Alpha-Emitting Isomeric State of Tb^{149}†

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Bombardment of La^{139} targets with 80- to 140-Mev O^{16} ions has resulted in the identification of a new alpha activity decaying with a half-life of 4.3±0.2 min and an alpha-particle energy of 3.99±0.03 Mev. Subsequent experiments have established that the activity is due to an isomeric state of Tb^{149}. It is proposed that the isomerism due to the filling of an $h_{11/2}$ proton state for the spherical Tb isotopes.

I. INTRODUCTION

THE appearance of alpha radioactivity among nuclides in the rare-earth region is caused by a decrease in neutron binding energies of isotopes lying just above the 82-neutron closed shell. This results in an increase in alpha-decay energy. Two terbium isotopes are known to decay by alpha-particle emission[1]: Tb^{149} and Tb^{151}. Terbium-149, which decays to the closed shell of 82 neutrons should possess the largest alpha-decay energy of the terbium isotopes in this region. According to the systematics of other rare-earth nuclides,[2,3] the alpha-decay energy is considerably reduced at $N=83$ and reaches a minimum at $N=82$. Thus, Tb^{148} and Tb^{147} will probably not exhibit a measurable alpha-decay branch.

In the present investigation, which is a part of a study of rare-earth alpha emitters produced by heavy-ion reactions, the $La^{139}+O^{16}$ reaction was studied yielding isotopes up to terbium. Alpha-particle spectra of the activity revealed the presence of a new alpha group, which has been ascribed to an isomeric state of Tb^{149}.

II. EXPERIMENTAL PROCEDURE

A. Targets

Rare-earth targets (~2 mg/cm²) were prepared by evaporating an alcohol solution of the rare-earth nitrate onto a 0.006-mm-thick aluminum foil and heating the foil over a hot plate to convert the nitrate to an adhering deposit of the oxynitrate. The foils were then mounted on a brass wheel containing eight equally spaced 2.5-cm-diam holes. A similar wheel was used to hold aluminum degrading foils. Both wheels were then mounted in an assembly that fitted in the target chamber and made possible the changing of targets and absorbers by externally dialing them into a position in front of the beam. The positions of the target and absorber wheels in relation to the rest of the target chamber are shown in Fig. 1. The data of Northcliffe for heavy ions were used to convert range in aluminum to energy.[4]

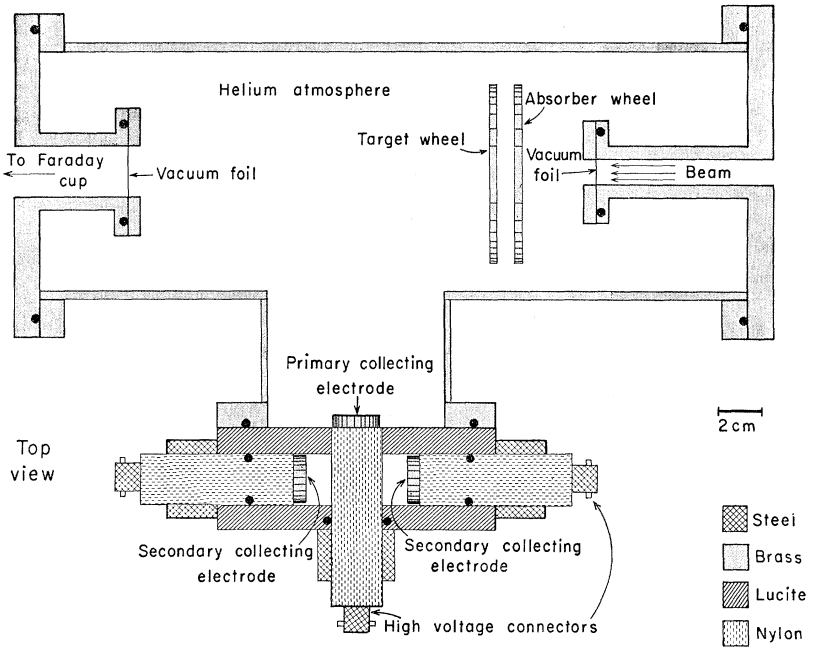

FIG. 1. Target chamber and "recoil-milking" apparatus.

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[1] J. O. Rasmussen, Jr., S. G. Thompson, and A. Ghiorso, Phys. Rev. **89**, 33 (1953).
[2] R. D. Macfarlane, thesis, Department of Chemistry, Carnegie Institute of Technology, NYO-7687, 1959 (unpublished).
[3] K. S. Toth and J. O. Rasmussen, Nuclear Phys. **16**, 474 (1960).
[4] L. C. Northcliffe, Phys. Rev. **120**, 1744 (1960).

