Radioactive Isotopes of the Rare Earths. III. Terbium and Holmium Isotopes

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A study has been made of neutron deficient radioactive isotopes of terbium and holmium produced respectively by α-particle bombardments of europium and terbium, and respectively by proton bombardments of gadolinium and dysprosium, using the 60-inch Crocker Laboratory cyclotron. Five new isotopes of each element have been characterized.

I. INTRODUCTION

THE experimental techniques of bombardments, chemical separations, and radioactivity measurements used in the study of radioactive rare earth isotopes have been previously described.[1] In the study of terbium and holmium activities, europium and terbium oxides were bombarded with α-particles of energy 38, 31, and 19 Mev, while gadolinium and dysprosium oxides were bombarded with 10-Mev protons. With the exception of two short-lived holmium activities, the chemical identity of the isotopes was proved by chemical separation using ion exchange resin columns. In europium bombardments, the target was dissolved and the bulk of the europium first removed by reduction of the boiling $6N$ hydrochloric acid solution with zinc amalgam and precipitation of europium III sulfate by addition of dilute sulfuric acid. A small amount of lanthanum was added to the solution before reduction to act as a holdback carrier for terbium activities. Rare earth activity in the supernatent solution was collected by coprecipitation with ferric hydroxide. The hydroxide was dissolved in dilute hydrochloric acid, the ferric chloride removed by ether extraction and, after adjustment of the acidity to $1N$, the rare earths adsorbed on ion exchange resin for subsequent column separation.

In Table I the production and radiation properties of the terbium and holmium activities are summarized. All of the activities, with the exception of the 34-min. Ho^{164} activity decay with the emission of electromagnetic radiations of half-thickness 7 to 8.5 mg/cm^2 aluminum (6.3 to 6.8 kev), and 60 to 70 mg/cm^2 lead (46 to 48 kev), which correspond to the average energies of L and K x-radiation of gadolinium and dysprosium, respectively. The energies of the electromagnetic radiations were obtained from aluminum and lead absorption measurements; electron energies were obtained from both aluminum absorption measurements and from measurements on a simple beta-ray spectrometer. The latter allowed the decay of positive and negative electrons to be followed separately. In the resolution of aluminum absorption curves, the contribution of soft electromagnetic radiation was obtained by removal of electrons by beryllium absorbers; a correction for absorption of the L x-rays in beryllium was made before resolving the curves. The approximate ratios of the various components of the complex radiations found in the orbital electron capture isotopes studied were obtained from absorption and magnetic spectrometer data. For the standard argon-alcohol counter tubes used, counting efficiencies of 6.5 and 7.5 percent were taken for gadolinium and dysprosium L x-radiation, 0.5 percent for K x-radiation and one percent per Mev for harder gamma-radiation; fluorescence yields of 0.5 and 0.8 were assumed for L and K x-radiation, respectively. Corrections for absorption of soft radiations in counter windows, etc. were made, and backscattering of electrons was minimized by mounting carrier-free, column-separated samples on very thin backing.

II. TERBIUM ISOTOPES

In the bombardment of europium with α-particles, activities of half-lives 5.1 days, 17.2 hr., 190 days, and 5.0 hr. were observed; from the yields in bombardments using α-particles of energies 38, 31, and 19 Mev, allocation was made to masses 153, 154, 155, and 156, respectively. No evidence has been obtained for Tb^{152} which would be formed by α, $3n$ reaction on Eu^{151}, and an upper limit of about 5 min. can be set for the half-life. The 86-day Gd^{153} activity has been observed in the column separated gadolinium fractions from $Eu+\alpha$ bombardments, and the formation of this isotope from the 5.1-day terbium allocated to mass 153 has been confirmed. In the bombardment of gadolinium with 10-Mev protons, the 17.2-hr. activity was observed in very low yield agreeing with its allocation to Tb^{154}; the 5.0-hr. positron emitting activity Tb^{156} and the well-known beta-particle emitting 3.9-hr. and 73.5-day Tb^{160} activities were found in high yields, as would be expected from the abundances of the gadolinium isotopes. A new activity of half-life 4.7 days was found in this bombardment; since the present work was completed, Butement[2] reported a 5.9-day activity which appears to have radiation characteristics agreeing with those of the 4.7-day activity. Since the 4.7-day isotope was found neither in α-particle bombardments of europium nor in fast neutron bombardments of terbium, allocation

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[1] G. Wilkinson and H. G. Hicks, Phys. Rev. 75, 1370 (1949).
[2] D. S. Butement, Phys. Rev. 75, 1276 (1949).

