce}^{137} \) CONVERSION  
ELECTRON 0.24 MEV  
\( H_p = 1836 \text{ GAUSS-CM} \)
FIG. 2
MAGNETIC COUNTER
DECAY OF THE 22 HR
Ce 135 POSITRON.

TIME (HOURS)  MU 199
FIG. 3
Ce$^{135}$ POSITRON 0.80 MEV
$H\rho = 4060$ GAUSS - CM
FIG. 4
BERYLLIUM ABSORPTION
OF Ce$^{135}$ AND Ce$^{137}$
A- ELECTROMAGNETIC RADIATION
B- Ce$^{135}$ 0.80 MEV POSITRON
C- Ce$^{137}$ 0.24 MEV CONVERSION ELECTRON

% TRANSMISSION

mg Be/cm$^2$  MU 201
Lanthanum fractions showed only the 19.5-hour La$^{135}$ and the very long La$^{137}$ which confirms the mass assignment.

In all bombardments with protons of 60- to 80-Mev energy a 72.0-hour cerium activity remained after the other cerium activities had decayed out and their daughter activities had decayed or had been removed by chemical means (Fig. 5). Chemical separation of the lanthanum daughter gave a 6.5-min. activity (Fig. 6).

The La$^{133}$ (4 hour) and La$^{135}$ (19.5 hour) mass assignments have been confirmed mass spectrographically. Since La$^{134}$ is an odd-odd type nucleus, and since La$^{136}$ has a half-life of 10 min., a value of approximately 5 min. was anticipated for La$^{134}$. This evidence plus the approximate threshold energy of 60 Mev for the 72.0-hour cerium constitute the basis for the mass assignment of this pair of isotopes.

A beryllium absorption (Fig. 7) of Ce$^{134}$ and La$^{134}$ in equilibrium revealed a particle of range 1340 mg. Be/cm$^2$ = 2.7 Mev. The magnetic counter proved it to be a positron of maximum energy 2.7 Mev; an end-point of 1.06 $\cdot$ 10$^4$ gauss-cm. (Fig. 8), and positron decay through ten half-lives gave a 72.0-hour half-life. Analysis of the electromagnetic radiation by Pb absorption gave only K x-rays and annihilation radiation (Fig. 9). The absorption curve data gave the following ratios for the radiations of the two isotopes in equilibrium:

\[
2.7 \text{ Mev } \beta^+: \text{ K x-rays: } 0.5 \text{ Mev } \gamma = 0.44: 1.56: 0.44.
\]

The L x-rays being of only a few kilovolts energy escape detection by these methods which constitutes a major error in the radiation analysis.

That the La$^{134}$ daughter emitted the positrons was verified by following the decay of a separated La$^{134}$ sample on the magnetic counter. This was also shown by beryllium absorption (Fig. 10) which was done on the chemically separated daughter. Since the necessarily quick chemistry did not remove all the parent,
FIG. 5
GROSS DECAY OF 72.0 HR Ce\(^{134}\)
FIG. 6
GROSS DECAY OF $\text{La}^{134}$
A - $\text{Ce}^{134}$ IMPURITY
B - 6.5 MIN $\text{La}^{134}$
FIG. 7
Be ABSORPTION OF Ce$^{134}$ AND La$^{134}$ IN EQUILIBRIUM
A. ELECTROMAGNETIC RADIATION
B. 2.7 MEV POSITRON
FIG. 8
L0\textsuperscript{134} POSITRON 2.7 MEV
$H_p = 1.06 \times 10^4$ GAUSS-CM
FIG. 9
Pb. ABSORPTION OF
Ce$^{134}$ AND La$^{134}$ IN
EQUILIBRIUM.
A- 0.5 MEV GAMMA
B- K X-RAYS
FIG. 10
Be ABSORPTION OF 6.5 MIN. $^{134}$La
A - ELECTROMAGNETIC RADIATION
B - 2.7 MEV POSITRON

% TRANSMISSION

0 300 600 900 1200 1500 1800 2100
mg Be/cm$^2$ MU 207
the beryllium absorption was repeated on the sample as soon as the 6.5-min. activity decayed out, and a point-by-point resolution of the absorption curve was made. This method was found to give reproducible results in obtaining absorption data for short-lived activities.

