# Production of Rare-Earth $\alpha$ Emitters with Energetic <sup>3</sup>He Particles; New Isotopes: <sup>151</sup>Er, <sup>156</sup>Yb, and <sup>157</sup>Yb<sup>†</sup>

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 $\alpha$ -active isotopes in the rare-earth region were investigated by bombarding  $^{156}\text{Dy}$  and  $^{162}\text{Er}$ targets with <sup>3</sup>He ions accelerated in the Oak Ridge isochronous cyclotron. In the experiments recoil nuclei ejected from the thin targets were stopped in helium gas, swept through an orifice, and deposited on catcher foils. After each bombardment the activity collected on the catchers was assayed with an  $\alpha$ -particle Si(Au) spectrometer. In the case of the <sup>156</sup>Dy target relative yields were obtained as a function of bombarding energy for known erbium, holmium, and dysprosium nuclides. Excitation functions determined from statistical-model calculations were found to be in reasonable agreement with the shapes of the experimental  $({}^{3}\text{He}, xn)$  and  $({}^{3}\text{He}, pxn)$ curves but not with those of the  $({}^{3}\text{He}, \alpha xn)$  curves. Experimentally, the yields for  $({}^{3}\text{He}, pxn)$ and  $({}^{3}\text{He}, \alpha xn)$  products were found to be ~3 to 5 times greater than those for  $({}^{3}\text{He}, xn)$  products. The statistical-model calculations, contrastingly, indicated the  $({}^{3}\text{He}, xn)$  and  $({}^{3}\text{He}, pxn)$  cross sections to be comparable and ~5 times greater than those for  $({}^{3}\text{He}, \alpha_{Xn})$  products. Similar yields were measured for nuclides observed in bombardments of the <sup>162</sup>Er target and were used in the assignment of two new  $\alpha$ -emitting isotopes to <sup>156</sup>Yb and <sup>157</sup>Yb. Their half-lives and  $\alpha$ decay energies were measured to be : (1)  $^{156}$ Yb,  $24 \pm 1$  sec,  $4.80 \pm 0.01$  MeV, and (2)  $^{157}$ Yb,  $34\pm3$  sec,  $4.50\pm0.01$  MeV. The assignment of the 4.80-MeV  $\alpha$  group to <sup>156</sup>Yb was clearly established by noting a generic relationship between it and its known  $\alpha$ -decay daughter, <sup>152</sup>Er. A similar clear-cut confirmation, based on a generic relationship, was not possible for  $^{157}$ Yb. The previously unknown nuclide,  $^{151}$ Er, was found in the  $^{156}$ Dy bombardments by noting an initial growth period in the 4.60-MeV  $\alpha$ -particle group known to follow the decay of its daughter, <sup>151m</sup>Ho. From this growth the half-life of <sup>151</sup>Er was found to be  $23 \pm 2$  sec. The existence of 5.3-min  $^{155}$ Er, previously observed by us in  $^{4}$ He bombardments of  $^{156}$ Dy, was confirmed. Its  $\alpha$ -decay energy was measured to be 4.01 ± 0.01 MeV, in agreement with the value reported earlier.

### I. INTRODUCTION

The work reported here is part of a program that deals with the production and characterization of new rare-earth  $\alpha$  emitters with N = 86 and 87. Earlier publications<sup>1,2</sup> discussed the properties of <sup>153,154</sup>Ho and <sup>155</sup>Er, nuclides produced by bombarding enriched targets of <sup>156</sup>Dy with protons and <sup>4</sup>He ions. The maximum proton and <sup>4</sup>He energies available at the Oak Ridge isochronous cyclotron (ORIC) are  ${\sim}65$  and  ${\sim}80$  MeV, respectively. These energies are not high enough to produce the thulium and ytterbium isotopes with N = 86 and 87 by bombarding the most neutron-deficient stable isotope of erbium, <sup>162</sup>Er. Reaction energetics indicated, however, that 100-MeV <sup>3</sup>He ions incident on <sup>162</sup>Er could induce the  $({}^{3}\text{He}, 8n)$  and  $({}^{3}\text{He}, 9n)$  reactions necessary to produce the ytterbium nuclides of interest. (The design characteristics of the ORIC are such that 100-MeV  $^{3}$ He ions can be accelerated at the radio frequency and magnetic field needed for 80-MeV <sup>4</sup>He ions.)

A beam of several  $\mu A$  of doubly charged 100-MeV <sup>3</sup>He ions was developed and a search for <sup>156</sup>Yb and <sup>157</sup>Yb was initiated. It soon became clear that a separate investigation with <sup>156</sup>Dy as the target nucleus was necessary to: (1) confirm the previously reported<sup>3</sup> decay characteristics of <sup>152</sup>Er and <sup>153</sup>Er, which are the  $\alpha$ -decay daughters of <sup>156</sup>Yb and <sup>157</sup>Yb, and (2) establish excitation functions for  $({}^{3}\text{He}, xn)$ . (<sup>3</sup>He, pxn), and (<sup>3</sup>He,  $\alpha xn$ ) reactions that would lead to known erbium, holmium, and dysprosium nuclides.<sup>4</sup> These excitation-function data could then be used in the mass assignment of the new ytterbium nuclides. It was also noted that the  ${}^{156}$ Dy( ${}^{3}$ He, 8n) reaction could produce a new erbium activity, <sup>151</sup>Er. From systematics this 83-neutron nuclide was not expected to exhibit an observable  $\alpha$ -decay branch. Its  $\beta$  decay, however, could be observed by noting a growth period followed by decay in the  $\alpha$  spectrum of its 84-neutron holmium daughter.

