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DYSPROSIUM-154, A LONG-LIVED ALPHA EMITTER

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ABSTRACT

Gadolinium oxide, enriched in Gd^{154} , was bombarded with 48-Mev alpha particles to form dysprosium isotopes. Alpha pulse-analysis of the chemically separated dysprosium resulted in the observation of a new weak alpha activity of 2.85 ± 0.05 Mev energy. This activity was assigned to Dy^{154} on the basis of a rough excitation function and consideration of alpha-decay systematics of the neighboring dysprosium isotopes.

The alpha half life was calculated as approximately 1×10^6 years by estimating the amount of Dy^{154} produced and comparing it with the alpha-count rate.

A lower limit of 10 years for the beta-decay half life was established on the basis of the absence of the prominent gamma radiation of Tb^{154} in the photon spectrum.

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INTRODUCTION

The observation of alpha activity in the rare earth elements is the result of an enhancement of alpha-decay energy due to the decrease in neutron binding energy of nuclides containing between 84 and 88 neutrons.

In 1953, Rasmussen et al. reported results on alpha emitters of europium, gadolinium, terbium, and dysprosium.¹ The dysprosium alpha emitters were later studied in greater detail by Toth and Rasmussen, who identified the alpha activities of Dy¹⁵⁰, Dy¹⁵¹, Dy¹⁵², Dy¹⁵³, and Dy¹⁵⁴.^{2,3} The alpha activity attributed to Dy¹⁵⁴ decayed with a half life of 13 hours and possessed an alpha energy of 3.35 Mev. From the observed alpha-counting rate, it appeared that the alpha-to-beta branching ratio was indeed very small. However, no evidence for a beta decay branch was observed: neither a 13-hour decay component in the gamma spectrum nor the appearance of the daughter, Tb¹⁵⁴, in the gamma spectrum. Toth postulated that this alpha activity was due to an isomer of Dy¹⁵⁴ and that the ground state actually was long-lived.⁴

The purpose of this investigation was to test, in part, this hypothesis by searching for a long-lived alpha-emitting Dy¹⁵⁴.

EXPERIMENTAL DETAILS

Two samples of 3 mg each of gadolinium oxide, enriched in Gd¹⁵⁴ to 33%, were irradiated in a stacked-foil arrangement with 30 microampere-hours of alpha particles in the Berkeley 60-inch cyclotron. The first sample was bombarded at 48 Mev, and the second sample at 37 Mev.

Separation of dysprosium from the target material was accomplished by a cation-exchange method described elsewhere.⁵ The eluting agent was 0.4 M alpha-hydroxy isobutyric acid buffered to a pH of 3.95.

After the rare earth separation, the dysprosium fraction was evaporated to dryness and the residue dissolved in concentrated hydrochloric acid. The

solution was then passed through a Dowex-1 X10 anion-exchange column in order to remove some of the inert material which had passed through the cation-exchange column (mostly iron). The eluate was then taken to dryness and treated with aqua regia to remove most of the organic material present.

Finally, the residue was taken up in a couple of drops of concentrated nitric acid and transferred to a 1-inch-diameter platinum disc. The disc was heated to dryness and ignited to red heat. This procedure resulted in the formation of an essentially weightless deposit suitable for alpha-pulse-analysis.

Alpha spectra were obtained by using a Frisch-grid argon-methane flow-type ion chamber. Electronic amplification was provided by a Berkeley "ball and chain" preamplifier and a "Miemar" main amplifier.⁶ The spectrum was displayed on a 100-channel pulse-height analyser.⁷

Energy calibration was provided by Sm^{147} (2.23 Mev)⁸ and Gd^{148} (3.16 Mev).¹

