THE
PHYSICAL REVIEW
A Journal of Experimental and Theoretical Physics Established by E. L. Nichols in 1893
VOL. 53, NO. 6 MARCH 15, 1938 SECOND SERIES

Radioactivity Induced in the Rare Earth Elements by Fast Neutrons

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(Received January 15, 1938)

With the exception of illinium, thulium and lutecium all the rare earth elements have been bombarded with slow and fast neutrons. The fast neutrons were obtained from the Li+D reaction. Deuterons of 6.3 Mev were produced by the cyclotron at the University of Michigan. The following periods for electron emission have been observed: La^{140} (31 hr.), Pr^{142} (19 hr.), Nd^{147} (84 hr.), Nd^{149} (2.0 hr.), Nd^{151} (21 min.), Il^{144} (12.5 hr.), Sm^{151} (21 min.), Sm^{153} (46 hr.), Eu^{152} (9.2 hr.), Tb^{160} (3.3 hr.), Dy^{165} (2.5 hr.), Ho^{164} (47 min.), Ho^{166} (30 hr.), Er^{169} (12 hr.) and Er^{171} (5.1 hr.) The following periods for positron emission have been observed: Ce^{139} (2.1 min.), Pr^{140} (3.5 min.), Eu^{150} (27 hr.), Tb^{158} (3.6 min.), Dy^{159} (2.2 min.) and Er^{165}(1.1 min.). There seems to be at least two isotopes which are isomeric: one is Gd^{159} (3.5 min., 17 hr.) and the other is Yb^{175} (2.1 hr., 14 hr.). From radioactivity data it is necessary to postulate the existence of four new stable isotopes: Gd^{152}, Dy^{160}, Er^{164} and Yb^{170}. With the fast neutron bombardment seventeen n-$2n$ type of reactions have been found.*

INTRODUCTION

THE initial work of Fermi[1] and of Amaldi *et al.*[2] in 1934 showed that certain of the rare earth elements could be made weakly radioactive by neutrons from a radon-beryllium source. These workers and others extended the study and in 1936 Hevesy and Levi summarized all such findings.[3] Since 1936 very few new data have been added.

This lack of additional work is probably due to the unusual number of difficulties encountered with this group of elements. Many of the elements can be had only in small quantities. The task of chemically separating one of the rare earth elements from others of the same group is in general long and tedious since their chemical reactions are so nearly identical that fractional separation is usually necessary. Inability to separate rapidly one rare earth from its neighbors obviously inhibits the analysis of the radioactive products formed in samples bombarded with neutrons or deuterons.

The influences of the above-named difficulties are seen in the existing induced radioactivity data as compiled by Hevesy and Levi. For many of the elements the data show contradicting periods, an extremely weak intensity of the radioactivity and meager evidence for associating a period with a definite nucleus.

Clearly much exacting work with pure and strongly radioactive samples is needed before the induced radioactive properties of the rare earths can be ascertained. The cyclotron, conceded to

* *Note added in proof:* Samples of gadolinium, erbium and ytterbium used in these experiments were sent to Professor A. J. Dempster of the University of Chicago who has kindly analyzed them in his mass spectrograph. He replies that the predicted stable isotopes Gd^{152}, Er^{164}, and Yb^{170} have been found as well as three other new isotopes. With his own dysprosium the existence of stable Dy^{160} is confirmed and in addition another dysprosium isotope is found. The high purity of gadolinium, erbium and ytterbium is also verified. Details will be published in a paper by Professor Dempster in the near future.

[1] Fermi, Ricerca Scient. **1**, 330 (1934).

[2] Amaldi, Fermi, Rasetti and Segrè, Nuovo Cimento **11**, 442 (1934).

[3] Hevesy and Levi, K. Dansk-Acad. **14**, No. 5 (1936).

produce slow neutrons in much greater abundance than the strongest radon-beryllium source, answers the need of strongly radioactive samples. A number of the rare earths of unusual purity had been prepared by one of us (L.L.Q.). Furthermore, it has recently been pointed out that fast neutrons, ranging in energies up to 20 Mev, induce strong radioactivity in a large number of elements and are of great aid (because of the n-$2n$ process) in assigning radioactive periods to nuclei.[4]

It is the primary purpose of this paper to report the action of fast neutrons on the rare earth group of elements. However, most of the rare earth elements were also irradiated with slow neutrons. In a few cases deuteron bombardment was employed.

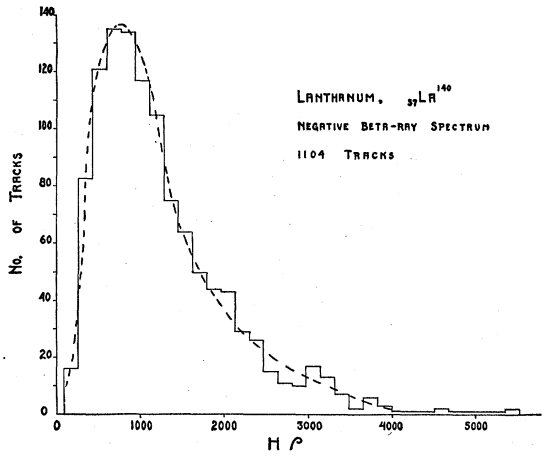


FIG. 1. Beta-ray spectrum of La^{140}. By inspection the upper limit is $4080H$ (0.8 Mev).

EXPERIMENTAL

The fast neutrons were obtained by bombarding a block of metallic lithium with 6.3 Mev deuterons. The lithium was securely fastened to a water cooled copper plate. The material to be irradiated with fast neutrons was placed in an aluminum, copper or glass container and attached to the copper plate. The container, copper plate, and lithium target were in a chamber that could be evacuated.

The slow neutrons were obtained by bombarding metallic beryllium with deuterons. The material to be irradiated with slow neutrons was surrounded by 15 cm or more of paraffin and placed about two feet from the beryllium target.

The deuterons of 6.3 Mev were furnished by the cyclotron at the University of Michigan.