The assumption that, since La$^{134}$ is a positron-emitter, it emits all the positrons, and hence the 0.5-Mev gamma-rays, coupled with the data of Figs. 9 and 10, allows the following ratios for the La$^{134}$ radiations to be calculated:

$$2.7 \text{ Mev } \beta^+: \text{ K x-rays: } 0.5 \text{ Mev } \gamma = 0.4: 0.6: 0.4.$$  

From the above data it can be postulated that the 72.0-hour Ce$^{134}$ decays entirely by orbital electron capture, and that the 6.5-min. La$^{134}$ decays 44 percent by positron-emission and 56 percent by electron capture. There are no conversion electrons, and the only gamma is the annihilation radiation.

Ce$^{133}$

In the bombardments with 70- and 80-Mev protons a cerium activity of shorter half-life which emitted a hard gamma was formed. Since Ce$^{134}$, Ce$^{135}$, and Ce$^{137}$ have no gamma radiation of energy greater than 0.8 Mev, an unambiguous half-life determination was possible. The decay of the gamma-radiation through 20 g. of lead yielded a 6.3-hour half-life through nine half-lives with a 36-hour tail from the 0.75-Mev gamma of Ce$^{137}$ (Fig. 11).

The mass number of the 6.3-hour cerium activity was determined by separating its lanthanum daughter. The 4.0-hour La$^{133}$, which has been run on the mass spectrograph, was shown to grow in only during the decay of the 6.3-hour activity which must then be Ce$^{133}$. The approximate threshold of 70 Mev for its formation is also compatible with this assignment.

A satisfactory analysis of the relative abundances of Ce$^{133}$ radiations was not obtained since the data are complicated by the presence of four cerium activities and the growth of their four daughter activities.
FIG. II
DECAY OF Ce$^{137}$ THROUGH
20 g OF Pb.
A - 36 HR Ce$^{137}$
B - 6.3 HR Ce$^{133}$
The decay of positrons on the magnetic counter had two components, the 72.0-hour Ce\textsuperscript{134} and an approximately 6-hour one. The positron spectrum indicated two particles as shown in Fig. 12, where curve A was obtained when both activities were present and curve B is the distribution of the 72.0-hour component only. The estimated end-point of the 6.3-hour positron is \( H \rho = 5680 \text{ gauss-cm.} = 1.3 \text{ Mev.} \)

The energy was confirmed by beryllium absorption which showed a particle of range 600 mg. Be/cm\textsuperscript{2} = 1.3 Mev in addition to the La\textsuperscript{134} positron and the Ce\textsuperscript{137} 0.24-Mev electron. The data also indicate a conversion electron of about the same energy as the Ce\textsuperscript{137} electron, but this was not unambiguously demonstrated.

The above mentioned hard gamma had a half-thickness in lead of 15 g/cm\textsuperscript{2} which corresponds to 1.8 Mev.

Thus the 6.30-hour Ce\textsuperscript{133} decays by orbital electron capture and positron-emission and emits gamma-rays of 1.8-Mev energy.

IV. PRAESEODYMIUM ISOTOPES

Cerium metal of 10-mil thickness prepared by Dr. F. H. Spedding was found to be the most satisfactory target material for the study of light praeseodymium isotopes. Traces of calcium and iron were the only impurities detected by spectrographic analysis. An air tight wrapping of aluminum foil minimized oxidation so that the reaction \( ^{18}\text{F}(p,n)^{18}\text{F} \) was not detectable, as it was with the oxide.