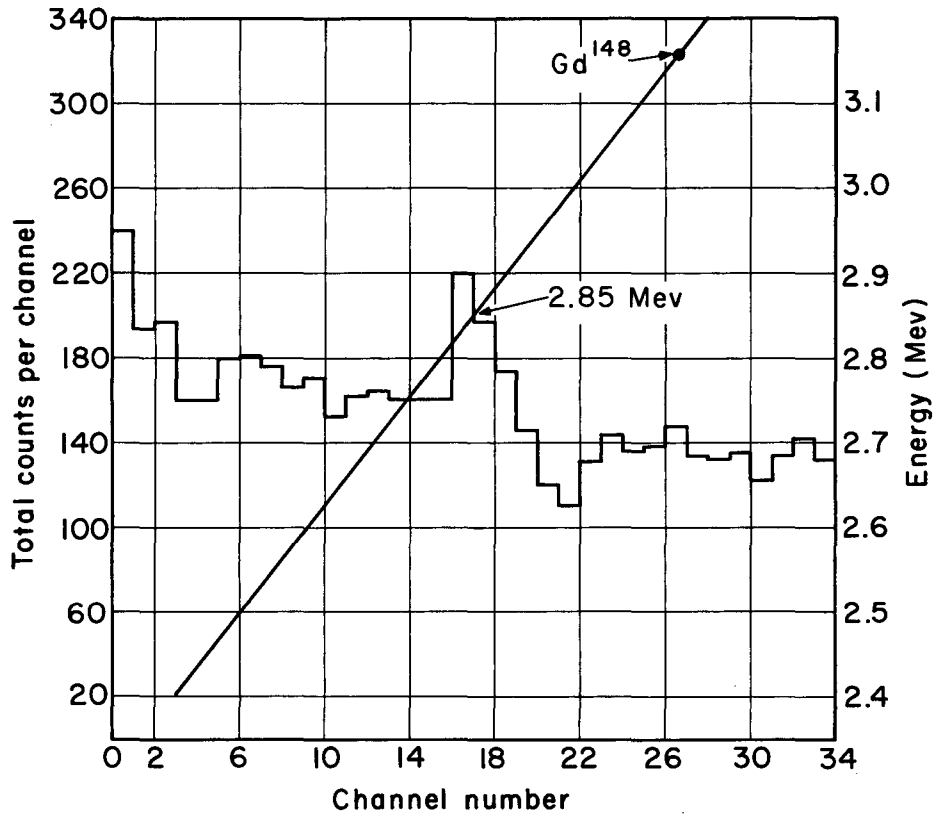
TABLE I

Isotopic abundance of target gadolinium.							
	<u>152</u>	<u>154</u>	<u>155</u>	<u>156</u>	<u>157</u>	<u>158</u>	<u>160</u>
Enriched Gd^{154}	0.32	33.19	38.57	15.92	5.49	4.50	2.05
Natural Gd	0.20	2.15	14.73	20.47	15.68	24.87	21.90

RESULTS

1. Alpha-Decay Properties

Fig. 1 shows the alpha spectrum of the dysprosium fraction from the 48-Mev bombardment taken 3 days after the end of the bombardment. A weak alpha group of 2.85 Mev energy with an intensity of ~ 0.07 count per minute was observed. This activity was not observed in the sample from the 37-Mev bombardment.



MU-21098

Fig. 1. Alpha Particle Spectrum of Dy¹⁵⁴. Counting time: 4269 minutes.

Assuming that the alpha activity was due to Dy^{154} , produced by an $(\alpha, 4n)$ reaction on Gd^{154} , the total number of atoms of Dy^{154} produced was calculated by using an estimate of 1 barn for the cross section.⁹

From these data, the alpha half life of Dy^{154} was determined to be 1×10^6 years, uncertain by a factor of 3.

2. Search for Beta-Decay Branch

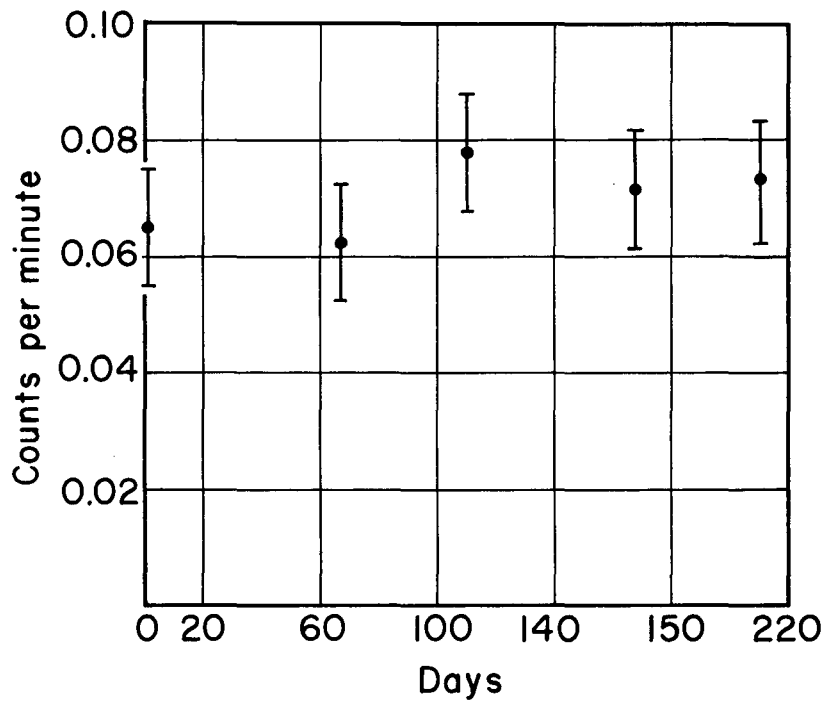
The alpha spectrum was recorded at different intervals over a period of 10 months. As shown in Fig. 2, no decrease in the alpha-particle counting rate was observed.