TABLE I. Production and characteristics of terbium and holmium isotopes.

Isotope	Type of radiation	Half-life	Energy of radiation in Mev Particles	γ-rays	Produced by
Tb^{153}	K, e^-, γ	5.1 ± 0.1 days	0.15, $\sim$0.3	0.23, 1.2	$Eu-\alpha-2n$
Tb^{154}	$K, e^-, \gamma, \beta^+(\sim0.5\%)$	17.2 ± 0.2 hr.	0.13, $\sim0.8(e^-)$, $2.6(\beta^+)$	1.3	$Gd-p-n$ $Eu-\alpha, n, 3n$
Tb^{155}	K, e^-, γ	190 ± 5 days	0.1	1.4	$Eu-\alpha-2n$
Tb^{156}	$K, \beta^+(<25\%)$	5.0 ± 0.1 hr.	$\sim$1.3		$Eu-\alpha-n$ $Gd-p-n$
Tb^{157}	K, e^-, γ	4.7 ± 0.1 days	0.09, 0.2	1.4	$Gd-p-n$
Ho^{160}	$K, e^-, \gamma, \beta^+(0.5\%)$	22.5 ± 0.5 min.	$0.17(e^-)$, $\sim1.3(\beta^+)$	$\sim$1.2	$Tb-\alpha-3n$
Ho^{161}	K, e^-, γ	4.6 ± 0.1 hr.	$0.1(e)$	1.1	$Tb-\alpha-2n$ $Dy-p-n$ $Dy-d-n$
Ho^{162}	$K, e^-, \gamma, \beta^-(\sim15\%)$	65.0 ± 0.5 days	$\sim0.1(e^-)$, $0.8(\beta^-)$	$\sim$1	$Tb-\alpha-n$ $Dy-p-n$ $Dy-d-2n$
Ho^{163}	K, e^-, γ	5.2 ± 0.05 days	0.4	0.4 to 0.5 1.4	$Dy-p-n$ $Dy-d-n, 2n$
Ho^{164}	β^-	34.0 ± 0.5 min.	0.95		$Dy-p-n$

to mass 157 seems reasonable. The existence of further long-lived isotopes of terbium of masses 156, 7, or 8 is possible but the presence of long-lived Tb^{155} and Tb^{160} activities in proton-bombarded gadolinium makes recognition of a further activity by simple measurment a difficult task. No difference in the decays of the long-lived activities from the various $Eu+\alpha$ bombardments has been noted yet and all are decaying with the 190-day activity allocated to mass 155 on the basis of reaction yields.

5.1 ± 0.1-Day Tb^{153}

The decay of this activity which was found in high yields in 38- and 31-Mev α-particle bombardments of europium was followed through ten half-lives. The radiations, which differ considerably from those of the 4.7-day activity described below consist of electrons ranges 32 mg/cm^2 (0.15 Mev), and $\sim$90 mg/cm^2 ($\sim$0.3 Mev), and electromagnetic radiations of half-thicknesses 7 mg/cm^2 (6.3 kev) aluminum, 60 mg/cm^2 (46 kev), 570 mg/cm^2 (0.23 Mev), and 11.0 g/cm^2 lead (1.2 Mev). The approximate ratios of various radiations are 0.17 Mev e^-: $\sim$0.3 Mev e^-; L x-ray:K x-ray:0.23-Mev γ-ray:1.2-Mev γ-ray; 0.02:0.001:0.3:1:0.1:0.02. The aluminum and lead absorption curves are shown in Fig. 1.