#### II. EXPERIMENTAL METHOD

The experimental assemblies used in this inves-

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tigation are based upon a system first reported by Macfarlane and Griffioen<sup>5</sup> and used by others.<sup>6</sup> The fundamental idea inherent in these systems (illustrated schematically in Fig. 1) is that radioactive recoil nuclei ejected from thin targets are stopped in helium gas in a "high-pressure" chamber (~0.2 to 2 atm) and then carried by the gas through a very small orifice into a "low-pressure" chamber (~0.2 to 0.5 Torr), where they are deposited on a catcher. In one of the assemblies used in the present study this catcher consisted of a foil mounted on a polyethylene "rabbit." After the end of an irradiation the "rabbit" was transferred in ~5 sec through a pneumatic tube into a chamber where the  $\alpha$  activity collected on the foil was then assayed with a Si(Au) detector. In the other system a rapidly driven wheel conveyed the collected radioactivity to a position in front of a Si(Au) detector in  $\sim 0.2$  sec.

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Targets consisted of thin layers of rare-earth oxide, ~0.6 mg/cm<sup>2</sup>, electrodeposited onto platinum or nickel supporting foils. The isotopic compositions of the enriched oxides were such that the target nuclei of interest, <sup>156</sup>Dy and <sup>162</sup>Er, made up 12.6 and 20.4% of the target materials, respectively. During irradiation, targets were positioned so that first the backing foil and then the oxide deposit intercepted the beam. It should be pointed out that the rare-earth oxide deposits were thicker than the recoil ranges calculated on the basis of full momentum transfer.

 $\alpha$ -particle spectra were measured with the Si(Au) detectors coupled through a low-noise charge preamplifier, linear amplifier, and postamplifier to

a multichannel analyzer. A 1600-channel analyzer was used in conjunction with the "rabbit" system. This pulse-height analyzer could be used as 8 200channel or 16 100-channel analyzers to store spectra as a function of decay time. Spectra measured with the apparatus that utilized the rapidly moving wheel were stored in a multiparameter 20 000-channel analyzer. This analyzer was ordinarily used in a  $100\!\times\!200$  mode to measure spectra at different decay times. A precision pulser together with a <sup>244</sup>Cm source was used to set the energy scale of these spectrometers, but internal calibration standards were used to determine precisely the energies of the various  $\alpha$ -particle peaks observed. The full width at half-maximum (FWHM) for the various  $\alpha$ -particle peaks observed during the experiments was typically 30 to 35 keV.

## III. DATA FROM <sup>3</sup>He INCIDENT ON <sup>156</sup>Dy

#### A. Yields for Various Reaction Products

With the use of the "rabbit" assembly, a series of 20-sec bombardments followed by 6 min of counting was made on a <sup>156</sup>Dy target. The primary purpose was to obtain relative yields for the various  $\alpha$  emitters produced as a function of the <sup>3</sup>He bombarding energy. While the initial <sup>3</sup>He energy was 102.1 MeV, the maximum available energy, without any absorbers, was 97.3 MeV, since the <sup>3</sup>He ions had to pass through an aluminum window foil and a 2.5-mil platinum target-backing foil.

Figure 2 shows spectra taken over a period of time following the bombardment of <sup>156</sup>Dy with 97.3-MeV <sup>3</sup>He ions. Part (a) of Fig. 2 represents the



SCHEMATIC OF GAS-JET SYSTEM (Top View)

FIG. 1. Schematic illustration of one of the systems used in this investigation. The other system utilized a foil mounted on a "rabbit" to collect the recoils. The "rabbit" could then be transferred through a pneumatic tube into a vacuum chamber and placed in front of a Si(Au) detector.



FIG. 3. Yields as a function of bombarding energy for activities produced from <sup>3</sup>He incident on <sup>156</sup>Dy. Curves labeled by various reactions are calculated from the statistical model and are normalized to the data points.

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sum of the first two 10-sec counts, part (b) the sum of four spectra measured over the next 110 sec of counting, and part (c) the sum of a 1- and 2-min count. Eight main  $\alpha$  groups were observed and on the basis of  $\alpha$  energies and half-lives were assigned to: <sup>152</sup>Er, <sup>153</sup>Er, <sup>151</sup>Ho, <sup>151m</sup>Ho, <sup>152</sup>Ho, <sup>152m</sup>Ho, <sup>150</sup>Dy, and <sup>151</sup>Dy.

Yields as a function of incident energy are shown in Fig. 3. The experimental counting rates were corrected for differences in beam intensity, bombardment time, and available<sup>4</sup>  $\alpha$ /total branching ratios. The ordinate scale is expressed in relative units because the efficiency for ejection of the recoil products from the <sup>156</sup>Dy target and their subsequent collection on the platinum catcher foil is not known. Furthermore, the recoil range is expected to increase with bombarding energy; this increase should not be large in going from 70 to 100 MeV and the data shown in Fig. 3 are not corrected for this effect.

Curves shown in Fig. 3 are excitation functions calculated by using the statistical theory<sup>7</sup> of compound-nuclear reactions and are normalized to the data points. It is seen that the energy dependence exhibited by the  $({}^{3}\text{He}, xn)$  and  $({}^{3}\text{He}, pxn)$  data is reproduced reasonably well by the calculated excitation functions. The particular values of the level density and radius parameters used in the calculations were a = A/20 and  $r_0 = 1.5$  F; these same values were used in our previous studies on holmium<sup>1</sup> and erbium<sup>2</sup> isotopes produced in bombardments of <sup>156</sup>Dy with protons and <sup>4</sup>He ions. As is seen in Fig. 3, the  $({}^{3}\text{He}, pxn)$  yields appear to be three or four times greater than those from the  $({}^{3}\text{He}, xn)$  reactions, while the calculations indicate that the  $(^{3}\text{He}, xn)$  and  $(^{3}\text{He}, pxn)$  products in the region of 70-100 MeV should have comparable cross sections. Part or all of this discrepancy may be due to uncertainties in available  $\alpha$ -decay branching ratios. (The  $\alpha$ /total branching ratios for <sup>152</sup>Er and <sup>153</sup>Er have been reported<sup>3</sup> to be 0.90 and 0.95, while those for the holmium  $\alpha$  emitters are reported<sup>8</sup> to be between 0.20 and 0.30.) Another possible explanation is that recoils originating from  $({}^{3}\text{He}, pxn)$ reactions have greater forward momentum than those resulting from  $({}^{3}\text{He}, xn)$  reactions. This additional momentum could be due to the effect of proton evaporation on the velocity of the recoils<sup>9</sup> or to the fact that a mechanism other than compound-nucleus decay is involved in (<sup>3</sup>He, pxn) reactions. Since the <sup>156</sup>Dy target used was thicker than the ranges of the recoil nuclei, the additional momentum would mean that a greater portion of the target material would be available for  $({}^{3}\text{He}, pxn)$ products.