The bombarded metal was dissolved in dilute hydrochloric acid, lanthanum carrier for the praeseodymium was added, and then the hydroxides were precipitated with ammonium hydroxide. After dissolving these with concentrated nitric acid and adding potassium bromate, the bulk of the cerium was precipitated as ceric iodate. The lanthanum carrier was then subjected to fluoride hydroxide cycles, and the final step of purification consisted of adding inactive cerium and precipitating to remove the remaining traces of active cerium.
FIG. 12
A - Ce\textsuperscript{133} AND La\textsuperscript{134} POSITRONS
B - La\textsuperscript{134} POSITRON
The cerium bombardments were made on the linear accelerator using 10, 20, and 32-Mev protons for $(p,n)$, $(p,2n)$, and $(p,3n)$ reactions. Even though cerium has an even atomic number, its isotopic abundances are suitable. The relative abundances are $\text{Ce}^{142}$, 11.07; $\text{Ce}^{140}$, 88.48; $\text{Ce}^{138}$, 0.250; $\text{Ce}^{136}$, 0.193. The contributions of the last two are negligible, and $(p,n)$ on $\text{Ce}^{142}$ gives the well known 19.5-hour $\text{Pr}^{142}$. $(p,2n)$ gives stable $\text{Pr}^{141}$. Thus the reactions of the abundant $\text{Ce}^{140}$ can be used to study the neutron deficient praeodymiums.

At 10 Mev the 19.5-hour $\text{Pr}^{142}$ and a 3.5-min. period were observed. The 3.5-min. half-life has previously been reported for $\text{Pr}^{140}$. At 20 Mev a 4.50-hour, the 19.5-hour, and the 3.5-min. activities were formed. A 120-min. half-life was seen at 32 Mev only. The 4.50-hour and the 120-min. activities have been assigned to $\text{Pr}^{139}$ and $\text{Pr}^{138}$, respectively.

$\text{Pr}^{140}$

Pool and Quill reported a 3.5-min. positron-emitter from the reaction $\text{Pr}^{141}(n,2n)\text{Pr}^{140}$. Since a short $\text{Pr}^{139}$ was suspected from the decay of $\text{Nd}^{139}$, it was necessary to verify this value. Cerium plus 10-Mev protons gave a 3.5-min. half-life through many half-lives as did cerium plus 20-Mev protons.

$\text{Pr}^{139}$

The 4.50-hour praeodymium activity (Fig. 13) was formed in bombardments of 20- and 32-Mev protons on cerium but did not appear at 10 Mev. It was thus assigned to $\text{Pr}^{139}$ as the product of a $(p,2n)$ reaction on $\text{Ce}^{140}$.

Confirmation of the mass assignment was made by showing it to be the parent of the 140-d. $\text{Ce}^{139}$. The proton beam of the linear accelerator is much too small to produce sufficient 4.50-hour activity to determine the growth curve of a 140-d. activity. A solution containing mainly the 4.50-hour $\text{Pr}^{139}$ and a very small amount of the 19.5-hour $\text{Pr}^{142}$ was purified and then allowed to decay for
FIG. 13
ELECTROMAGNETIC DECAY
OF 4.50 HR. Pr $^{139}$

TIME (HOURS)

ACTIVITY

MU 210

0 4 8 12 16 20 24
approximately 20 hours. A cerium fraction was then precipitated in which the only cerium activity appearing was Ce$^{139}$, which must necessarily have grown in from the 4.50-hour activity.

A beryllium absorption, (Fig. 14), showed one particle of range 400 mg. Be/cm$^2$ which corresponds to 1.0 Mev. The Feather range was found to be the same. This was verified by a magnetic counter sweep which gave a positron of maximum energy 1.0 Mev = 4750 gauss-cm. (Fig. 15).

In addition to the expected K x-rays and annihilation radiation, a 1.0-Mev gamma-ray appeared in the lead absorption (Fig. 16).

From absorption data, the relative abundances of these radiations are as follows: 1.0-Mev $\beta^+$: K x-rays: 0.5-Mev $\gamma$: 1.0-Mev $\gamma$ = 0.06: 1: 0.06: 0.04. Since no conversion electrons were observed, it can be assumed that each quantum of K x-radiation represents one disintegration by orbital electron capture. And from the above data it can be postulated that Pr$^{139}$ decays mainly by orbital electron capture with approximately 6 percent positron branching.