The intensity of the induced radioactivity was measured by a Wulf string electrometer. A six-inch Wilson cloud chamber filled with hydrogen and placed in a magnetic field was used to examine the beta-rays.

RESULTS

Lanthanum

Amaldi *et al.* found lanthanum inactive after strong irradiation under water.[5] However, Marsh and Sugden, using 400 millicuries of radon, found a rather weak activity with a half-life of 46 hours.[6] In the following research lanthanum oxide, La_2O_3, was used. Arc spectrum analysis showed the absence of other rare earths. A bombardment of five to ten microampere-hours with deuterons gave an activity that could easily be followed five half-lives.

This activity, which had a half-life of 31 hours, was found to be associated with the lanthanum fraction in the following series of chemical separations: The bombarded lanthanum was dissolved in dilute hydrochloric acid. To this solution was then added a small amount of barium chloride and cerium chloride solutions. The lanthanum and cerium were precipitated with oxalic acid from the resulting solution as the oxalates. The barium was precipitated from the filtrate as barium sulfate, which was found to be inactive. The lanthanum and cerium oxalates were dissolved in concentrated nitric acid, the resulting solution then being almost neutralized with ammonium hydroxide (just acid to litmus). A small amount of calcium oxide and some potassium bromate were added and the mixture evaporated almost to dryness. The residue was boiled with water, filtered and washed. The cerium remained in the precipitate and the lanthanum was precipitated from the filtrate by oxalic acid. The cerium precipitate was virtually inactive, whereas the lanthanum oxalate precipitate showed the 31-hr. activity.

[4] Pool, Cork and Thornton, Phys. Rev. **51**, 890 (1937); **52**, 239 (1937).

[5] Amaldi, D'Agostino, Fermi, Pontecorvo, Rasetti and Segrè, Proc. Roy. Soc. **149**, 522 (1935).

[6] Marsh and Sugden, Nature **136**, 102 (1935).

The reactions for this activity are,

$$_{57}La^{139}+{}_1d^2\rightarrow{}_{57}La^{140}+{}_1p^1$$
$$_{57}La^{140}\rightarrow{}_{58}Ce^{140}+{}_{-1}e^0 \text{ (31 hr.)}.$$

A histogram based upon the measurement of 1104 negative beta-ray tracks is shown in Fig. 1. By inspection the upper limit is placed at 0.8 Mev. This value of the energy together with the period gives on the Sargent plot a point which falls on the first Sargent curve. The interpretation is that the beta-ray transition is a permitted one.

Fast neutron bombardment of lanthanum also gave the 31-hr. period and in addition a period of 85.6 min. This latter period is undoubtedly due to $_{56}Ba^{139}$ since an 85.6-min. period was obtained by bombarding barium with deuterons and observing the activity in the barium sulphate precipitate.[7] This interpretation is substantiated by Amaldi *et al.*[5] who obtained with slow neutron bombardment of barium a period of 80 min. in the barium chemical separation. The fast neutron reaction equations are,

$$_{57}La^{139}+{}_0n^1\rightarrow{}_{56}Ba^{139}+{}_1p^1$$
$$_{56}Ba^{139}\rightarrow{}_{57}La^{139}+{}_{-1}e^0 \text{ (85.6 min.)}.$$

Cerium

Amaldi *et al.*[5] and Marsh and Sugden[6] found cerium inactive. The question of neutron excited alpha-ray activity in cerium has been discussed by Döpel and by Fünfer.[8]

In the following study the cerium used was the dioxide, CeO_2. Although it was found to be free of other rare earths, the purest available sample had a natural beta- and gamma-activity which was probably due to thorium contamination. The intensity of the activity in a ten gram sample of CeO_2 was about equal to the background of the ionization chamber. Since thorium is strongly activated by fast neutrons, this impurity becomes a serious problem.

In order to compensate as much as possible for this contamination a sample of NH_4OH+Th was made which had the same natural activity as the cerium sample. Both samples were then irradiated with fast neutrons at the same time. The NH_4OH+Th decay curve was subtracted from the cerium decay curve and the resultant decay attributed to the cerium. This procedure gave a 2.1-min. positron period which was about ten times as intense as the induced thorium radioactive background. Since fast neutron bombardment of thorium alone resulted in an induced thorium activity which was totally electron emitting, the carrier of the 2.1-min. positron activity is very probably Ce^{139}. The reaction equations are

$$_{58}Ce^{140}+{}_0n^1\rightarrow{}_{58}Ce^{139}+2{}_0n^1$$
$$_{58}Ce^{139}\rightarrow{}_{57}La^{139}+{}_1e^0 \text{ (2.1 min.)}.$$

Deuteron bombardment of cerium gave longer periods in the cerium chemical separation, but since thorium is also strongly activated by deuterons the results are too confusing for analysis at present.

Praseodymium

Amaldi *et al.*[5] found a 5-min. and a 19-hr. period, the latter being water sensitive. Marsh and Sugden[6] verified the 19-hr. period and attributed the activity to Pr^{142}. However, they were unable to detect the 5-min. period.

In the following study praseodymium oxide, Pr_2O_3, containing about 99 percent praseodymium and 1 percent lanthanum, was used. The presence of lanthanum did not interfere since it is not easily activated.

With fast neutrons a very strong 3.5-min. period was observed which is by far the strongest positron period in the rare earth group of elements. The 19-hr. period was also observed and measured as 18.7 hr. Since praseodymium has only one stable isotope, the reaction equations for the short period may be written:

$$_{59}Pr^{141}+{}_0n^1\rightarrow{}_{59}Pr^{140}+2{}_0n^1$$
$$_{59}Pr^{140}\rightarrow{}_{58}Ce^{140}+{}_1e^0 \text{ (3.5 min.)}.$$

With slow neutron bombardment only the 19-hr. period was observed and the assignment made by Marsh and Sugden is confirmed.

Neodymium

Amaldi *et al.*[5] found a very weak 1-hr. period. Marsh and Sugden[6] were unable to find any induced radioactivity with neutrons from a 400-millicurie radon-beryllium source. With a 500-millicurie source McLennan and Rann[9] observed

[7] Pool and Cork, Phys. Rev. **51**, 1010 (1937).
[8] Döpel, Zeits. f. Physik **99**, 161 (1936); Fünfer, Physik. Zeits. **37**, 693 (1936).
[9] McLennan and Rann, Nature **136**, 831 (1935).

