Radioactive Europium, Gadolinium, and Terbium

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An activity with a half-life of 18.0±0.2 hours and a weaker one of 5.5±0.2 days have been produced by neutron bombardment of gadolinium, resulting in a saturation intensity ratio of 25. A deuteron bombardment of gadolinium gave a ratio of 3 and produced 72-day Tb^{160} with a saturation intensity equal to that of the 5.5 day. The 18.0-hour activity decays by the emission of negative β-particles of 0.85 Mev and a γ-ray of 0.3 Mev. The 5.5-day activity decays by the emission of negative β-particles of 0.5 Mev and γ-rays of 1.28 Mev. Neither the 18.0-hour nor the 5.5-day substance is produced by fast neutron bombardment of Tb, but there is some evidence that both are produced by fast neutron bombardment of dysprosium. In accordance with the above data both the 18.0-hour and the 5.5-day activities are assigned mass 161. An activity with a minimum half-life of 20 years and one with a minimum half-life of 75 days have been produced by a deuteron bombardment of europium. The decay of the 20-year activity is by the emission of β-particles of 1.4 Mev and a γ-ray. The 75-day activity is probably attributable to Gd^{153} and the 20-year activity to Eu^{154}.

I. INTRODUCTION

USING neutrons from a radon beryllium source, several investigations of neutron induced activities in the rare earth elements have been made.[1–4] Later, using cyclotron techniques, experiments were performed using both neutrons and deuterons as bombarding particles.[5–7] Because of the difficulties of obtaining very pure rare earths, there remains some disagreement as to the radioactive isotopes existing in these elements. In addition, certain known activities of the rare earths have not yet been firmly placed because of the difficulty of obtaining chemical separation of these elements from each other.

It is the purpose of this paper to report a series of experiments in the europium, gadolinium, and terbium region which have extended over the past six years. Some of the characteristic radiations associated with the above-mentioned activities are described. Although the experiments give some evidence as to the isotopic location of the activities, exact placing has not been done.

Unless otherwise stated, Hilger materials were used. For all neutron bombardments the sample material was passed directly from the original shipping container to a gelatin capsule. At the conclusion of the bombardment the sample was removed from the capsule for decay readings and the capsule checked for activity.

II. THE 72-DAY ACTIVITY

An activity which decayed with a half-life of 72 days was produced in the rare earth fraction of a deuteron bombardment of gadolinium. Cloud-chamber observations showed that decay occurred by the emission of negative β-particles. By means of aluminum absorption measurements the energy of the β-particle was determined to be 0.71 Mev. Lead absorption measurements indicated that a γ-ray was associated with this decay. The above data are in agreement with the known[8] characteristic radiations of Tb^{160}. This activity is therefore presumed to have been produced by the reaction $Gd^{160}(d,2n)Tb^{160}$.

III. THE 5.5-DAY ACTIVITY

Figure 1 shows the decay of a 5.5-day activity produced by a neutron bombardment of Gd_2O_3.

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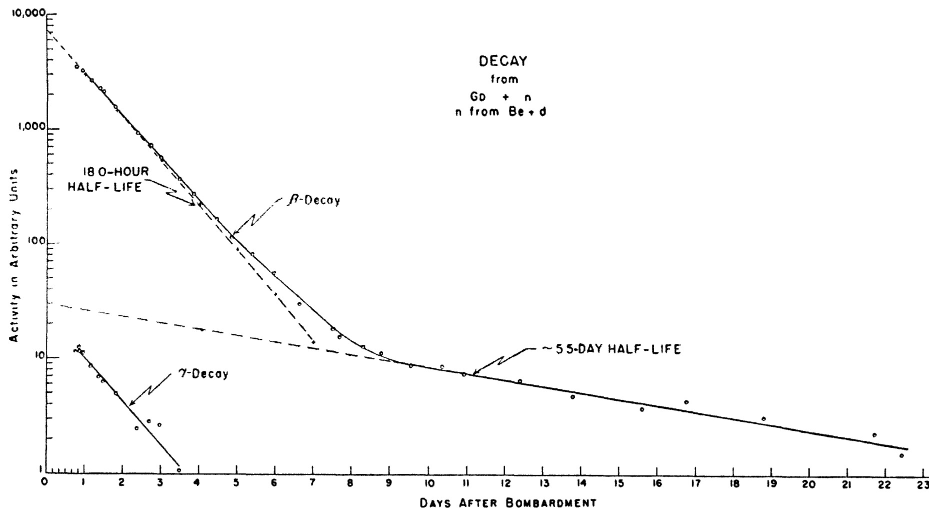