17.2 ± 0.2-Hr. Tb^{154}

In all bombardments of europium with α-particles, and in low yields in proton bombardments of gadolinium a 17.2-hr. positron emitting activity was observed. The decay of negative and positive electrons and γ-radiation was separated followed through ten half-lives. Using a simple beta-ray spectrometer, the positron of maximum energy 2.6 Mev as well as two groups of negative electrons corresponding to those measured in aluminum absorptions were studied, and the abundances measured by integrating the distribution curves. The positrons are in such low abundance that no annihilation radiation would be observed in absorption measurements. From the various measurements the following ratios were obtained: 0.13-Mev e^-: $\sim$0.8-Mev e^-:2.6-Mev β^+:L x-ray:K x-ray:1.3-Mev γ-ray$=$0.1:0.02:0.004:0.3:1:0.03. It seems reasonable to conclude that the isotope decays at least 98 percent by orbital electron capture to metastable or excited states of the daughter nucleus.

190 ± 5-Day Tb^{155}

This activity was observed in all $Eu+\alpha$ bombardments after decay of the shorter-lived activities. The decay has been followed through three half-lives. The approximate ratios of the radiations were: 0.1-Mev e^-: L x-ray:K x-ray:1.4-Mev γ-ray$=$0.4:0.3:1:0.3; in the calculation of yields it was assumed that $\sim$0.5 of the K x-ray quanta represent one disintegration by orbital electron capture.

5.0 ± 0.1-Hr. Tb^{156}

In bombardments of europium with 19-Mev α-particles, an activity of half-life 5.0 ± 0.1 hr. measured through nine half-lives was observed in high yield. The aluminum absorption showed only a hard electron range 600 mg/cm^2 ($\sim$1.3 Mev), soft electromagnetic radiation 7 mg/cm^2 half-thickness (6.3 kev), and hard quantum radiation background. On a simple beta-ray spectrograph, no negative electrons were observed, but only positrons of maximum energy ~1.4 Mev, which decayed with the 5.0-hr. half-life. Insufficient activity was obtained for measurement of a lead absorption curve of gamma-rays. Assuming 0.5 percent average counting efficiency for gamma-radiation, the ratios of the radiations obtained from aluminum absorptions gives: 1.3-Mev β^+: L x-ray: K x-ray and γ-ray radiation $=\sim 0.2:\sim 0.1:1$.

The isotope thus appears to decay mainly by orbital electron capture with less than 25 percent positron branching.

4.7±0.1-Day Tb^{157}

In the decay of the chemically separated terbium fraction from 10-Mev proton bombardments of gadolinium, an activity of half-life 4.7±0.1 days measured through seven half-lives has been observed. The radiation characteristics obtained by resolution of aluminum and lead absorption curves (Fig. 2) are quite different from those of the 5.1-day activity described above. The latter has been allocated to Tb^{153} on the basis of yields in the α-particle bombardment of europium, and hence would not be present in the terbium fraction from proton-bombarded gadolinium. The aluminum beryllium and lead absorption data show electrons total range ~11 mg/cm² (~0.09 Mev) and 40 mg/cm² (0.22 Mev), electromagnetic radiation half-thicknesses 7.5 mg/cm² aluminum (6.3 kev), ~60 mg/cm² lead (~46 kev), and ~12.7 g/cm² lead (1.4 Mev); the 46-kev ray was in very low abundance and may be a mixture of K x-radiation and unconverted gamma-radiation. The corrected ratios of the various radiations obtained were 0.09-Mev e^-:0.2-Mev e^-:L x-ray:K x-ray:1.4-Mev γ-ray$=0.2:0.1:0.4:1:0.3$.

A search for α-particle emission in radioactive isotopes of terbium from both Eu+α and Gd+d bombardments has been made using very active, ion exchange resin column separated samples. No α-particle emission was observed, and approximate upper limits for decay by α-particle branching can be set for the isotopes assuming that the measured K x-radiation represents decay by orbital electron capture; the upper limit for α-branching are: 5.1-day Tb^{153}, 5×10^{-8}; 17.2-hr. Tb^{154}, 10^{-9}; 190-day Tb^{155}, 2×10^{-7}; 5.0-hr. Tb^{156}, 5×10^{-6}; 4.7-day Tb^{157}, 2×10^{-8}; 73.5-day Tb^{160}, 10^{-7}.