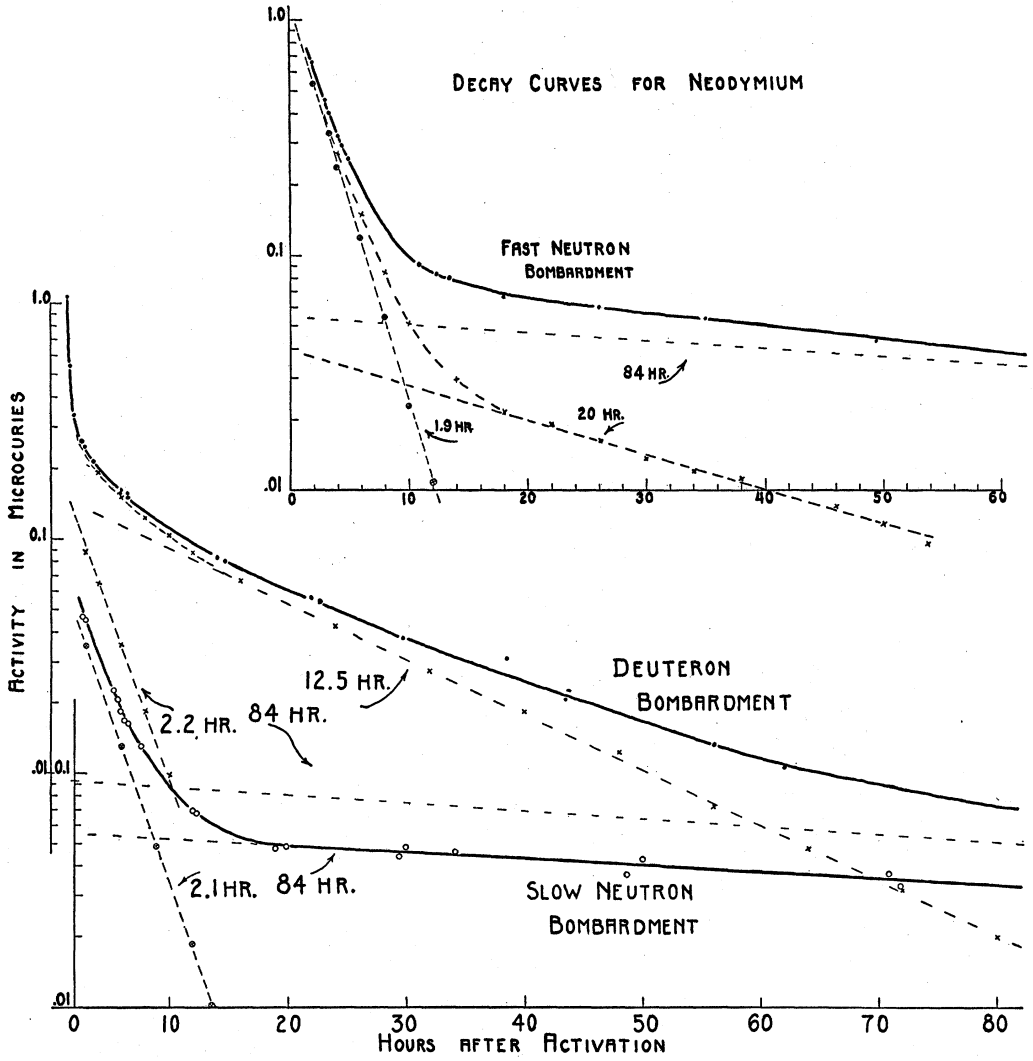


FIG. 2. Decay curves for neodymium. The periods shown are 84 hr. (Sm^{147}), 2 hr. (Sm^{149}), 20 hr. (Pr^{142}) and 12.5 hr. (Il^{144}). The curves extending beyond 80 hr. after activation are not shown. The 21-min. (Sm^{151}) slow neutron period is also not shown.

a 35-min. period which gave an initial intensity of eight impulses per minute in a Geiger counter.

In the following experiments the neodymium oxide, Nd_2O_3, used showed, by arc spectrum analysis, the absence of other rare earths.

The very much greater neutron equivalent of the cyclotron is easily evident from the fact that in four hours of slow neutron bombardment three periods were easily evident, 21 min., 2.0 hr. and 84 hr. They all emitted electrons. The relative rates of formation of the nuclei responsible for these radioactive periods are, respectively, 0.8 : 1 : 1.7. With fast neutron bombardment the observed periods were 2.0 hr., 20 hr. and 84 hr.; the relative rates of formation are 1 : 0.22 : 1.2. The decay curves are shown in Fig. 2.

Since the 21-min. period is produced by slow but not by fast neutron bombardment and since Nd^{150} is the heaviest neodymium isotope,[10] it is reasonable to assign this activity to Nd^{151}.

In order to assign the 2.0-hr. and 84-hr. periods the relative abundance of the stable

[10] Dempster, Phys. Rev. **51**, 289 (1937).

nuclei and the rate of formation of the radioactive nuclei (branching ratio) must be compared. The ratio of the abundance of Nd^{146}/Nd^{148} is 2.4.[11] For slow neutron bombardment the ratio of the rate of formation of the 84-hr. period to the 2-hr. period is 1.7. This ratio for fast neutron bombardment is 1.2 and the ratio of abundance of Nd^{148}/Nd^{150} is 1.15. Consequently, these data suggest, in view of no other guiding information, that the 84-hr. period should be attributed to Nd^{147} and the 2-hr. period to Nd^{149}. The reaction equations would then be

for slow n $\quad {}_{60}Nd^{146}+{}_0n^1\rightarrow{}_{60}Nd^{147}+{}_0\gamma^0$

for fast n $\quad {}_{60}Nd^{148}+{}_0n^1\rightarrow{}_{60}Nd^{147}+2{}_0n^1$

$\qquad {}_{60}Nd^{147}\rightarrow{}_{61}Il^{147}+{}_{-1}e^0$ (84 hr.)

for slow n $\quad {}_{60}Nd^{148}+{}_0n^1\rightarrow{}_{60}Nd^{149}+{}_0\gamma^0$

for fast n $\quad {}_{60}Nd^{150}+{}_0n^1\rightarrow{}_{60}Nd^{149}+2{}_0n^1$

$\qquad {}_{60}Nd^{149}\rightarrow{}_{61}Il^{149}+{}_{-1}e^0$ (2.0 hr.).