Let us now examine the yields observed for <sup>150</sup>Dy and <sup>151</sup>Dy. Because the bombardments were only

20-sec in duration the counting statistics for the 6-min (<sup>150</sup>Dy) and 18-min (<sup>151</sup>Dy) activities were rather poor. This fact accounts for the large scatter in the <sup>150</sup>Dy and <sup>151</sup>Dy data points. Nevertheless, it is clear that their yields are comparable to those of the  $({}^{3}\text{He}, pxn)$  products. The calculations, on the other hand, predict that the (<sup>3</sup>He,  $\alpha xn$ ) yields should be smaller than the  $({}^{3}\text{He}, xn)$  yields by about a factor of 5. Here again part of the discrepancy may be due to uncertainties in  $\alpha$ /total ratios<sup>4</sup> (0.18 and 0.06 for <sup>150</sup>Dy and <sup>151</sup>Dy, respectively) or to a greater forward momentum imparted to the (<sup>3</sup>He,  $\alpha xn$ ) recoils. In contrast to the (<sup>3</sup>He, pxn) and (<sup>3</sup>He, xn) results, however, the shapes of the calculated curves do not reproduce the (<sup>3</sup>He,  $\alpha xn$ ) data points. A further complicating factor here is that part of the observed yields for <sup>150</sup>Dy and <sup>151</sup>Dy could be due to electron-capture decay from <sup>150</sup>Ho and <sup>151</sup>Ho. As seen in Fig. 3, <sup>151</sup>Ho (with an electron-capture/ total ratio<sup>8</sup> of  $\sim$ 75%) is produced in large yields for incident energies >90 MeV and, indeed, indication of an initial growth period was observed in the <sup>151</sup>Dy  $\alpha$  peak. The 83-neutron nuclide <sup>150</sup>Ho, which should not decay by  $\alpha$  emission, has been tentatively reported<sup>8</sup> as having a 20-sec half-life. On the basis of the yields shown in Fig. 3, it should begin to be produced in appreciable amounts only at energies above 95 MeV and, indeed, no growth period was readily apparent in the  $^{150}\text{Dy}~\alpha$  peak.

Before leaving the matter of the (<sup>3</sup>He,  $\alpha xn$ ) yields, some additional data obtained at 53.1, 48.3, and 43.2 MeV should be discussed. Figure 4 shows spectra observed at 53.1 and 43.2 MeV. These spectra represent the sum of three 2-min counts taken after 5-min irradiations. As might be expected from the results shown in Fig. 3,  $\alpha$  peaks due to <sup>152</sup>Er, <sup>151</sup>Ho, and <sup>152</sup>Ho are not seen in Fig. 4. Erbium-153 is observed at 53.1 MeV but not at 43.2 MeV. The (<sup>3</sup>He,  $\alpha xn$ ) products <sup>150</sup>Dy and <sup>151</sup>Dy are clearly present even at 43.2 MeV. Because this energy is below the threshold for the production of <sup>150</sup>Ho and <sup>151</sup>Ho, the dyprosium isotopes must be either produced independently in (<sup>3</sup>He,  $\alpha xn$ ) reactions or from the  $\alpha$  decay of <sup>154</sup>Er and <sup>155</sup>Er. It is evident from Fig. 4 that even at 43.2 MeV, where the <sup>155</sup>Er peak is prominent, there are far too many <sup>151</sup>Dy events to be accounted for by the  $\alpha$  decay of <sup>155</sup>Er. Also, if the branching ratio for <sup>150</sup>Dy is 0.18, then for every 100 <sup>154</sup>Er  $\alpha$  counts 18 will appear in the <sup>150</sup>Dy  $\alpha$  peak. It is clear that at 53.1 MeV there are too many <sup>150</sup>Dy events to be accounted for by the  $\alpha$  decay of <sup>154</sup>Er. (But at 43.2) MeV, growth was observed in the <sup>150</sup>Dy  $\alpha$  peak and the data were consistent with all  $^{\rm 150}{\rm Dy}$  events being due to  $^{154}$ Er  $\alpha$  decay.) In considering the evidence presented in Figs. 3 and 4 one must conclude that in the bombarding energy region investigated most



FIG. 4. Spectra measured for 53.1- and 43.2-MeV <sup>3</sup>He ions incident on <sup>156</sup>Dy. The spectra represent the sum of three 2-min counts.

of the observed  $^{150}$ Dy and  $^{151}$ Dy yields are independently produced.