III. HOLMIUM ISOTOPES

In the bombardment of terbium with α-particles of various energies, radioactive isotopes of half-lives 22.5 min., 4.6 hr., and 65 days were observed. Short bombardments of dysprosium with 10-Mev protons allowed characterization of a 34-min. beta-emitting isotope, together with the 4.6-hr. and longer-lived activities; in long bombardments of dysprosium with 10-Mev protons and 19-Mev deuterons, activities of 4.6-hr. and 65-day half-life, identical with those from Tb+α-bombard-

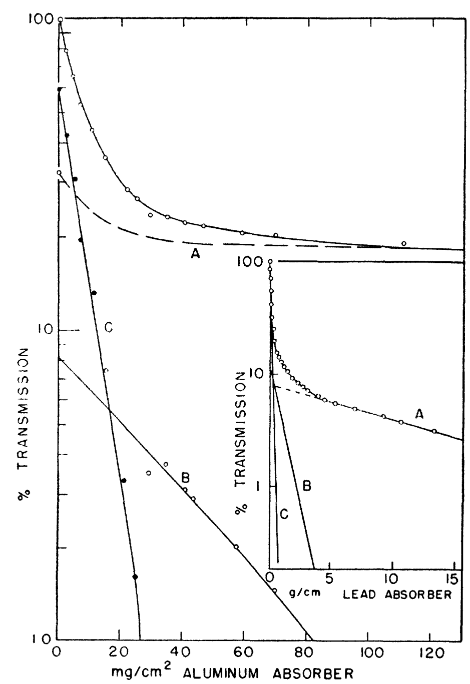

FIG. 1. Aluminum and lead absorptions of 5.1-day Tb^{153} activity from Eu+α-bombardment. Aluminum absorption A, B, and C are, respectively, the electromagnetic radiation contribution, ~0.3- and 0.15-Mev electrons. Lead absorption A, B, and C are, respectively, 1.2 Mev, 0.22-Mev gamma-rays, and K x-rays.

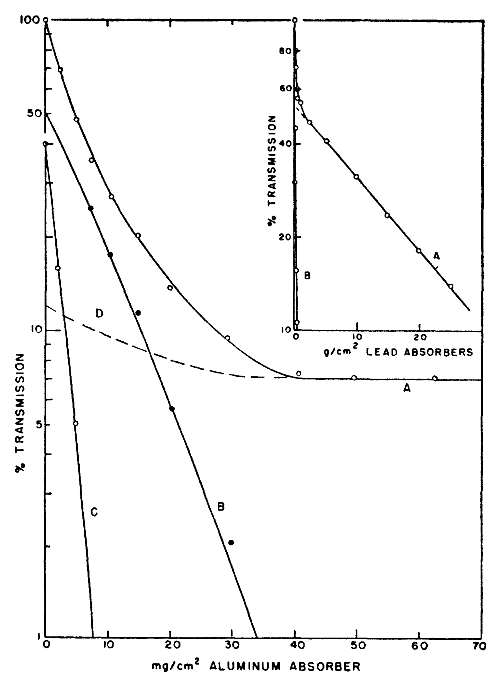

FIG. 2. Aluminum and lead absorptions of 4.7-day Tb activity from Gd+p bombardment. Aluminum absorption, K x-ray and γ-ray background (A), 0.2-Mev electron (B), 0.09-Mev electron (C), L x-rays (D). Lead absorption, 1.4-Mev γ-ray (A), K x-rays (B).

ments, were found together with an activity of 5.2-day half-life.