The weak 20-hr. period obtained with fast neutron bombardment is best assigned to Pr^{142}

$${}_{60}Nd^{142}+{}_0n^1\rightarrow{}_{59}Pr^{142}+{}_1p^1$$
$${}_{59}Pr^{142}\rightarrow{}_{60}Nd^{142}+{}_{-1}e^0 \text{ (19 hr.).}$$

With deuteron bombardment the 2-hr. and 84-hr. periods were observed as well as a new period of 12.5 hours. The relative rate of formation of the nuclei responsible for these periods are, respectively, 1 : 1.5 : 3.7.

In order to eliminate the effects of sodium, potassium and other contaminations easily activated by deuterons, chemical separation of neodymium oxide was made as follows. The irradiated neodymium oxide was dissolved in warm hydrochloric acid to which was added a small amount of salt. The neodymium was precipitated with oxalic acid. The mixture was cooled to room temperature and filtered. The activity of the precipitated neodymium oxalate was then measured.

Because of the time required for the chemical separation, periods appreciably shorter than the 2-hr. period could not be observed. The 12.5-hr. period was quite strong and has been followed for six half-lives. However, it is uncertain to which nucleus this period should be assigned. The best assignment seems to be Il^{144} according to the equations

$${}_{60}Nd^{143}+{}_1d^2\rightarrow{}_{61}Il^{144}+{}_0n^1$$
$${}_{61}Il^{144}\rightarrow{}_{62}Sm^{144}+{}_{-1}e^0 \text{ (12.5 hr.).}$$

If a stable samarium isotope should be found at mass number 145 or 146, the assignment of the 12.5-hr. period to Il^{145} or Il^{146} would be equally plausible.

[11] Mattauch and Hauk, Naturwiss. **48**, 780 (1937).

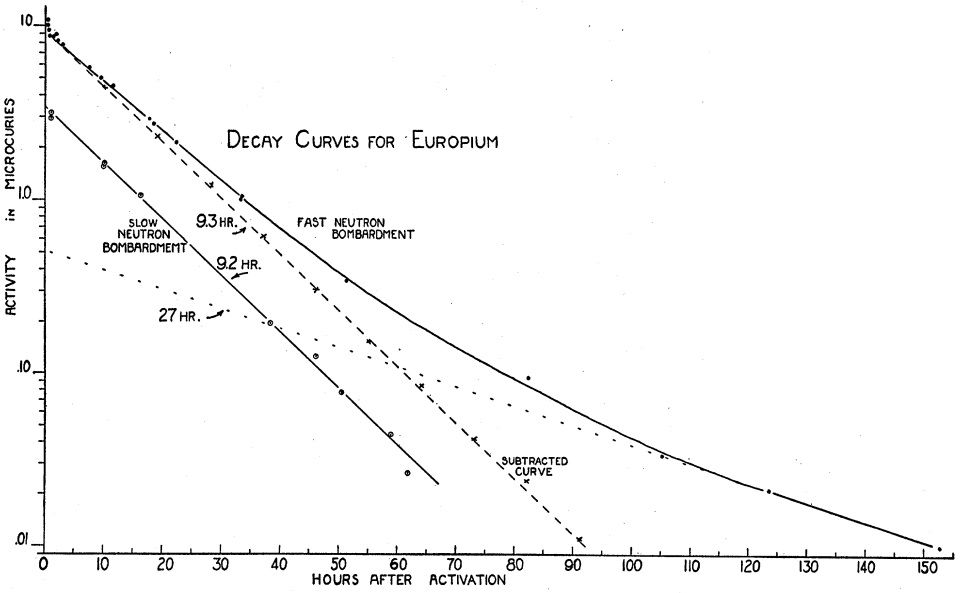


FIG. 3. Decay curves for europium.

Samarium

A 40-min.[5, 6] period and a "longer period" [6] have been reported.

By arc spectrum analysis the samarium oxide, Sm_2O_3, used in the following study showed no other rare earths present.

Fast and slow neutrom bombardment gave the same periods, 21-min. and 46-hr. For slow neutron bombardment the branching ratio of the 46-hr. period to the 21 min. period is 7.7. For fast neutron bombardment this ratio is 1.4. The ratio of the abundance of the stable isotopes Sm^{152}/Sm^{150} is 5.2 and that for Sm^{154}/Sm^{152} is 0.8. These data strongly suggest that the 21-min. period should be assigned to Sm^{151} and the 46-hr. period to Sm^{153}. The reaction equations are

for slow n $_{62}Sm^{150}+{}_0n^1\rightarrow{}_{62}Sm^{151}+{}_0\gamma^0$

for fast n $_{62}Sm^{152}+{}_0n^1\rightarrow{}_{62}Sm^{151}+2{}_0n^1$

$_{62}Sm^{151}\rightarrow{}_{63}Eu^{151}+{}_{-1}e^0$ (21 min.)

for slow n $_{62}Sm^{152}+{}_0n^1\rightarrow{}_{62}Sm^{153}+{}_0\gamma^0$

for fast n $_{62}Sm^{154}+{}_0n^1\rightarrow{}_{62}Sm^{153}+2{}_0n^1$

$_{62}Sm^{153}\rightarrow{}_{63}Eu^{153}+{}_{-1}e^0$ (46 hr.).

Europium

Marsh and Sugden[6] found a strong 9.2-hr. slow neutron period.

Arc spectrum and magnetic susceptibility measurements of the europium oxide, Eu_2O_3, used in the present research showed the europium to be of exceptional purity.