FIG. 1. Decay curve of the β-particle and γ-ray activity induced in Gd_2O_3 by neutrons produced from a deuteron bombardment of beryllium.

The neutron flux was obtained from a deuteron bombardment of beryllium. The beryllium target was made sufficiently thick to completely intercept the deuteron beam, and the Gd_2O_3 was placed directly behind the target. Within experimental error an identical decay curve was obtained using the neutron flux from a deuteron bombardment of lithium.

Figure 2 shows the γ-ray decay of the rare earth activity produced when Gd_2O_3 was bombarded with deuterons. A similar curve was obtained for the β-particle decay. From the lead absorption measurements shown in Fig. 3 the energy of the γ-ray was found to be 1.28 Mev. Cloud-chamber observations showed that decay occurred with the emission of negative β-particles. The energy was determined to be 0.5 Mev by aluminum absorption measurements.

A sample of very pure Tb was exposed for 7.5 hours to an intense neutron flux produced by a deuteron bombardment of lithium. No 5.5-day activity was observed. When Dy was similarly bombarded there was some evidence that this half-life was present in the resulting activity.

IV. THE 18.0-HOUR ACTIVITY

A sample of Gd_2O_3 was placed in the center of a four-foot cube of paraffin with a 500-millicurie radon-beryllium source and allowed to remain for thirty days. No measurable activity was obtained.

When a similar sample enclosed in 5 inches of paraffin was placed near the target section of the cyclotron during a deuteron bombardment of lithium, the induced activity decayed with a half-life of approximately 18 hours.

As shown in Fig. 1 this activity was produced also by a neutron bombardment of Gd_2O_3 when the neutron flux was obtained from a deuteron bombardment of beryllium. A γ-ray of low intensity was found to be associated with this decay. Cloud-chamber observations showed that decay took place by the emission of negative β-particles. By means of aluminum absorption measurements the energy was found to be 0.85 Mev. Within experimental error identical results

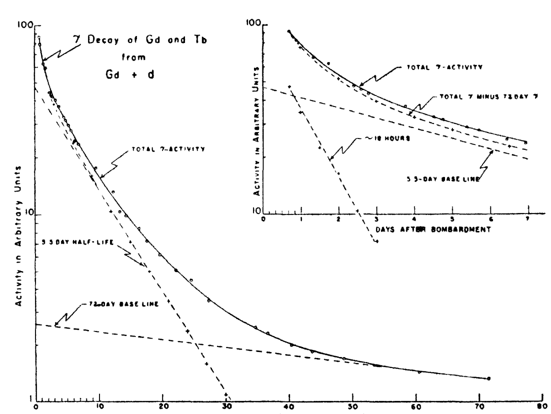

FIG. 2. γ-ray activity induced on the rare earth fraction by a deuteron bombardment of Gd_2O_3.

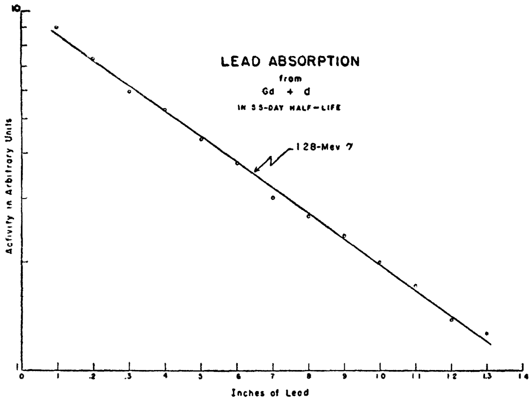

FIG. 3. Lead absorption curve showing 1.28-Mev γ-ray associated with the decay of the 5.5-day activity.