#### B. Results for Previously Known Holmium and Erbium Isotopes

The isotope <sup>155</sup>Er was initially found<sup>2</sup> in <sup>4</sup>He bombardments of <sup>156</sup>Dy. The apparatus used was such that the recoils were not stopped in a gas; instead they were allowed to embed directly in a catcher foil. As a result, the resolution for the observed  $\alpha$  peaks was ~135 keV FWHM. The  $\alpha$  decay of <sup>155</sup>Er was observed in the present investigation (see Fig. 4), and with the improved resolution of the detector system its  $\alpha$ -decay energy was found to be  $4.01 \pm 0.01$  MeV. This value is in agreement with the one of  $4.01 \pm 0.03$  MeV reported earlier.<sup>2</sup>

The magnetic spectrometer measurements by Golovkov, Khvan, and Chumin<sup>10</sup> for the  $\alpha$ -decay energies of  $^{150}$ Dy,  $^{151}$ Dy, and  $^{154}$ Er were used as internal calibration standards in the present investigation. Their reported energy values, accurate to  $\pm 5$  keV, are as follows: <sup>150</sup>Dy, 4.232 MeV; <sup>151</sup>Dy, 4.067 MeV; and <sup>154</sup>Er, 4.166 MeV. The  $\alpha$ -decay energies measured in this work for the two isomers of <sup>151</sup>Ho and <sup>152</sup>Ho and for <sup>152</sup>Er, <sup>153</sup>Er, and <sup>155</sup>Er are compared in Table I with previously reported values. Our energies are accurate to within  $\pm 10$  keV. Thus they are more precise than values published in Refs. 2, 3, and 8. Note, however, that within the error limits the literature values do not disagree with our energies. In Table I we also compare the half-lives measured in this investigation with those reported earlier.<sup>2,3,8</sup> It is felt that more accurate half-lives were obtained for  ${}^{152}$ Er,  ${}^{153}$ Er, and  ${}^{151m}$ Ho.

### C. <sup>151</sup>Er

In 1963 Macfarlane and Griffioen<sup>8</sup> reported the existence of two  $\alpha$ -decaying states in <sup>151</sup>Ho. One of these, assigned to the ground state, emitted  $\alpha$  particles of 4.52 MeV, while the other, an isomeric state located at an undetermined excitation energy, decayed by emitting 4.60-MeV  $\alpha$  particles. The activities were produced in heavy-ion-induced reactions, and on the basis of displacements in the peaks of the two excitation functions the authors<sup>8</sup> concluded that the ground state had a higher spin than the isomeric state. From shell-model considerations the two proton orbitals most probably involved in this N=84, Z=67 system are  $h_{11/2}$  and  $d_{5/2}$ .

As mentioned in the Introduction, a search was made for the 83-neutron nuclide <sup>151</sup>Er by looking for an initial growth period in the two  $\alpha$  groups of <sup>151</sup>Ho. None was observed for the more intense (see Fig. 2) 4.52-MeV  $\alpha$  peak. The 4.60-MeV group, contrastingly, did show an initial increase

TABLE I. Decay information for previously known  $\alpha$  emitters.

Nuclide	Present work		Previous data		
	<i>T</i> <sub>1/2</sub>	$E_{\alpha}$ (MeV)	<b>T</b> <sub>1/2</sub>	$E_{\alpha}$ (MeV)	Ref.
<sup>152</sup> Er	$9.8 \pm 0.3  \sec$	$4.80 \pm 0.01$	$10.7 \pm 0.5$ sec	$4.80 \pm 0.02$	3
$^{153}$ Er	$36 \pm 1$ sec	$4.67 \pm 0.01$	$36 \pm 2$ sec	$4.67 \pm 0.02$	3
$^{155}\mathrm{Er}$	$5.5 \pm 1.0 \text{ min}$	$4.01 \pm 0.01$	$5.3 \pm 0.3$ min	$4.01 \pm 0.03$	$^{2}$
<sup>51</sup> Ho	$36 \pm 2$ sec	$4.52 \pm 0.01$	$35.6 \pm 0.4$ sec	$4.51 \pm 0.02$	8
<sup>51m</sup> Ho	$47 \pm 2$ sec	$4.60 \pm 0.01$	$42 \pm 4$ sec	$4.60 \pm 0.02$	8
<sup>52</sup> Ho (high spin)	$50 \pm 2$ sec	$4.46 \pm 0.01$	$52.3 \pm 0.5$ sec	$4.45 \pm 0.02$	8
<sup>52</sup> Ho (low spin)	$2.0 \pm 0.3 \text{ min}$	$4.38 \pm 0.01$	$2.36 \pm 0.16$ min	$4.38 \pm 0.02$	8

in intensity at <sup>3</sup>He incident energies above 90 MeV. The growth period was most pronounced at the highest bombarding energy.

A separate experiment was made to emphasize the initial growth period so that an accurate determination could be made of its half-life. The wheel assembly and the two-parameter 20 000-channel analyzer were used in this experiment. Twenty bombardments were made, each 10 sec in duration, followed, within ~0.2 sec, by forty 10-sec counts. The analyzer, set up in a  $100 \times 200$  mode, was programmed to scale automatically through 40 planes of 200 channels each. The decay data for the 4.60-MeV peak, shown in Fig. 5, are characteristic of radioactive growth and decay. Least-squares analyses of these data indicated that the 47-sec <sup>151m</sup>Ho arises in part from the decay of a nuclide



FIG. 5. Decay data obtained for the 4.60-MeV  $\alpha$  peak following 20 bombardments, each 10 sec in duration, of <sup>156</sup>Dy with 97.3-MeV <sup>3</sup>He ions.

with a half-life of  $23 \pm 2$  sec. This new activity, based on the parent-daughter relationship, was assigned to <sup>151</sup>Er. Again from shell-model considerations the ground-state spin of <sup>151</sup>Er is probably due to the  $f_{7/2}$  orbital of its odd 83rd neutron.  $\beta$  decay from this state would proceed via a firstforbidden transition to the  $d_{5/2}$  isomeric state in <sup>151</sup>Ho. The transition to the  $h_{11/2}$  ground state, on the other hand, would be highly forbidden.