The decay curves for slow and fast neutron bombardment of europium are shown in Fig. 3. The 9.2-hr. period is confirmed. For fast neutron bombardment a weak 27-hr. positron period in addition to the 9.2-hr. period is also present. The 27-hr. period is attributed to Eu^{150} instead of to Il^{148} or Il^{150} primarily because positron activity is not found on the heavy isotope side of an element.

The 9.2-hr. period is assigned to Eu^{152} instead of to Eu^{154} since this period is strongly produced by fast neutron bombardment. The equations are

for slow n $_{63}Eu^{151}+{}_0n^1\rightarrow{}_{63}Eu^{152}+{}_0\gamma^0$

for fast n $_{63}Eu^{153}+{}_0n^1\rightarrow{}_{63}Eu^{152}+2{}_0n^1$

$_{63}Eu^{152}\rightarrow{}_{64}Gd^{152}+{}_{-1}e^0$ (9.2 hr.)

for fast n $_{63}Eu^{151}+{}_0n^1\rightarrow{}_{63}Eu^{150}+2{}_0n^1$

$_{63}Eu^{150}\rightarrow{}_{62}Sm^{150}+{}_1e^0$ (27 hr.).

Since gadolinium is not known to have a stable isotope with mass number 152 and since the 9.2-hr. period emits electrons and is assigned to Eu^{152}, it is necessary to postulate the existence of a stable Gd^{152}. Radioeuropium decays into this new stable isotope as shown in the third equation above.

Gadolinium

Amaldi *et al.*[5] observed an 8-hr. period which was water-sensitive. Marsh and Sugden[6] found gadolinium inactive. With fourteen grams of material McLennan and Rann[9] observed a 6.4-hr. period which gave an initial intensity of sixteen impulses per minute in a Geiger counter.

In this present study gadolinium oxide, Gd_2O_3, was used. High purity was indicated by the absence, in arc spectrum analysis, of any other rare earth elements.

Periods of 3.5-min. and 17-hr. were obtained with both slow and fast neutron bombardment. The branching ratio of these periods for both methods of production is also nearly the same. It is therefore likely that both the 3.5-min. and 17-hr. activity should be assigned to Gd^{159}. This assignment is substantiated by fast neutron bombardment of terbium as indicated below.

Terbium

Marsh and Sugden obtained a weak 3.9-hr. period [6]

The terbium used in the following experiments was prepared by G. Urbain. Slow neutron bombardment gave a 3.3-hr. period which emitted electrons. With fast neutron bombardment a 3.6-min. and 17-hr. period were obtained and the 3.3-hr. period was not evident. About half of the 3.6-min. activity was due to positrons.

Since the 3.6-min. and 17-hr. periods appeared only with fast neutron bombardment of terbium, and with both slow and fast neutron bombardment of gadolinium, support is thereby added to the previous contention that Gd^{159} is isomeric.

The reaction equations for fast neutron bombardment of terbium may be written as follows:

$_{65}Tb^{159}+{}_0n^1\rightarrow{}_{65}Tb^{158}+2{}_0n^1$

$_{65}Tb^{158}\rightarrow{}_{64}Gd^{158}+{}_1e^0$ (3.6 min.)

$_{65}Tb^{159}+{}_0n^1\rightarrow{}_{64}Gd^{159}+{}_1p^1$

$_{64}Gd^{159}\rightarrow{}_{65}Tb^{159}+{}_{-1}e^0$ (3.5 min. 17 hr.).

Since the 3.3-hr. period emits electrons and is assigned to Tb^{160} and since there is no known stable isotope in dysprosium at mass number

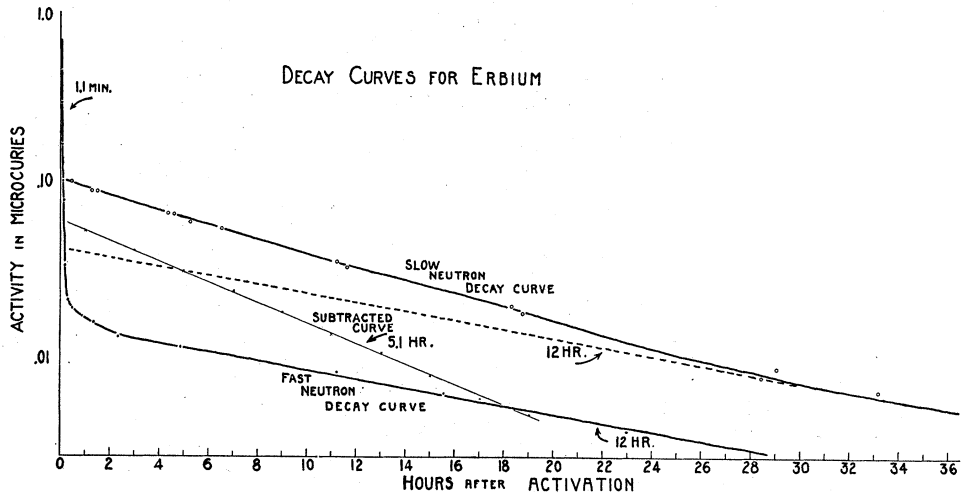


FIG. 4. Decay curves for erbium. The periods shown are 12 hr. (Er^{169}), 5.1 hr. (Er^{171}) and 1.1 min. (Er^{165}). The latter is a very strong positron period.

160, it is necessary to postulate the existence of stable Dy^{160}. Radioterbium then decays into this stable nucleus as follows:

$$_{65}Tb^{159}+{}_0n^1\rightarrow{}_{65}Tb^{160}+{}_0\gamma^0$$
$$_{65}Tb^{160}\rightarrow{}_{66}Dy^{160}+{}_{-1}e^0 \text{ (3.3 hr.)}.$$

Dysprosium

A 2.5-hr. period, the strongest slow neutron period yet known, has been previously found.[6, 12]

For the following experiments Professor Hopkins loaned the authors very pure material that had been used for atomic weight determinations.