Pr$^{138}$

In the bombardments with 32-Mev protons a 120-min. praseodymium activity appeared in higher yield than the 4.50-hour Pr$^{139}$. On the basis of the approximate threshold of 30 Mev it was allocated to Pr$^{138}$ as the product of a $(p,3n)$ reaction on Ce$^{140}$.

Decay through 16 g. of lead of the abundant hard gamma-ray allowed the 120-min. activity to be observed through six half-lives before the amount of Pr$^{139}$ 1.0-Mev gamma became significant (Fig. 17).

This isotope emits both a positron and a conversion electron as shown by beryllium absorption (Fig. 18), and the magnetic counter, (Fig. 19). The range of the positron is 625 mg. Be/cm$^2$, which was verified by Feather analysis, and the visual end-point of the magnetic counter sweep is 6140 gauss-cm., both of
FIG. 14
Be ABSORPTION OF 4.50 HR. Pb $^{138}$

A - ELECTROMAGNETIC RADIATION

B - 1.0 MEV POSITRON

% TRANSMISSION vs mg Be/cm$^2$
FIG. 15

Pr\textsuperscript{139}, 1.0 MEV POSITRON
\(\mathcal{H} = 4750 \text{ GAUSS-CM}\)

\begin{center}
\begin{tabular}{|c|c|c|}
\hline
162 & 1300 & 2680 \\
\hline
366 & 4060 & 5440 \\
\hline
335 & 481 & 212 \\
\hline
\end{tabular}
\end{center}
FIG. 16
Pb ABSORPTION OF 4.50HR Pr$^{139}$

A - 1.0 MEV GAMMA
B - 0.5 MEV GAMMA
C - K X-RAYS

% TRANSMISSION

g Pb/cm$^2$  0  5  10  15  20  25  30  35

MU 213
FIG. 17

120 MIN. Pr\textsuperscript{138} DECAY THROUGH 16 g OF LEAD

TIME (HOURS) MU 214
FIG. 10

Be ABSORPTION OF 120 MIN $^{39}$P

A - ELECTROMAGNETIC RADIATION
B - 1.4 MEV POSITRON
C - 0.22 MEV CONVERSION ELECTRON
FIG. 19

A - \(^{138}\)Pr \(0.22\) MEV CONVERSION ELECTRON
\[ H_p = 1740\ \text{GAUSS - CM} \]

B - \(^{139}\)Pr \(1.4\) MEV POSITRON
\[ H_p = 6140\ \text{GAUSS - CM} \]
which correspond to a maximum energy of 1.4 Mev. The values 50 mg. Be/cm$^2$ and 1740 gauss-cm. show the energy of the conversion electron to be 0.22 Mev.

Gamma-rays of half-thicknesses 0.275, 4.5, and 12.0 g. Pb/cm$^2$, corresponding to 0.16, 0.5, and 1.3 Mev, and K x-radiation appeared in the lead absorption (Fig. 20). From absorption data the relative abundances of the radiations are as follows: 0.22-Mev e$^-$: 1.4-Mev $\beta^+$; K x-rays: 0.16-Mev $\gamma$: 0.5-Mev $\gamma$: 1.3-Mev $\gamma = 0.04$: 0.14: 1: 0.22: 0.36: 0.75.

If it is assumed that 4 percent of the K x-radiation arises from the process of conversion, an approximate branching ratio of positron-emission to electron capture of 0.13: 1 is obtained for the decay of Pr$^{138}$. The high abundance of the 0.5-Mev gamma-radiation indicates a gamma of approximately 0.5 Mev in addition to the annihilation radiation.

V. NEODYMIUM ISOTOPES

The target material for these experiments was praseodymium oxide from Johnson Matthey Co., sample no. 547 which had been further purified on an ion exchange column by Mr. R. C. Lilly of this laboratory. No impurities were detected by spectrographic analysis.

The bombarded material was dissolved in nitric acid and a pure rare earth fraction was obtained by fluoride hydroxide cycles.

The neodymium activities were separated from the praseodymium target material on an ion-exchange column.