## IV. DATA FROM <sup>3</sup>He INCIDENT ON <sup>162</sup>Er

As in the case of the <sup>156</sup>Dy results, the <sup>162</sup>Er target was first bombarded in the "rabbit" assembly. The energy of the ORIC beam in this set of runs was 101.2 MeV and because a 0.35-mil nickel backing was used for the Er<sub>2</sub>O<sub>3</sub> material the maximum <sup>3</sup>He energy on target with no absorbers was 100.0 MeV. A composite spectrum measured at this bombarding energy is shown in Fig. 6. The spectrum is the result of thirty 15-sec irradiations, each followed by 3 min of counting. Three  $\alpha$ groups stand out in Fig. 6, at 4.80, 4.67, and 4.50 MeV. Two smaller peaks appear at 4.46 and 4.38 MeV. Decay-curve analyses for the main three peaks indicated that: (1) The 4.80-MeV group had a 24-sec half-life and thus could not be  $^{152}$ Er, (2) the intense 4.67-MeV peak had the 36-sec half-life of <sup>153</sup>Er, and (3) the 4.50-MeV  $\alpha$  particle had a 34sec half-life, i.e., similar to that of the high-spin state of <sup>151</sup>Ho discussed above. The count rates for the peaks at 4.46 and 4.38 MeV were low; the decay curves, however, were consistent with the half-lives of the <sup>152</sup>Ho isomers.



FIG. 6. Composite spectrum measured following 100-MeV <sup>3</sup>He bombardments of <sup>162</sup>Er.

## A. <sup>156</sup>Yb

Before discussing the remainder of the results obtained in the series of runs mentioned above, attention will now be focused on the 4.80-MeV  $\alpha$ peak. It was clear from previously available data<sup>4</sup> that this  $\alpha$  group represented a new  $\alpha$ -decaying isotope. The decay data for the peak indicated an initial growth period. A series of 10-sec irradiations was made with the wheel arrangement to emphasize the growth period. The 20 000-channel analyzer was set up in a  $100 \times 200$  mode and 40 planes of 200 channels, each representing 4 sec of counting time, were utilized. 100 such irradiations were made and the summed decay data are shown in Fig. 7.

Least-squares analysis indicated a generic relationship between two radioactive components, one with a half-life of  $24 \pm 1$  sec and the other with a 9.8-sec half-life. Because this latter value is that of <sup>152</sup>Er (see Table I), the parent-daughter relationship establishes the existence of a new ytterbium nuclide, <sup>156</sup>Yb. The  $\alpha$ -decay energy,  $4.80 \pm 0.01$  MeV, within the resolution capabilities of the detection system used, is the same as that of its  $\alpha$ -decay daughter, <sup>152</sup>Er.

## B. <sup>157</sup>Yb

To determine the nuclidic assignments of the 4.67- and 4.50-MeV  $\alpha$  groups it is necessary to consider the relative yields that were measured as a function of bombarding energy. The bombardments were 30 sec in duration and were followed by 24 min of counting. By degrading the cyclotron beam with absorber foils the range from 100.0 to 85.2 MeV was covered. It was also possible to raise the beam energy slightly and yields were measured at 102.0 MeV.

The relative yields for the three major  $\alpha$  groups (see Fig. 6) are shown in Fig. 8. Statistical-model calculations were made for <sup>3</sup>He ions incident on <sup>162</sup>Er. The data points assigned to <sup>156</sup>Yb show the yield still increasing at 102 MeV and, as seen in Fig. 8, are consistent with the shape of the calculated excitation function for a (<sup>3</sup>He, 9*n*) reaction. The only calculated curve that was consistent with the 4.50-MeV data points was the one for a (<sup>3</sup>He, 8*n*) reaction. There is a strong indication then that the 4.50-MeV  $\alpha$  group is due to the decay of <sup>157</sup>Yb.

Arguments against the assignment of the 4.50-MeV peak nuclides other than <sup>157</sup>Yb will now be



FIG. 7. Decay data obtained for the 4.80-MeV  $\alpha$  group following 100 bombardments, each 10 sec in duration, of <sup>162</sup>Er with 100-MeV <sup>3</sup>He ions.



FIG. 8. Yields as a function of bombarding energy for isotopes produced from <sup>3</sup>He ions incident on <sup>162</sup>Er. The two curves labeled (<sup>3</sup>He, 8n) and (<sup>3</sup>He, 9n) are calculated from the statistical model and are normalized to the data points for the 4.50- and 4.80-MeV  $\alpha$  groups, respectively.

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The assignment of the  $\alpha$  group to <sup>151</sup>Ho is possible if one considers only the  $\alpha$ -decay energy and half-life. The experimental excitation function for the 4.50-MeV group, however, clearly peaks at ~98 MeV, a result that is unreasonable for the (<sup>3</sup>He,  $\alpha p9n$ ) reaction needed to produce <sup>151</sup>Ho in light of the data shown in Fig. 3, where the  $({}^{3}\text{He},$ p7n) excitation function is just beginning to peak at 100 MeV. Another argument against the assignment of the 4.50-MeV peak to <sup>151</sup>Ho is that the 4.60-MeV  $\alpha$  group of <sup>151*m*</sup>Ho was not observed (see Fig. 6). Results obtained with the  $^{156}$ Dy target indicate that the isomer ratio  $(^{151}\text{Ho}/^{151m}\text{Ho})$  for the  $(^{3}\text{He},$ p7n) reaction is ~2 throughout the investigated region of bombarding energies. Thus, if the <sup>151</sup>Ho isomers were produced independently in the <sup>162</sup>Er- $({}^{3}\text{He}, \alpha p 9 n)$  reaction, one would expect that the ejection of an additional  $\alpha$  particle should remove more angular momentum from the compound system and thus increase the production of the lowspin isomer <sup>151m</sup>Ho (see, e.g., Williams and Toth<sup>11</sup>).

The possible production of <sup>151</sup>Ho from the  $\alpha$  decay of the unknown nuclide <sup>155</sup>Tm, as well as the suggestion that the 4.50-MeV  $\alpha$  group is due to the  $\alpha$  decay of <sup>155</sup>Tm itself, can also be ruled out because the thulium nuclide would have to be made in a (<sup>3</sup>He, p9n) reaction. Again the shape of the observed yield curve is inconsistent with the data shown in Fig. 3. Also, the half-life of <sup>155</sup>Tm should be >5 sec (that reported<sup>12</sup> for <sup>154</sup>Tm), but no initial growth period was observed for the 4.50-MeV peak.