With slow neutron bombardment the 2.5-hr. period was verified. With fast neutron bombardment a weak 2.2-min. positron period was found as well as the 2.5-hr. period. This 2.5-hr. period was unusually strong which suggests a stable Dy^{166} isotope. However, the intensity of the radioactivity was not sufficiently strong to force the postulation of the existence of such a stable dysprosium isotope.

The 2.2-min. period is best assigned to Dy^{159} since stable Dy^{160} has been postulated. The reaction equations are as follows:

$$_{66}Dy^{160}+{}_0n^1\rightarrow{}_{66}Dy^{159}+2{}_0n^1$$
$$_{66}Dy^{159}\rightarrow{}_{65}Tb^{159}+{}_1e^0 \text{ (2.2 min.)}.$$

Holmium

Periods of 2.6 hr.,[6, 9] 33 hr.,[3] and 35 hr.[12] have been reported.

For the following experiments Dr. Kremers loaned the authors a holmium-rich yttrium mixture.

Because the holmium contained yttrium the holmium sample and a pure yttrium sample were bombarded at the same time with fast neutrons. The holmium activity was so much stronger than the yttrium activity that no appreciable difficulties were encountered. A 30-hr. and a 47-min. period were observed. The 47-min. period emits electrons and is assigned to Ho^{164}. This assignment necessitates postulating a stable isotope of erbium at mass number 164. The reaction equations for the 47-min. period may be written

$$_{67}Ho^{165}+{}_0n^1\rightarrow{}_{67}Ho^{164}+2{}_0n^1$$
$$_{67}Ho^{164}\rightarrow{}_{68}Er^{164}+{}_{-1}e^0 \text{ (47 min.)}.$$

Erbium

The previous work on this element has resulted in the observation of a variety of radioactive periods, 4.5 min.,[9] 7 min.,[6] 2.9 hr.,[13] 12 hr.,[12] 13 hr.,[3] and 38 hr.[6]

For the following experiments erbium oxide, Er_2O_3, was used. The purity was about 99 percent or better; traces of holmium and yttrium were present.

Slow neutron bombardment gave two weak periods, 5.1 hr. and 12 hr., both electron active. Fast neutron bombardment gave a strong 1.1 min. positron period and a weak 12-hr. period

[12] Hevesy and Levi, Nature **136**, 103 (1935).

[13] Sugden, Nature **135**, 469 (1935).

and a very weak 51-min. period. The 51-min. period is probably due to a slight holmium impurity. The decay curves are shown in Fig. 4. Since the 5.1-hr. period is not present with fast neutron bombardment, it is assigned to Er^{171}. The 12-hr. period, obtained with both fast and slow neutron bombardment, is assigned to Er^{169}. Since the 1.1-min. period is positron active, it is assigned to Er^{165}. The reactions with fast neutrons are as follows:

$$_{68}Er^{170}+{}_0n^1\rightarrow{}_{68}Er^{169}+2{}_0n^1$$
$$_{68}Er^{169}\rightarrow{}_{69}Tu^{169}+{}_{-1}e^0 \text{ (12 hr.)}$$
$$_{68}Er^{166}+{}_0n^1\rightarrow{}_{68}Er^{165}+2{}_0n^1$$
$$_{68}Er^{165}\rightarrow{}_{67}Ho^{165}+{}_1e^0 \text{ (1.1 min.)}.$$

This strong 1.1-min. positron period distinguishes Er^{165} as being the heaviest element in the periodic system yet known to emit positrons. This observation is significant because the probability of K electron capture increases, supposedly, very rapidly with atomic number and as a competitive process in all positron radioactive decay may become even much greater than the probability of positron emission itself.

Thulium

With slow neutron bombardment half-life periods of 3.5 mo.,[12] 4 mo.[14] and 8 mo.[15] have been reported.

Since thulium was not available no additional information can be given in this paper.

Ytterbium

A period of 3.5-hr. has been reported.[6, 12]

In the following experiments ytterbium oxide, Yb_2O_3, was used. It was prepared by the method of Yntema,[16] being precipitated electrolytically as $YbSO_4$. No exact analysis was made but it was probably about 98–99 percent Yb_2O_3 with thulium and lutecium as probable impurities.

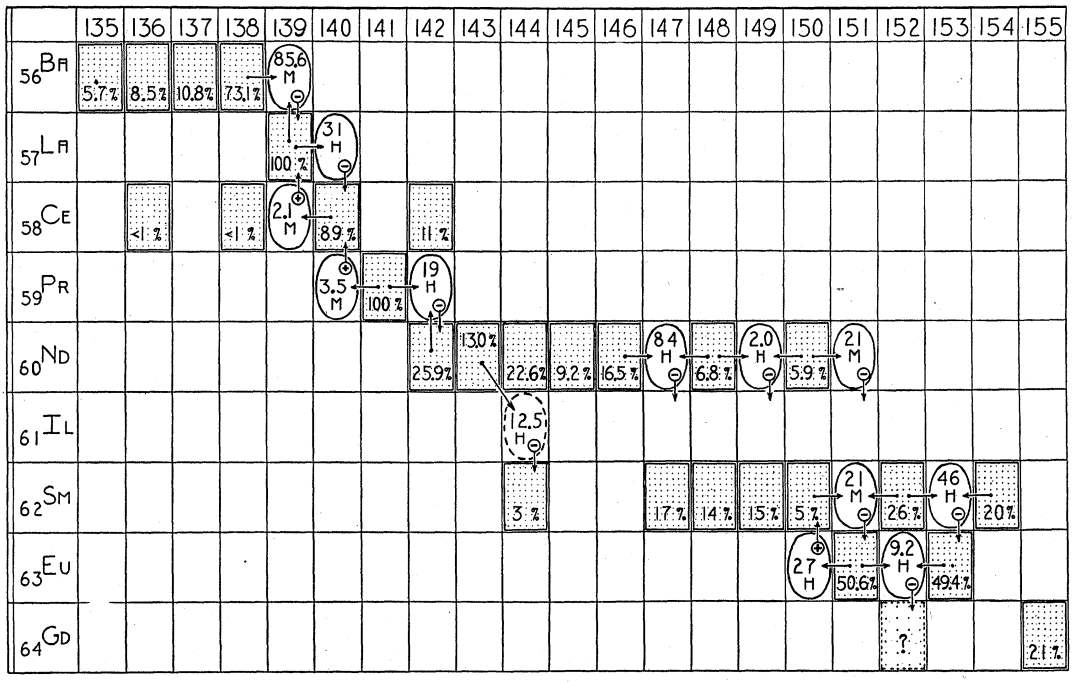
Both fast and slow neutron bombardment gave periods of 2.1 hr. and 41 hr. The branching ratio for these two periods was the same by both methods of activation, even though Yb^{174} is three times as abundant as Yb^{176}. Therefore, both periods are assigned to Yb^{175}. The fast neutron reaction equations are