B. Recoil Collection and Counting

The beam of the Berkeley heavy-ion linear accelerator (Hilac) entered the target chamber through a 0.025-mm-thick Dural foil window, struck the absorber and target wheels, continued through the chamber, and was finally collected and measured in a Faraday cup (see Fig. 1). Recoils from nuclear reactions were ejected from the target and slowed down in 1 atm of helium. When a voltage was applied to a primary collecting electrode, many of the recoils were deposited on its surface.[5] The electrode assembly consisted of a solid nylon cylinder with a 2.5-cm-diam brass plate attached to one end and connected to a high-voltage supply. The collecting efficiency was measured by comparing the intensity of the collected activity with that of a sample made by catching the recoils on a foil in back of the target in vacuum. It was found to be dependent upon bombarding energy, varying a factor of 3 between 80 and 140 Mev. Above 1500 v, the collecting efficiency is independent of the voltage.

A rather surprising finding was that the collecting efficiency was a factor of 3 larger at +1500 v than at −1500 v, implying that the collection of the activity as negative ions is more efficient than collecting as positive ions. No attempt has been made to determine the mechanism of negative-ion formation in this system. It may be associated with the formation of highly reactive organic species produced from the interaction of organic impurities in the helium with the heavy-ion beam. These might react with the recoils forming a stable negative-ion complex. The best collecting efficiency obtained has been 30%.

In most of the experiments, recoils were collected on a 2.5-cm-diam platinum disk, which was held against the collecting electrode by a ring clamp. After sufficient activity was collected, the electrode assembly was pulled out of the target chamber and the platinum disk placed in a Frisch-grid ionization chamber to obtain alpha-particle spectra. The grid chamber was calibrated for

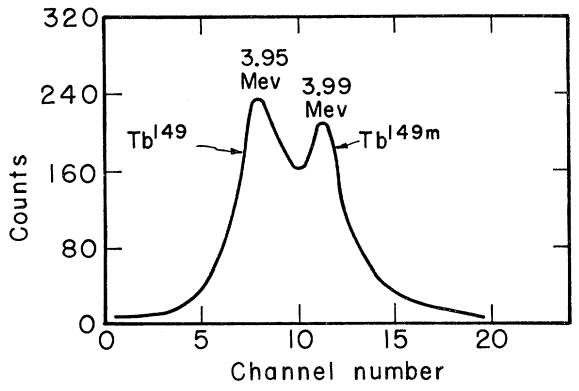

FIG. 2. Alpha-particle spectrum of Tb^{149} and Tb^{149m} produced by bombarding La^{139} with 104-Mev O^{16} ions.

[5] A. Ghiorso, T. Sikkeland, J. R. Walton, and G. T. Seaborg, Phys. Rev. Letters **1**, 18 (1958).

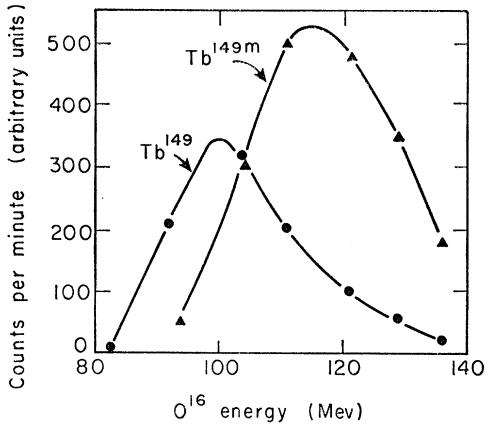

FIG. 3. Excitation functions for Tb^{149} (●) and Tb^{149m} (▲).

energy by using alpha particles from Dy^{152} (3.66 Mev), Dy^{151} (4.06 Mev), and Dy^{150} (4.21 Mev).[3]

C. Collection of Recoils from Radioactive Decay

Since the recoils from the target are deposited only on the surface of the collecting electrode, the sample layer is extremely thin—thin enough to collect recoils from radioactive decay.[5] By detecting the presence of these recoils collected at two different times, one can establish a parent-daughter relationship.

The experimental arrangement is shown in Fig. 1. The primary collecting electrode was modified so that it could be motor driven. Activity from the target was collected on this electrode, which was rotated to insure a nearly homogeneous collection across the face of the electrode. The voltage was then removed from this electrode, and the target chamber pumped down to a pressure of 100 μ. Then the electrode was driven back to a position in front of the secondary collecting electrodes. A voltage of −1500 v was then applied consecutively to the secondary electrodes for a timed period. Recoils were collected on platinum plates which fitted on to the secondary electrodes, and from these plates alpha-particle spectra were obtained of the daughter activities.

III. RESULTS

A natural lanthanum oxide target was bombarded with 80- to 140-Mev O^{16} ions to produce nuclides up to terbium. In addition to the 4.1-hr Tb^{149} (3.95 Mev), a new alpha group was observed at 3.99±0.03 Mev, decaying with a half-life of 4.3±0.2 min. An alpha-particle spectrum of the new activity produced at a bombarding energy of 104 Mev is shown in Fig. 2. Excitation functions were obtained for the two groups at bombarding energies between 80 and 140 Mev. The results are shown in Fig. 3.