The allocation of the 22.5-min., 4.6-hr., and 65-day holmium activities to masses 160, 161, and 162, respectively, was made on the basis of yields in bombardments of terbium with 38-, 30-, and 19-Mev α-particles; the yields follow a pattern similar to that found for the production of thulium activities in α-particle bombardments of holmium.[1] The measured K x-radiation was taken as representing decay by orbital electron capture for the comparison of yields. The yields of the active isotopes formed in 10-Mev proton bombardments of dysprosium are approximately equal as would be expected from the similarity in abundance of the dysprosium isotopes of masses 161 to 164 inclusive. The 5.2-day activity is allocated to mass 163 since it was not observed in α-particle bombardments of terbium, or in fast neutron bombardments of holmium, but was formed in yields approximately the same as the other isotopes in proton bombardments of dysprosium.

34.0±0.5-Min. Ho^{164}

An activity of 47-min. half-life produced by fast neutron bombardment of holmium has been reported in an early paper.[3]

The 34-min. isotope here described was produced by short bombardments of pure dysprosium oxide with 10-Mev protons. Identification of the activity with holmium has not been proved by chemical separation but no activities due to known dysprosium isotopes have been observed, and longer-lived activities from the same bombardment have been shown to follow holmium chemistry. A half-life of 34.0±0.5 min. was obtained through eight periods, after subtraction of the longer-lived holmium activities, principally the 4.6-hr. activity, from the decay curves. The resolved aluminum absorption curve of the 34-min. activity shows this to have a simple beta-particle. The Feather range, 370 mg/cm^2 aluminum (0.95 Mev), agrees with the maximum energy obtained for the negative beta-particle on a simple beta-ray spectrometer. An upper limit of 0.05 γ-ray quanta per beta-particle can be set; no γ-radiation or conversion electrons attributable to the 34-min. activity were observed even in very active samples.

5.20±0.05-Day Ho^{163}

This activity whose decay was followed through six periods was found only in long bombardments of dysprosium with deuterons or protons. The ratio of the various radiations obtained from absorption measurements and corrected for counting efficiency, absorption in counter window, etc. are approximately, 0.4-Mev e^-; L x-ray,:K x-ray:$\sim$0.5-Mev γ-ray:1.4-Mev γ-ray, 0.04:0.6:1:$\sim$0.1:$\sim$0.25. The isotope thus appears to decay predominantly by orbital electron capture; the 0.04-Mev electron probably results from conversion of the weak 0.5-Mev γ-ray.

65±0.5-Day Ho^{162}

This activity, the decay of which has been followed for eight periods, is produced both in Tb+α and Dy+p bombardments and is identical in both cases. The Feather range of 300 mg/cm^2 (0.8 Mev) for the hard negative electron corresponds well with the maximum energy of 0.8 Mev obtained from the spectrometer. The ratios of the radiations are 0.1-Mev e^-:0.8-Mev β^-:L x-ray:K x-ray:1-Mev γ-ray$=\sim$0.1:$\sim$0.15:$\sim$1:1:<0.1. The isotope thus appears to decay predominantly by orbital electron capture with $\sim$15 percent branching decay by negative beta-particle emission. The shape of the aluminum absorption curve of the 0.8-Mev particle and also its distribution on the magnetic spectrometer is that of a negative beta-particle.

4.6±0.1-Hr. Ho^{161}

This activity was observed with identical radiation characteristics in both Tb+α and Dy+p bombardments. The decay of both electron and electromagnetic was followed through seven half-lives. The approximate ratios of the various radiations obtained are: $\sim$0.1-Mev e^-:L x-ray:K x-ray:1.1-Mev γ-ray$\approx\sim$0.1:$\sim$1:1:$\sim$1.

22.5±0.5-Min. Ho^{160}

This activity was observed only in short 38-Mev α-particle bombardments of terbium together with the 4.6-hr. and 65-day activities. The decay was followed through six periods. The approximate ratios of the radiation are 0.17 e^-:β^+:L x-ray:γ-ray$\approx$0.1:$\sim$0.005:$\sim$0.4:1:$\sim$1. Assuming that one K x-ray quantum represents one disintegration by orbital electron capture, branching decay by positron emission is about 0.5 percent.

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[3] M. L. Pool and L. L. Quill, Phys. Rev. **53**, 437 (1938).