$$_{70}Yb^{176}+{}_0n^1\rightarrow{}_{70}Yb^{175}+2{}_0n^1$$
$$_{70}Yb^{175}\rightarrow{}_{71}Lu^{175}+{}_{-1}e^0 \text{ (2.1 hr., 41 hr.)}.$$

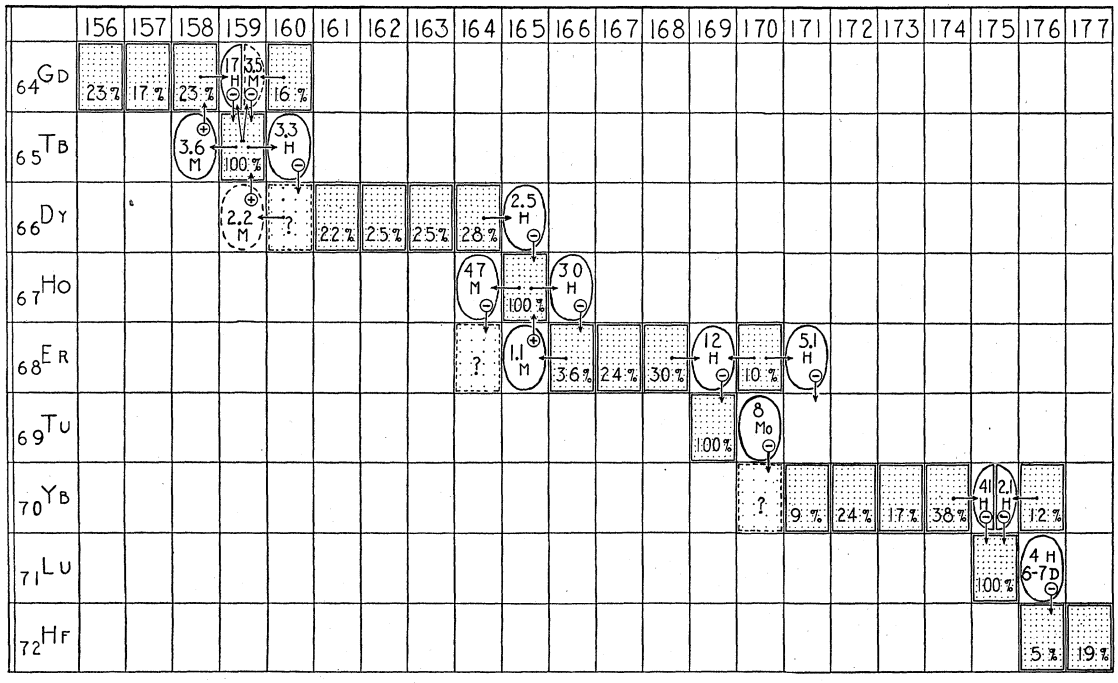
[14] Neuninger and Rona, Wiener Akad. Anzeiger. **73**, 159 (1936).
[15] Curie and Preiswerk, Comptes rendus **203**, 787 (1936).
[16] Yntema, J. Am. Chem. Soc. **52**, 2782 (1930).

TABLE I. *Induced radioactivities in the rare earth elements. Arrangement and notation similar to that used by Livingston and Bethe.*[17]

Raa Element					
Z	A	Class	Raa Particle	Half-life T	Produced by
56	Ba^{139}	A	e^-	85.6 min.	Ba-n-γ, Ba-d-p, La-n-p
57	La^{140}	A	e^-	31 hr.	La-n-γ, La-d-p
58	Ce^{139}	B	e^+	2.1 min.	Ce-n-$2n$
59	Pr^{140}	A	e^+	3.5 min.	Pr-n-$2n$
60	Nd^{147}	A	e^-	84 hr.	Nd-n-γ, Nd-n-$2n$
	Nd^{149}	A	e^-	2.0 hr.	Nd-n-γ, Nd-n-$2n$
	Nd^{151}	A	e^-	21 min.	Nd-n-γ
61	Il^{144}	C	e^-	12.5 hr.	Nd-d-n
62	Sm^{151}	A	e^-	21 min.	Sm-n-γ, Sm-n-$2n$
	Sm^{153}	A	e^-	46 hr.	Sm-n-γ, Sm-n-$2n$
63	Eu^{150}	B	e^+	27 hr.	Eu-n-$2n$
	Eu^{152}	A	e^-	9.2 hr.	Eu-n-γ, Eu-n-$2n$
64	Gd^{159}	B	e^-	3.5 min., 17 hr.	Gd-n-γ, Gd-n-$2n$, Tb-n-p
65	Tb^{158}	B	e^+	3.6 min.	Tb-n-$2n$
	Tb^{160}	A	e^-	3.3 hr.	Tb-n-γ
66	Dy^{159}	B	e^+	2.2 min.	Dy-n-$2n$
	Dy^{165}	A	e^-	2.5 hr.	Dy-n-γ, Dy-d-p
67	Ho^{164}	B	e^-	47 min.	Ho-n-$2n$
	Ho^{166}	A	e^-	30 hr.	Ho-n-γ
68	Er^{165}	A	e^+	1.1 min.	Er-n-$2n$
	Er^{169}	A	e^-	12 hr.	Er-n-γ, Er-n-$2n$
	Er^{171}	A	e^-	5.1 hr.	Er-n-γ
69	Tu^{170}	A	e^-	8 mo.	Tu-n-γ
70	Yb^{175}	A	e^-	2.1 hr., 41 hr.	Yb-n-γ, Yb-n-$2n$
71	Lu^{176}	E	e^-	3.6 hr., 6 da.	Lu-n-γ



(a)



(b)

FIG. 5. Rare earth chart. Stable isotopes are shown by rectangles. Predicted stable isotopes are shown by question mark. Radioactive isotopes are shown by ellipses. Arrows indicate method of production or disintegration. Isomeric nuclei are represented by half-ellipses.