Protons of energies 40 and 50 Mev gave a 22-min. activity, a 5.50-hour one, and the 3.3-d. Nd$^{140}$ reported by Hicks and Wilkinson. The 145-min. Nd$^{141}$ was not observed at these energies. The 5.50-hour Nd was shown to be the grandparent of the 140-d. Ce$^{139}$ and thus is Nd$^{139}$. The 22-min. period is tentatively assigned to Nd$^{138}$ since its yield relative to Nd$^{139}$ is greater at 50 Mev than at 40 Mev.
FIG. 20
Pb ABSORPTION OF
120 MIN. Pr\textsuperscript{139}
A: 1.3 MEV GAMMA
B: 0.5 MEV GAMMA
C: 0.16 MEV GAMMA
D: K X-RAYS

% TRANSMISSION

\[ \text{g Pb/cm}^2 \]
The 5.50-hour activity formed by bombarding praeoseodymium with 40- and 50-Mev protons was found to emit hard gamma-radiation of greater relative abundance than the 3.3-day Nd$^{140}$ which was also formed. This made possible a good half-life determination by following decay through 16 g. of lead (Fig. 21).

The chemical identification was made by means of an ion exchange column. The target material, after being purified by fluoride hydroxide cycles and dissolved in dilute hydrochloric acid, was warmed with a small amount of resin for about five minutes, after which time it was placed on the resin in the column and eluted with citrate.

Since the elution of light rare earths is quite slow, a short column was necessary in the identification of a 5.50-hour activity. A length of 4 cm. and a cross section of 0.317 cm$^2$ for the resin, which was the ammonium form of Dowex-50 spheres, an aromatic hydrocarbon polymer containing nuclear sulfonic acid groups as the only active exchange groups, gave satisfactory separation. The flow rate was 0.06 ml. min.$^{-1}$ cm.$^{-2}$, and the eluting agent was 0.5M citric acid to which sufficient ammonium hydroxide was added to obtain a pH of 3.15.

The elution curve is shown in Fig. 22. Curve A shows the position of the target material and was determined by spectrographic analysis. Curve B represents the elution of the activity. It is not an accurate representation of the shape of the curve since it was not corrected for decay. However, it is an accurate representation of the position of the peak since decay of samples taken at the peak and on both sides of it when extrapolated to an arbitrary time showed a constant ratio of 5.50-hour activity to 3.3-d. Nd$^{140}$. This proves that the 5.50-hour activity is neodymium.

The broad shoulder of Curve B under Curve A is in part the 4.50-hour Pr$^{139}$ daughter, but the elution time is too long relative to the time for growth and
FIG. 21
Nd$^{159}$ DECAY THROUGH 16 g OF LEAD
A - 3.3 D Nd$^{140}$
B - 5.5 HR. Nd$^{139}$
FIG. 22
COLUMN SEPARATION OF Nd$^{139}$
A - AMOUNT Pr BY SPECTROGRAPHIC ANALYSIS
B - ACTIVITY

HOURS AFTER BEGINNING OF RUN
MU 219
decay to detect any distinct separation with the similarity of half-lives. After many hours the 140-d. Ce\textsuperscript{139} granddaughter appeared.

The mass number was confirmed by separating Ce\textsuperscript{139} from an approximately equilibrium mixture of Nd\textsuperscript{139} and Pr\textsuperscript{139}. The 140-day Ce\textsuperscript{139} was found to grow in with a 5.50-hour half-life (Fig. 23).

In the analysis of the Nd\textsuperscript{139} radiations an uncertainty was introduced by the presence of a small but unknown amount of Pr\textsuperscript{139} daughter. In short bombardments a \((p,np)\) reaction to give the 3.5-min. Pr\textsuperscript{140} was detected at 50 Mev but not at 40 Mev. From this it was assumed that the amount of \((p,pn)\) reaction to give Pr\textsuperscript{139} was negligible at 40 Mev. The absorption data were obtained as soon after bombardment as possible when a minimum of Pr\textsuperscript{139} was present.