In the series of 100 irradiations at 100 MeV made with the wheel arrangement (see the section on <sup>156</sup>Yb), no  $\alpha$  activity was observed due to the known<sup>4</sup> 1.65-sec <sup>155</sup>Yb nuclide, even though measurements were begun within ~0.2 sec after the end of the bombardment. Thus the 4.50-MeV  $\alpha$  group cannot be due to either of the two decay sequences <sup>155</sup>Yb  $\stackrel{\alpha}{\rightarrow}$  <sup>151</sup>Er  $\stackrel{\text{E.C. 151}}{\rightarrow}$ Ho or <sup>155</sup>Yb  $\stackrel{\text{E.C. 155}}{\rightarrow}$ Tm  $\stackrel{\alpha}{\rightarrow}$  <sup>151</sup>Ho.

The assignment of the 4.50-MeV peak to <sup>156</sup>Tm is also unlikely. The shape of the observed yield curve is unreasonable for the (<sup>3</sup>He, p8n) reaction needed to produce <sup>156</sup>Tm. Also, if the assumption is made that the 4.50-MeV  $\alpha$  peak is due to <sup>156</sup>Tm, then one should see the <sup>152</sup>Ho isomers. Since the  $\alpha$ /total ratios for the <sup>152</sup>Ho isomers are between 0.2 and 0.3, the intensisties of the two peaks should be  $\sim_4^1$  the intensity of the 4.50-MeV peak. Two peaks at the correct energies, 4.46 and 4.38 MeV, are indeed seen in Fig. 6; their intensities, however, are too low to arise from the  $\alpha$  decay of <sup>156</sup>Tm. Figure 9 shows a spectrum measured at 94.3 MeV. The 4.67- and 4.80-MeV  $\alpha$  groups have decreased in intensity relative to the 4.50-MeV  $\alpha$  peak, as expected from the yield data shown in Fig. 8. Since there is no sign of the peaks at 4.46 and 4.38 MeV, one must conclude that at 100 MeV they are produced independently and not by the decay of the 4.50-MeV  $\alpha$  group. Correspondingly, the 4.50-MeV  $\alpha$  group is not <sup>156</sup>Tm.

Additional corroboration for the assignment of the 4.50-MeV peak to  $^{157}$ Yb comes from the data obtained for the 4.67-MeV  $\alpha$  group. At bombarding energies close to 100 MeV this latter  $\alpha$  group, as indicated by its half-life and decay energy, is apparently due to the  $\alpha$  decay of <sup>153</sup>Er. Other assignments are unreasonable. Because of its large intensity (see Fig. 6) the 4.67-MeV peak cannot be assigned to either <sup>155</sup>Tm or <sup>156</sup>Tm; their  $\alpha$ -decay daughters, <sup>151</sup>Ho and <sup>152</sup>Ho, should but do not appear in the measured spectra with  $\sim \frac{1}{4}$  the intensity of the parent nuclides. The  $\alpha$  group cannot be assigned to  $^{157}$ Yb either because: (1) The yield points do not agree with the shape of the calculated (<sup>3</sup>He, 8n) excitation function, and (2) there was no discernible growth in the peak, which would have to be the case if <sup>157</sup>Yb and its  $\alpha$ -decay daughter, <sup>153</sup>Er, had the same decay energy. One must conclude that at energies ~100 MeV the bulk of the counts observed in the 4.67-MeV peak are due to <sup>153</sup>Er independently produced in a (<sup>3</sup>He,  $\alpha 8n$ ) reaction.

Because <sup>153</sup>Er has been reported<sup>3</sup> to be essentially a pure  $\alpha$  emitter, then for every <sup>157</sup>Yb  $\alpha$  decay one should see one  $\alpha$  event due to <sup>153</sup>Er. The actual counts observed at ~100 MeV (see Fig. 6) were such that the independently produced <sup>153</sup>Er obscured any counts that would have grown from the 4.50-MeV  $\alpha$  group, which we have assigned to <sup>157</sup>Yb. At energies below 90 MeV, count rates for both the 4.50- and 4.67-MeV groups were extremely low. An attempt was made with the wheel assembly and the 20 000-channel analyzer to observe growth in the 4.67-MeV  $\alpha$  peak at a <sup>3</sup>He en-



FIG. 9. Composite spectrum measured after 94.3-MeV  $^3\mathrm{He}$  bombardments of  $^{162}\mathrm{Er.}$ 

ergy of 90.3 MeV. Sixty 20-sec irradiations were made, each followed by 320 sec of counting.

The decay data from these experiments are shown in Fig. 10 for the 4.50- and 4.67-MeV peaks. As is seen in the lower portion of the figure, the 4.50-MeV  $\alpha$  activity has the same 34-sec half-life that was measured at higher bombarding energies. But the data for the 4.67-MeV  $\alpha$  peak, in the upper part of Fig. 10, no longer exhibit the characteristic half-life of <sup>153</sup>Er; instead, the apparent halflife is longer than 36 sec.

One could argue that the 4.67-MeV peak observed at 90.3 MeV is due not only to 36-sec <sup>153</sup>Er, but to some other nuclide as well, with a half-life of ~70 sec. The combination of these two components could lead to the experimental decay curve (Fig. 10), *if* the intensity of the longer component were comparable to that of <sup>153</sup>Er. One would expect to see this result reflected in the behavior of the ex-



FIG. 10. Decay data for the 4.50- and 4.67-MeV  $\alpha$  groups obtained after 60 bombardments, each 20 sec in duration, of <sup>162</sup>Er with 90.3-MeV <sup>3</sup>He ions. The curves labeled  $A_1(t)$  and  $A_2(t)$  are decay curves calculated as follows: (a)  $A_1(t)$  represents the 36-sec decay of <sup>153</sup>Er (4.67-MeV  $\alpha$  peak) present at the end of bombardment, and (b)  $A_2(t)$  represents the growth and decay of <sup>153</sup>Er consistent with the intensity of the 4.50-MeV  $\alpha$  group which we assign to <sup>157</sup>Yb. The sum of the two curves is seen to be in fair agreement with the data points for the 4.67-MeV  $\alpha$  group.

citation function for the 4.67-MeV peak (see Fig. 8). Nothing striking, however, occurs in the excitation function at 90 MeV to indicate that another nuclide besides <sup>153</sup>Er is making a marked contribution to the yield of the 4.67-MeV peak.