TABLE II. *Rate of formation of radioactive nuclei.*

SUBSTANCE BOMBARDED	TYPE OF BOMBARDMENT	PERIOD T	ASSIGNMENT	TYPE OF REACTION	RELATIVE RATE OF FORMATION
La	fast n	85.6 min. 31 hr.	Ba^{139} La^{140}	n-p n-$2n$	1.0 4.7
Pr	fast n	3.5 min. 19 hr.	Pr^{140} Pr^{142}	n-$2n$ n-γ	3.5 1.0
Nd	slow n	21 min. 2 hr. 84 hr.	Nd^{151} Nd^{149} Nd^{147}	n-γ n-γ n-γ	0.8 1.0 1.7
	fast n	2 hr. 19 hr. 84 hr.	Nd^{149} Pr^{142} Nd^{147}	n-$2n$ n-p n-$2n$	1.0 0.22 1.2
	deuteron	2 hr. 12.5 hr. 84 hr.	Nd^{149} Il^{144} Nd^{147}	d-p d-n d-p	1.0 3.7 1.5
Sm	slow n	21 min. 46 hr.	Sm^{151} Sm^{153}	n-γ n-γ	1.0 7.7
	fast n	21 min. 46 hr.	Sm^{151} Sm^{153}	n-$2n$ n-$2n$	1.0 1.4
Eu	fast n	9.3 hr. 27 hr.	Eu^{154} Eu^{152}	n-$2n$ n-$2n$	6.5 1.0
Gd	slow n	3.5 min. 17 hr.	Gd^{159} Gd^{159}	n-γ n-γ	1.0 3.6
	fast n	3.5 min. 17 hr.	Gd^{159} Gd^{159}	n-$2n$ n-$2n$	1.0 5.7
Tb	fast n	3.6 min. 17 hr.	Tb^{158}, Gd^{159} Gd^{159}	 n-p n-p	 1.0 2.6
Dy	fast n	2.2 min. 2.5 hr.	Dy^{159} Dy^{165}	n-$2n$ n-γ	1.0 24.0
Ho	fast n	47 min. 30 hr.	Ho^{164} Ho^{166}	n-$2n$ n-γ	1.0 3.0
Er	slow n	5.1 hr. 12 hr.	Er^{171} Er^{169}	n-γ n-γ	0.7 1.0
(Ho)	fast n	1.1 min. 51 min. 12 hr.	Er^{165} Ho^{164} Er^{169}	n-$2n$ n-$2n$ n-$2n$	8.0 .043 1.0
Yb	slow n	2.1 hr. 41 hr.	Yb^{175} Yb^{175}	n-γ n-γ	1.0 1.24
	fast n	2.1 hr. 41 hr.	Yb^{175} Yb^{175}	n-$2n$ n-$2n$	1.0 1.27

Lutecium

Radioactive periods of 3.6 hr.,[9] 4 hr.,[6, 3] and 6 to 7 days[3] have been reported.

No observations on lutecium were made in this study.

SUMMARY AND DISCUSSION

Livingston and Bethe[17] have summarized in Table LXIX of their article the available data on induced radioactivities as of July 1, 1937. Following the same plan of presentation and using the same symbols, we have in Table I summarized the data collected in this research on the rare earth group of elements.

Since the relative rate of formation of radioactive nuclei for a given type of bombardment was useful in assigning some of the radioactive periods, Table II shows the relative rates for all the induced radioactive reactions involving two or more periods.

In the rare group of elements the n-$2n$ type of reaction is very prominent in the production of new radioactive periods. Seventeen such reactions were observed. Fig. 5 shows the rare earth region of the periodic table with the stable isotopes represented by rectangular blocks. The radioactive isotopes are represented by ellipses. A dotted ellipse signifies that the assignment is questionable. The isomeric nuclei are represented by half-ellipses.

That Nd^{151} and Sm^{151} both have a 21-min. period is worthy of note. Provided Il^{151} is unstable, which seems quite likely, it is possible that Nd^{151} has a very short life compared to 21 min. It would thus change to Il^{151} which also, in a very short time, would change to Sm^{151}. This samarium isotope which has the real 21 min. period would change to stable Eu^{151}. This proposed series of transformations from Nd^{151} to Eu^{151} involves a triple beta-ray process.

If a stable illinium isotope exists it will have to be isobaric with either neodymium or samarium. However, no examples of stable neighboring isobars in the rare earths are known; the nearest example is Sb and Te. Should no stable illinium isotopes exist at mass numbers 147 and 149, then Nd^{147} (84 hr.) and Nd^{149} (2.0 hr.) would decay by a double beta-ray process into samarium.

ACKNOWLEDGMENTS

The authors are very grateful to Professor J. M. Cork and Dr. R. L. Thornton for the opportunity of one of us (M.L.P.) to work in their laboratory with the cyclotron at the University of Michigan. To Professor B. S. Hopkins of the University of Illinois we give thanks for the loan of a large quantity of very pure dysprosium. Likewise we thank Dr. H. C. Kremers of Cleveland for the loan of the holmium-rich material. Mr. E. Rosenbaum made some of the chemical separations. Mr. B. R. Curtis kindly helped with some of the cloud chamber pictures. One of us (M.L.P.) is very grateful to have been the recipient of the Elizabeth Clay Howald Scholarship for 1936–37.

[17] Livingston and Bethe, Rev. Mod. Phys. 9, 245 (1937).