Because of the short half-life, it was not possible to determine the chemical properties of the new activity. Rather an alternative procedure was used where the

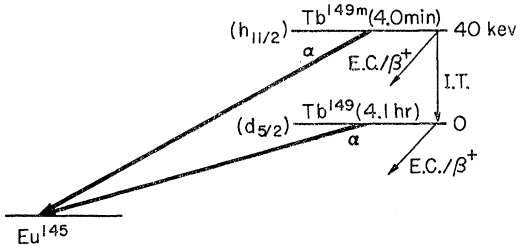

FIG. 4. Proposed alpha decay scheme of Tb^{149}.

element was identified by a process of elimination. A natural barium oxide target was bombarded with O^{16} ions over the same energy range. This should produce all the nuclides formed in the $La^{139}+O^{16}$ bombardments with the exception of the isotopes of terbium. Alpha-particle spectra of samples obtained in these bombardments did not show the presence of the 4-min activity or the 4.1-hr Tb^{149}, indicating that the 4-min activity was due to an isotope of terbium.

From the excitation-function data, it appeared that the mass number of the 4-min activity was less than 149, probably 148, because its excitation function peaked about 15 Mev higher than the excitation function for the 4.1 hr Tb^{149} (Fig. 3). This would be a surprising result because the alpha-particle energy of Tb^{148} is expected to be much lower than Tb^{149}. To obtain a check on the energetics of the O^{16}, $6n$ reaction in this region, the excitation function for the reaction $Ce^{140}(O^{16},6n)Dy^{150}$ was studied. It was found to peak at 118 Mev, which is 18 Mev higher than the corresponding $La^{139}(O^{16},6n)Tb^{149}$ reaction giving rise to the 4.1-hr Tb^{149}. However, the excitation function for the 4-min activity peaks at approximately the same energy as the $Ce^{140}(O^{16},6n)Dy^{150}$ reaction. [The difference in Q between $Ce^{140}(O^{16},6n)Dy^{150}$ and $La^{139}(O^{16},6n)Tb^{149}$ as calculated from the semiempirical atomic-mass formula of Seeger is $\sim$3 Mev].[6] The possibility that the 4.1-hr activity is Tb^{150} and not Tb^{149} can be ruled out because the mass number has been reliably established by using a mass spectrometer.[7] A possible explanation for the apparent shift in the 4.1-hr Tb^{149} excitation function is that it might be the low-spin member of an isomer pair, with the 4-min activity representing the high-spin state. Such shifts, much smaller in magnitude, have previously been observed in isomer-pair formation with low-energy deuteron reactions.[8]

Proof that the 4-min activity was due to an isomeric state of Tb^{149} would be obtained if it could be shown that the 4-min activity exhibited some decay to the 4.1-hr Tb^{149}. To do this, the "recoil-milking" experiment described in Sec. II.C was used. Activity was collected from a $La^{139}+120$-Mev O^{16} bombardment. Recoils from decay were collected for 4 min on each secondary plate. Alpha-particle analysis of the secondary plates showed the presence of a very small amount of the 4.1-hr Tb^{149}. The ratio of the activity on the two plates indicated that its precursor had a half-life of a few minutes. This provides good evidence that the 4-min activity is an isomeric state of Tb^{149}.

IV. DISCUSSION

The "recoil-milking" experiment establishes that the 4-min activity is the upper state. If the 4-min activity decays to the ground state of Eu^{145} (no alpha groups of an energy higher than 3.99 Mev were observed), then the energy level of the isomeric state can be obtained from the difference between the alpha-particle energies of the two states (40 kev).

Terbium-149, with 84 neutrons, lies close to the 82-neutron closed shell and most likely possesses a stable spherical shape. The existence of a long-lived isomeric state can possibly be explained then by a consideration of shell-model proton states with zero deformation. Above the 50-proton closed shell, the proton-level sequence suggested by Mottelson and Nilsson is $g_{7/2}$, $d_{5/2}$, $h_{11/2}$, $d_{3/2}$, and $s_{1/2}$.[9] If the order of filling is according to this sequence, the sixty-fifth proton (Tb) should begin the filling of the $h_{11/2}$ level. If this level is only filled by pairs of nucleons, as is observed with the $h_{11/2}$ neutron shell in the region around $Z=50$,[10] then the ground state of Tb^{149} would probably be $d_{5/2}$ and the isomeric state $h_{11/2}$. This would give rise to an $E3$ isomer. A proposed decay scheme is shown in Fig. 4.

Terbium-149 is probably not a unique case in this region. The other spherical isotopes of Tb and higher members in the rare-earth region that fill the $h_{11/2}$ state may also show the same kind of isomerism.

The peak of the excitation function for Tb^{149} is shifted downward approximately 15 Mev relative to Tb^{149m}. This is the largest shift that has been observed for an isomer pair. A possible explanation for this shift is that the high-spin isomer may be formed primarily from compound nuclei having high angular momenta where de-excitation by gamma-ray emission has been found to compete favorably with nucleon evaporation.[11] Gamma-ray de-excitation may be less important for compound nuclei with low angular momenta, which probably contribute most in the formation of the low-spin isomer.

ACKNOWLEDGMENTS

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