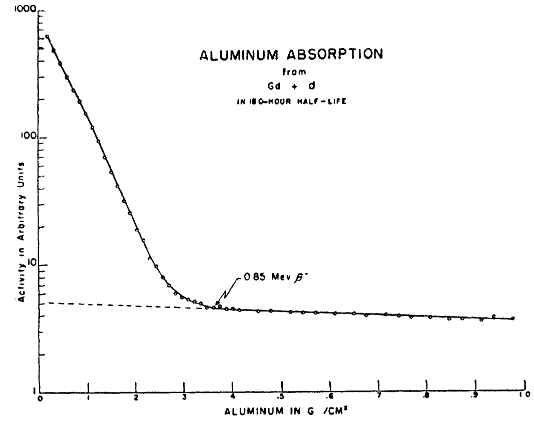

FIG. 5. Aluminum absorption curve showing 0.85-Mev β-particle associated with the decay of the 18.0-hour activity.

were obtained when Gd_2O_3 was bombarded with neutrons produced by a deuteron bombardment of lithium.

This activity was observed in the rare earth fraction of a deuteron bombardment of Gd_2O_3, as shown in the insert of Fig. 2. From the lead absorption measurements shown in Fig. 4 the energy of the γ-ray was found to be 0.3 Mev. No evidence of internal conversion of this γ-ray was found. The energy of the negative β-particle emitted was found to be 0.85 Mev, as shown in Fig. 5.

V. THE 75-DAY AND THE 20-YEAR ACTIVITY

Figure 6 shows the decay of the rare earth fraction of a deuteron bombardment of europium. The 20-year half-life is estimated as the minimum half-life of this activity from the decay observed. The very strong 9.2-hour activity of Eu^{152} present during the early part of decay is not shown. As shown by Fig. 7, aluminum absorption measurements showed that the activity of the 20-year half-life material decayed with the emission of β-particles of 1.4 Mev. By means of lead absorption measurements a γ-ray of approximately 0.9 Mev was found to be associated with this decay. When this activity was subtracted from the total activity curve shown in Fig. 6 an additional activity with a minimum half-life of 75 days was observed.

VI. DISCUSSION

Early investigators found that the use of a radon-beryllium source with as much as 500 millicuries of radon induced very weak activities

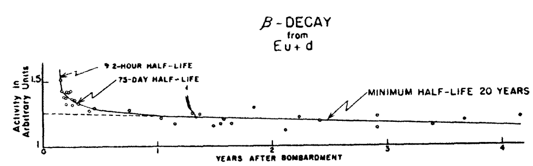

FIG. 6. β-particle decay curve of the rare earth activity induced by a deuteron bombardment of europium. The 75-day half-life was observed when the activity of the 20-year half-life was subtracted from the total β-particle activity shown in this Fig.

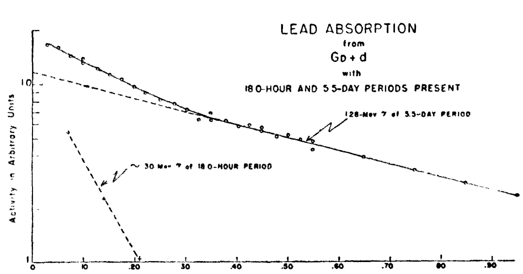

FIG. 4. Lead absorption curve showing 0.3-Mev γ-ray associated with the decay of the 18.0-hour activity.