The explanation for the results in Fig. 10 that seems to be most consistent with all of the data is based on our assignment of the 4.50-MeV peak to the decay of <sup>157</sup>Yb. In Fig. 10 it is seen that the initial intensities of the 4.50- and 4.67-MeV peaks are comparable, so that the <sup>153</sup>Er growing from the  $\alpha$  decay of <sup>157</sup>Yb can have a striking effect upon the observed decay curve for the 4.67-MeV  $\alpha$  peak. (As noted earlier, at higher energies, where the <sup>153</sup>Er yield is much larger than that for <sup>157</sup>Yb, the effect of growth and decay upon the 4.67-MeV peak would be small.)

Because parent and daughter have approximately the same half-life, the equation for the growth and decay for  $^{153}$ Er has a special form,

$$A_d(t) = A_p(0)\lambda t e^{-\lambda t} .$$
<sup>(1)</sup>

Here  $A_d(t)$  is the daughter activity at some time t after the end of the irradiation,  $A_p(0)$  is the parent activity at the end of bombardment, and  $\lambda$  is the decay constant, which is taken to be equal for parent and daughter. Note that this expression contains the multiplicative factor  $\lambda t$ , so that it does not exhibit the simple exponential dependence upon t that is usually associated with radioactive decay.

The calculated growth and decay curve for <sup>153</sup>Er, consistent with the observed intensity of the 4.50-MeV peak that we assign to  $^{157}$ Yb, is labeled  $A_{2}(t)$ in Fig. 10. The curve labeled  $A_1(t)$ , with the characteristic 36-sec half-life, is that ascribed to the <sup>153</sup>Er present at the end of bombardment. [It is worth noting that the <sup>153</sup>Er is produced in two ways during the irradiation; by the (<sup>3</sup>He,  $\alpha 8n$ ) reaction and by decay of <sup>157</sup>Yb.] The sum of the two curves, labeled  $A_1(t) + A_2(t)$ , is seen to agree quite well with the data; at decay times >120 sec the growth of <sup>153</sup>Er from <sup>157</sup>Yb [curve  $A_2(t)$ ] has an important effect upon the shape of the observed decay curve. Thus, the data for the 4.67-MeV peak are certainly consistent with our assignment of the 4.50-MeV  $\alpha$  particle to <sup>157</sup>Yb.

Before going on to the next section some remarks should be made concerning the yields shown in Fig. 8. These data were not corrected for  $\alpha/\text{to-}$ tal branching ratios, since these are not known for <sup>156</sup>Yb and <sup>157</sup>Yb. (The value for <sup>153</sup>Er is reported<sup>3</sup> to be 0.95.) Based on previous information<sup>1,2,4</sup> for N = 86 and  $N = 87 \alpha$  emitters, their ratios should be between 0.01 and 0.001. Thus if branching ratios had been used to correct the data in Fig. 8, the <sup>156</sup>Yb and <sup>157</sup>Yb cross sections would be greater than that for <sup>153</sup>Er by a factor of 10-100. For the sake of completeness it should be added that the (<sup>3</sup>He,  $\alpha 7n$ ) and (<sup>3</sup>He,  $\alpha 6n$ ) products.<sup>154</sup>Er and <sup>155</sup>Er were also observed. While the  $\alpha$ /total branching ratio for <sup>155</sup>Er is not known, that for <sup>154</sup>Er has been reported<sup>10</sup> to be  $(1.7 \pm 1.0) \times 10^{-3}$ . When the <sup>153</sup>Er and <sup>154</sup>Er counting rates were corrected by their respective branching ratios it was found that the yield of <sup>154</sup>Er was ~30 times greater than that of <sup>153</sup>Er. This means then that the yield for the (<sup>3</sup>He,  $\alpha 7n$ )<sup>154</sup>Er reaction is comparable to the yields for the (<sup>3</sup>He, 8n) and (<sup>3</sup>He, 9n) reactions. Thus the yield data shown in Fig. 8 agree with the results obtained with the <sup>156</sup>Dy target, i.e., cross sections for (<sup>3</sup>He,  $\alpha xn$ ) reactions are as large or larger than those for (<sup>3</sup>He, xn) reactions.

### V. α-DECAY ENERGY SYSTEMATICS

The discovery of previously unknown isotopes is of significance because it contributes to the understanding of the systematics of radioactive decay and provides measurements of decay energies which can then be used to determine precise values of nuclear masses and binding energies.  $\alpha$ decay energies available<sup>4</sup> for rare-earth nuclei are plotted in Fig. 11 as a function of neutron number. Decay energies obtained for the new isotopes reported here and in Refs. 1 and 2 are indicated by large points. With the exception of the data shown for N = 83 nuclides, only experimentally determined decay energies are plotted in Fig. 11. Energies given for <sup>147</sup>Gd, <sup>145</sup>Sm, and <sup>143</sup>Nd were calculated by means of closed energy cycles. The method consists of constructing an energy-balance cycle from two  $\alpha$ - and two  $\beta$ -decay energies. If three of the four pieces of information that constitute a cycle are known, then the fourth can be calculated. In all three instances the  $\alpha$ -decay energy of a given N = 83 isotope is ~1.5 MeV less than that of the corresponding N = 84 nuclide. (In Fig. 11 the <sup>151</sup>Er  $\alpha$ -decay energy is thus estimated to be ~1.5 MeV less than that of <sup>152</sup>Er.) This sudden drop in energy shows the influence of the closed 82-neutron shell. As a result of the major closed shell, the maximum  $\alpha$ -decay energy for a given element is reached at 84 neutrons, owing to the abnormally low neutron binding energies just beyond the closed shell.