Next, the gamma spectrum was studied 10 months after bombardment in order to see whether the beta-decay daughter, 19-hour Tb^{154} , was present. It is characterized by a strong 123-keV gamma radiation.¹⁰ The only peaks present, however, were those due to Gd^{153} , arising initially from Dy^{153} . The absence of the 123-keV gamma radiation of Tb^{154} , was used to establish a lower limit for the beta half life of 10 years. (It was estimated that the 123-keV gamma was present in at least 20% abundance in the electron-capture decay of Tb^{154} .)

3. Mass Assignment

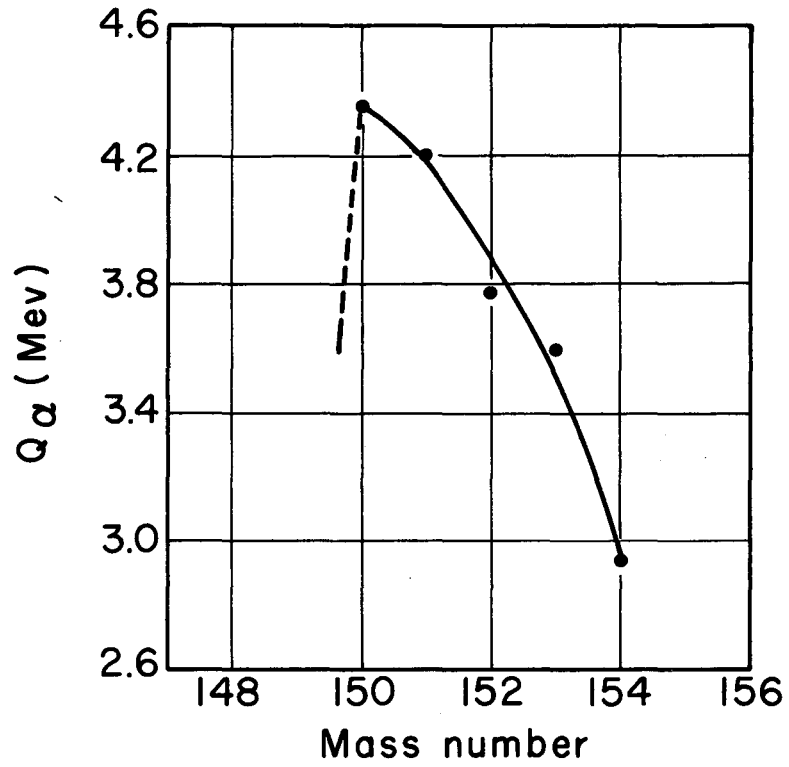
The assignment of the 2.85-MeV alpha activity to Dy^{154} was based on the following argument. That the activity was observed at a bombarding energy of 48 MeV and not at 37 MeV indicates that the activity was produced by an $(\alpha, 4n)$ but not an $(\alpha, 3n)$ reaction (very little $(\alpha, 4n)$ product is formed at 37 MeV in the rare earth region). Of all the gadolinium isotopes present in the target material (see Table I) only Dy^{152} , Dy^{154} , and Dy^{160} could be formed by an $(\alpha, 4n)$ and not an $(\alpha, 3n)$ reaction. Dy^{160} was eliminated because it is a naturally occurring isotope with a very long alpha half life. Dy^{152} was also eliminated because its alpha-decay properties have already been characterized and are different from those observed in this work.² This left Dy^{154} as the only other possibility.

From the trend of the alpha-decay energies of the other dysprosium isotopes, it appeared reasonable that Dy^{154} should have the observed alpha-particle energy (see Fig. 3).



MU - 21099

Fig. 2. Counting Rate of Decay Curve of Dy¹⁵⁴ Alpha Activity.



MU - 21100

Fig. 3. Variation of Alpha Decay Energy with Mass Number for the Dysprosium Isotopes.

DISCUSSION

The discovery of long-lived Dy¹⁵⁴ provides an answer to the puzzling result of Toth, who was unable to observe the growth of Tb¹⁵⁴ in samples known to contain Dy¹⁵⁴. It also relieves somewhat what appears to be an abnormally large difference in alpha-decay energy between Dy¹⁵⁴ and Dy¹⁵⁶ compared with what has been observed from mass data for Gd¹⁵² - Gd¹⁵⁴ and Sm¹⁵⁰ - Sm¹⁵².¹¹ (An upper limit of 2.0 Mev for the alpha-decay energy of Dy¹⁵⁶ has been established on the basis of the negative results obtained by Riezler and Kaww for natural dysprosium.¹²)

The origin of the 13-hour alpha emitter reported by Toth and Rasmussen is still a puzzle. Preliminary results confirm the apparent existence of this alpha activity, but there is some indication that it is not due to Dy¹⁵⁴.¹³ Further work is in progress on this problem.

ACKNOWLEDGMENTS

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