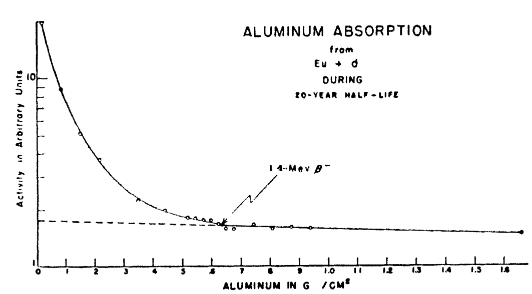

FIG. 7. Aluminum absorption curve showing 1.4-Mev β-particle associated with the decay of the 20-year activity.

in gadolinium. First experimenters using this technique found a weak gadolinium activity which decayed with a half-life of 8 hours.[1] Later, others used the same technique but did not confirm the presence of this half-life in gadolinium. In one case gadolinium was found to be inactive[2] and in another the induced activity was found to be 6.4 ± 0.3 hours.[4] In this latter experiment a 500-millicurie radon-beryllium source of neutrons was used with 14 grams of gadolinium oxalate to obtain an activity of 16 counts/minute with a Geiger counter. The use of cyclotron techniques to obtain greater induced activity in gadolinium also failed to confirm the 8-hour activity.[5] Half-lives of 3.5 minutes and 17 hours were obtained using both slow and fast neutrons. For the experiments described in this paper activities were obtained ranging from the equivalent of 60 counts/sec. to 18,000 counts/sec. on a Geiger counter.

In view of the manner in which neutron bombardments were carried out it is unlikely that contamination could have been added to the original material during preparation for bombardment. The specific purity of the gadolinium used to obtain the activity curves shown is given by Hilger laboratory report No. 11,817. The possibility that any of the periods observed might be due to impurities was further checked by performing each experiment at least twice with sample materials obtained on different shipments. This possibility is eliminated by four factors: the bombarding particle by which the activities were produced, the half-lives observed, the characteristic radiations of the observed activities, and a repetition of saturation intensity ratios over two or three bombardments performed in the same manner.

Since both the 18.0-hour and the 5.5-day activities are produced by slow neutron bombardment of gadolinium, assignment of either to Gd^{153}, Gd^{159}, or Gd^{161} is possible. Assignment of either activity to Gd^{153} is eliminated by the fact that both periods decay with the emission of a negative β-particle. Assignment of either activity to Gd^{159} is unlikely since neither could be produced by a fast neutron bombardment of Tb. Both the 18.0-hour and the 5.5-day activities are therefore assigned mass 161. This assignment is apparently substantiated by the appearance of both activities when dysprosium is bombarded with lithium neutrons. The saturation intensity ratio of the 18.0-hour half-life to the 5.5-day half-life produced by such a neutron bombardment of Gd_2O_3 was found to be 25. The large ratio eliminates the possibility that the 5.5-day activity is a daughter product of the 18.0-hour activity. Assignment of both to Gd^{161} is not supported by the fact that a deuteron bombardment of gadolinium produced the 5.5-day and the 72-day activities with equal intensities. Since the 72-day half-life is known to be produced by a $Gd^{160}(d,2n)Tb^{160}$ reaction, it appears most likely that the 5.5-day activity is produced by a $Gd^{160}(d,n)Tb^{161}$ reaction. Both the 18.0-hour and the 5.5-day activities are therefore tentatively assigned to Tb^{161}.

Fajans and Voigt found Eu^{154} decayed with a half-life of 4–8 years by the emission of a β-particle of 1.0 ± 0.1 Mev. This figure for the half-life of this activity was determined by observations made over a period of 8 months.[7] The half-life determined from Fig. 6 over the first 8 months is approximately 4 years. The fact that two activities are present in this decay is not evident until observations have been made for a number of years. Since the 75-day activity is present with comparatively small intensity, absorption measurements taken while the total activity was decaying with a half-life of 4–8 years would probably show a single "end point." The 20-year activity is therefore assigned Eu^{154}, which decays by the emission of a negative β-particle of 1.4 Mev and a γ-ray of approximately 0.9 Mev to Gd^{154}.

The 75-day activity could be assigned either to Gd^{151} or Gd^{153}. Since 75 days is a longer half-life than would be expected for Gd^{151}, it is tentatively assigned to Gd^{153}.

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