In addition to the trend of increasing decay energies with decreasing neutron number (up to 84) there is an indication of a pairing effect similar to the one that has been noted<sup>13</sup> for  $\alpha$  emitters in the N = 126 region. While nuclides with N = 84 and N= 85 have similar decay energies, the additional pair of neutrons in N = 86 nuclides seems to result in added stability and a consequent extra decrease in  $\alpha$ -decay energy. The effect is even more pronounced in going from N = 87 to N = 88 nuclides, and



Fig. 11.  $\alpha$ -decay energies for rare-earth nuclei plotted as a function of neutron number. All decay energies are experimentally measured ones except those for the 83-neutron nuclides <sup>147</sup>Gd, <sup>145</sup>Sm, and <sup>143</sup>Nd. These were calculated from closed energy cycles. Note that in all three instances these energies are ~1.5 MeV less than those for the corresponding 84-neutron isotopes. Consequently, the energy for <sup>151</sup>Er is estimated to be 1.5 MeV less than that for <sup>152</sup>Er. Large points in Fig. 11 indicate isotopes first reported here and in Refs. 1 and 2.

would account for the fact that a rather extensive search<sup>1</sup> for  $\alpha$  activity due to the 88-neutron nuclide, <sup>155</sup>Ho, proved fruitless. A straight line drawn through the energies for <sup>151,152,153,154</sup>Ho vields a decay energy for  $^{155}$ Ho or  $\sim 3.7$  MeV. Based on the sharp drop in energy between the 87- and 88-neutron isotopes of dysprosium, however, one would estimate the <sup>155</sup>Ho  $\alpha$ -decay energy to be only ~3.3 MeV. A decrease of 0.4 MeV in  $\alpha$ -decay energy leads to a greatly increased  $\alpha$ -decay half-life which in turn makes the  $\alpha$ /total branching ratio for <sup>155</sup>Ho much less than expected. The one clear exception in Fig. 11 to this pairing effect involves <sup>150</sup>Tb. This odd-odd nucleus has an unknown ground-state spin. From decayscheme studies  $^{14}$  the level structure in its  $\alpha\text{-decay}$ daughter, <sup>146</sup>Eu, has been proposed to be: 0 keV  $(4^+)$ , 115 keV  $(3^+)$ , 230 keV  $(2^+)$ , and 375 keV  $(1^+)$ . If the ground-state spin of <sup>150</sup>Tb were low, then  $\alpha$  decay would proceed to one of the low-spin excited states in <sup>146</sup>Eu rather than to the 4<sup>+</sup> ground state. The observed <sup>150</sup>Tb  $\alpha$  decay would therefore not correspond to the actual energy difference between the two nuclei.

One other point should be made concerning Fig. 11. On examination of the N = 84 nuclei one notes a "break" between the gadolinium (Z = 64) and terbium (Z = 65) nuclides. This "break" had been observed earlier<sup>15</sup> when it was suggested that the 65th proton is less tightly bound than the other protons in the region. The effect correlates nicely with the fact that there is a proton subshell occurring at Z = 64, where the  $d_{5/2}$  orbital is closed and the next available orbital is the  $h_{11/2}$ . As a result of the splitting between the two subshells, there would be an extra stabilization for the 64-proton configuration.

The decay characteristics of the three new isotopes found in this investigation are summarized

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 $^1\mathrm{R.}$  L. Hahn, K. S. Toth, and T. H. Handley, Phys. Rev. 163, 1291 (1967).

- <sup>2</sup>K. S. Toth, R. L. Hahn, M. F. Roche, and D. S. Brenner, Phys. Rev. 183, 1004 (1969).
- <sup>3</sup>R. D. Macfarlane and R. D. Griffioen, Phys. Rev. <u>131</u>, 2176 (1963).
- <sup>4</sup>P. Eskola, Arkiv. Fysik <u>36</u>, 477 (1966).
- ${}^{5}$ R. D. Macfarlane and R. D. Griffioen, Nucl. Instr. Methods 24, 461 (1963).
- <sup>6</sup>A. Ghiorso, K. Valli, and M. Nurmia, private com-

munication.

<sup>7</sup>I. Dostrovsky, Z. Fraenkel, and G. Friedlander, Phys. Rev. <u>116</u>, 683 (1960).

<sup>8</sup>R. D. Macfarlane and R. D. Griffioen, Phys. Rev. <u>130</u>,

TABLE II. Decay information for new isotopes.

Isotope	Half–life (sec)	α-decay energy (MeV)
<sup>151</sup> Er	$23 \pm 2$	no $\alpha$ emission
<sup>156</sup> Yb	$24 \pm 1$	4.80 ± 0.01
<sup>157</sup> Yb	$34 \pm 3$	4.50 ± 0.01

in Table II.

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1491 (1963).

- <sup>9</sup>See, for example, G. B. Saha and N. T. Porile, Phys. Rev. 149, 880 (1966).
- <sup>10</sup>N. A. Golovkov, S. K. Khvan, and V. G. Chumin, in Proceedings of the International Symposium on Nuclear Structure, Dubna, 1968 (International Atomic Energy

Agency, Vienna, Austria, 1969), p. 27.

<sup>11</sup>I. R. Williams and K. S. Toth, Phys. Rev. <u>138</u>, B382 (1965).

<sup>12</sup>R. D. Macfarlane, Phys. Rev. <u>136</u>, B941 (1964).

<sup>13</sup>See, for example, K. Valli, University of California Lawrence Radiation Laboratory Report No. UCRL 17723, 1967 (unpublished).

<sup>14</sup>M. J. Martin, Nucl. Data B2, (No. 4), 1 (1967).

<sup>15</sup>J. O. Rasmussen, S. G. Thompson, and A. Ghiorso, Phys. Rev. <u>89</u>, 33 (1953).