Beryllium absorption showed two particles of ranges 70 and 1560 mg. Be/cm\(^2\) corresponding to 0.28 and 3.1 Mev, respectively (Fig. 24). These were shown to be a conversion electron and a positron on the magnetic counter (Fig. 25), and the energies were verified, for the conversion electron, \(H\varphi = 2010\) gauss-cm. = 0.28 Mev, and for the positron, \(H\varphi = 1.19 \cdot 10^4\) = 3.1 Mev. The sweeps were taken a number of hours after bombardment and the curious shape of Curve B results from the 1.0-Mev Pr\textsuperscript{139} positron which had grown in, the presence of which in no way affects the determination of the Nd\textsuperscript{139} positron end-point.

In addition to the K x-rays and the annihilation radiation, a gamma-ray of half-thickness 12.5 g. Pb/cm\(^2\) = 1.3 Mev was detected by lead absorption (Fig. 26).

The relative abundances of these radiations, subject to the uncertainty introduced by daughter growth, are as follows: 0.28-Mev \(e^-\), 3.1-Mev \(\beta^+\), K x-rays: 0.5-Mev \(\gamma\), 1.3-Mev \(\gamma = 0.03: 0.11: 1: 0.11: 0.10\).

From these ratios it can be postulated that Nd\textsuperscript{139} decays mainly by electron capture, and, assuming that each K x-ray quantum represents one disintegration by electron capture, the positron branching is approximately 10 percent.
FIG. 23
GROWTH OF 140d Ce\(^{139}\)
FROM 5.50 HR Nd\(^{139}\)

ACTIVITY

TIME (HOURS) MU 220

14823-1
FIG. 24
Be ABORPTION OF 5.50 HR. Nd$^{159}$
A - ELECTROMAGNETIC RADIATION
B - 3.1 MEV POSITRON
C - 0.28 MEV CONVERSION ELECTRON
**FIG. 25**

A - Nd$^{139}$ CONVERSION ELECTRON  
0.28 MEV $H_p = 2010$ GAUSS-CM

B - Nd$^{139}$ POSITRON  
3.1 MEV  
$H_p = 1.19 \times 10^4$ GAUSS-CM
FIG. 26
Pb ABSORPTION OF
5.50 HR Nd$^{139}$
A - 1.3 MEV GAMMA
B - 0.5 MEV GAMMA
C - K X-RAYS
A 22-min. activity was formed in the bombardments of praeodymium with 40- and 50-Mev protons (Fig. 27), and appeared in greater yield relative to $\text{Nd}^{139}$ at 50 Mev. It was shown to be a rare earth by fluoride hydroxide cycles, and, since $\text{Nd}^{139}$, $\text{Pr}^{139}$, and $\text{Pr}^{138}$ have been identified, the 22-min. activity is tentatively assigned to $\text{Nd}^{138}$.

A beryllium absorption was taken of the 22-min. activity, and, after it was gone, the absorption was repeated on the $\text{Nd}^{139}$. A point by point resolution shows a particle of approximate range 1160 mg. Re/cm$^2$ corresponding to a maximum energy of 2.4 Mev (Fig. 28). It undoubtedly is a positron, and the shape of the curve indicates that there may also be a conversion electron.

VI. OSMIUM ISOTOPES

Rhenium metal powder prepared by Prof. A. D. Melavan served as target material for these experiments. The spectrographic analysis which accompanied the sample indicated a trace of silver as impurity. The chemistry employed sufficed to exclude errors from the silver impurity.

The bombarded rhenium was placed in a small distilling flask and osmium carrier added. The metal was dissolved with nitric acid; osmium tetraoxide was distilled in a stream of air and collected in 6N NaOH as sodium osmate. Both osmium and rhenium were precipitated as the sulfides by bubbling hydrogen sulfide through warm acidified solutions of each to which hydroxylamine hydrochloride had been previously added to reduce the nitric acid. Similar chemistry was used to determine the osmium rhenium parent daughter relationships.

Bombardments of rhenium ($\text{Re}^{185}$, 37.07 percent; $\text{Re}^{187}$, 62.93 percent) with 25-Mev protons in the linear accelerator produced the known 97-d. $\text{Os}^{185}$ and a 12.0-hour osmium activity which was shown to be the parent of the 120-d. $\text{Re}^{183}$. With 40-Mev protons in the 184-inch cyclotron an additional 24.0-hour activity
FIG. 27
GROSS DECAY OF Nd$^{138}$
A - 5.50 HR. Nd$^{139}$
B - 22 MIN. Nd$^{138}$
FIG. 28
Be ABSORPTION OF 22 MIN Nd $^{138}$
A - ELECTROMAGNETIC RADIATION
B - 2.4 MEV POSITRONS

% TRANSMISSION

0 300 600 900 1200 1500 1800 2100 2400 mg Be/cm$^2$ MU 225
which decayed to the 12.7-hour $^8\text{Re}^{182}$ was formed.

$\text{Os}^{183}$

The decay of the osmium fraction of the 25-Mev bombardment gave only the 12.0-hour activity and the long $\text{Os}^{185}$ (Fig. 29). The purified osmium was allowed to decay through several half-lives and then a rhenium fraction was separated which was found to contain the 120-d. $\text{Re}^{183}$. The 12.0-hour activity is thus assigned to $\text{Os}^{183}$ as the product of a $(p,3n)$ reaction on $\text{Re}^{185}$.

Aluminum absorption showed two electrons of ranges 27 mg. Al/cm$^2 = 0.15$ Mev and 115 mg. Al/cm$^2 = 0.42$ Mev (Fig. 30). The presence of two conversion electrons was confirmed by the magnetic counter and no positrons were observed. Gamma-rays of energy 0.34 Mev (2.2 g. Pb/cm$^2$) and 1.6 Mev (14 g. Pb/cm$^2$) appeared in the lead absorption (Fig. 31).

The relative abundances of the radiations of $\text{Os}^{183}$, which decays wholly by electron capture, are as follows:

- 0.15-Mev $e^-$: 0.42-Mev $e^-$: L x-rays: K x-rays: 0.34-Mev $\gamma$: 1.6-Mev $\gamma = 0.18$: 0.009: 0.53: 1: 0.18: 0.10.

$\text{Os}^{182}$

The additional osmium activity formed at 40 Mev was found to grow the 12.7-hour $\text{Re}^{182}$ in a series of milking experiments which extended over a week, and was assigned to $\text{Os}^{182}$.

The half-life determination was complicated by the decay of 12.0-hour $\text{Os}^{183}$ and the growth of the 12.7-hour $\text{Re}^{182}$, but all decay data ultimately gave a value of 24 hours. Fig. 32 is a representative curve, and depicts the decay of an osmium sample purified 30 hours after bombardment at which time 42 percent of the original $\text{Os}^{182}$ and 18 percent of the $\text{Os}^{183}$ remain. The first portion of the curve shows the growth of $\text{Re}^{182}$ which is partially compensated by the decay of $\text{Os}^{183}$. 
Fig. 29
Gross Decay of Os$^{183}$
A - 970. Os$^{185}$ and 1200. Re$^{183}$
B - 12.0 hr. Os$^{183}$
FIG. 30
AI ABSORPTION OF 12.0HR Os$^{183}$
A- ELECTROMAGNETIC RADIATION
B- L X-RAYS
C- 0.42 MEV CONVERSION ELECTRON
D- 0.15 MEV CONVERSION ELECTRON
FIG. 31
Pb ABSORPTION OF 12.0HR Os
A - 1.6 MEV GAMMA
B - 0.34 MEV GAMMA
C - K X-RAYS
FIG. 32
GROSS DECAY OF Os\(^{182}\)
A - 97 D. Os\(^{185}\)
B - 24.0 HR. Os\(^{182}\)
C - GROWTH OF Re\(^{182}\)
Decay of Os\textsuperscript{182} is by electron capture, no positrons having been detected on the magnetic counter. Absorption data is ambiguous and indicates that, if Os\textsuperscript{182} emits conversion electrons or gamma-rays, the energies are similar to those of Os\textsuperscript{183}.

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VIII. REFERENCES
