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New Rare-Earth Alpha Emitter, ^{148}Eu†

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A new alpha emitter with a decay energy of 2.63±0.03 MeV was discovered and assigned to the nuclide ^{148}Eu. The α/E.C. branching ratio was measured to be $(9.4\pm2.8)\times10^{-9}$, corresponding to a partial alpha half-life of $(1.6\pm0.5)\times10^{7}$ years. By using a Coulomb barrier penetration formula and the measured alpha-decay energy, a half-life of $(6.2_{-3.3}^{+6.7})\times10^{6}$ years was calculated. The agreement, within experimental error, between the measured and calculated partial alpha half-lives indicates that the ground-state spin and parity assignments are probably the same for ^{148}Eu and its daughter ^{144}Pm.

INTRODUCTION

ONE of the effects of the 82-neutron closed shell is to enhance the alpha-decay energies of the 84-neutron nuclides. The enhancement of decay energies extends to neighboring isotopes with slightly higher neutron numbers. The only europium alpha emitter known from previous work[1] is the 84-neutron nuclide ^{147}Eu. From rare-earth alpha-decay systematics[2] the 54-day 85-neutron isotope ^{148}Eu has been predicted to have a total alpha disintegration energy (measured alpha particle energy plus recoil energy plus orbital electron screening correction) of ~2.75 MeV. This predicted decay energy corresponds to a partial alpha half-life of approximately 10^7 years. Since the resultant α/E.C. branching ratio of $\sim10^{-8}$ is within present detection limits, a search was initiated for the alpha decay of ^{148}Eu.

EXPERIMENTAL METHOD

The ^{148}Eu activity was produced by bombarding for 8-h periods 10-mg targets of isotopically enriched samarium oxide with protons accelerated in the Oak Ridge National Laboratory 86-in. cyclotron. The maximum energy proton beam ($\approx$22 MeV) was used to produce ^{148}Eu in the reaction $^{149}\mathrm{Sm}(p,2n)$. By means of standard ion-exchange techniques, the europium fraction was chemically separated from the irradiated samarium. Samples for alpha counting were prepared by evaporating the eluant on platinum discs and then flaming off the excess organic matter.

The alpha particles were counted with a surface-barrier semiconductor detector. Although the detector and biased amplifier used in the experiment were capable of 20-keV resolution, the low counting rates encountered prevented the use of the detection system with this resolution. The alpha spectra were accumulated in a 256-channel pulse-height analyzer so that the apparent width of the ^{241}Am line was ~40 keV. The geometry was calculated for the finite source size and detector aperture; it was also measured by counting an ^{241}Am source with a known disintegration rate. The detector arrangement was thus found to have a geometry factor of $(4\pi/7.6\pm0.6)$. A 1.5×1.5-in. NaI crystal spectrometer was used to measure the γ-ray spectra. A relative photopeak efficiency curve for this NaI crystal was computed from the known decay characteristics of standard γ-ray sources; the curve was then made absolute by determining the disintegration rate of three of the sources. A more complete discussion of the procedure has been presented earlier.[3] The error in the absolute photopeak efficiencies is estimated to be 15%.

MEASUREMENTS

To investigate the sensitivity of the experimental equipment, a sample of ^{147}Eu was prepared by the $(p,2n)$ reaction on ^{148}Sm. The ^{147}Eu alpha-decay energy was measured to be 2.89±0.02 MeV; an ^{241}Am source

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[1] J. O. Rasmussen, S. G. Thompson, and A. Ghiorso, Phys. Rev. 89, 33 (1953).

[2] K. S. Toth and J. O. Rasmussen, Nucl. Phys. 16, 474 (1960).

[3] E. Newman and K. S. Toth, Phys. Rev. 129, 802 (1963).

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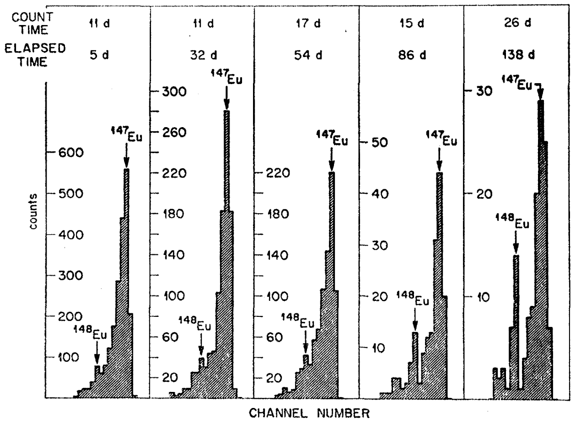

FIG. 1. Portions of alpha-particle spectra taken with the thin ^{148}Eu source during a 6-month period. In addition to the ^{147}Eu alpha particles, another peak is visible at a lower energy.

was used as an energy standard. This value is in agreement with the more accurate measurement of Siivola[4] who has reported the ^{147}Eu alpha-decay energy to be 2.91 ± 0.01 MeV. The total number of disintegrations of the ^{147}Eu sample was determined from the counting rate of the 122- and 198-KeV γ rays following the electron-capture decay of ^{147}Eu. The ^{147}Eu decay scheme published by Schwerdtfeger *et al.*[5] was used as a guide to determine what percentages of the total number of decays were represented by the two γ rays. The α/E.C. branching ratio for ^{147}Eu was in this manner found to be $(1.8 \pm 0.9) \times 10^{-5}$, which is in satisfactory agreement with Siivola's value[4] of $(2.2 \pm 0.6) \times 10^{-5}$.

The enrichment of the samarium oxide was 97.5% in ^{149}Sm, with the remaining 2.5% distributed among the other samarium isotopes. As a result, the ^{148}Eu samples also contained small amounts of ^{147}Eu. However, since the ^{147}Eu alpha branching ratio was expected to be $\sim 10^3$ times greater than that of ^{148}Eu, it was not surprising to find ^{147}Eu alpha activity dominant in the ^{148}Eu samples.

A strong ^{148}Eu source was found initially to be too thick to distinguish any ^{148}Eu alpha particles from those of ^{147}Eu. Thus, it was placed aside and a thin, weak source (6×10^6 dis/min) was prepared. The alpha spectrum of the weak source was studied during a period of six months; the results obtained at various times after sample preparation are shown in Fig. 1. To compensate for the electronic drift during the long periods of counting and for the poor statistics, the spectra are plotted as histograms where the number of counts from adjacent pairs of channels are added. In addition to the ^{147}Eu alphas, the spectra show a second peak whose half-life is longer than that of ^{147}Eu. By the use of the ^{241}Am source and by assuming Siivola's value of 2.91 MeV for the ^{147}Eu alphas, the decay energy of the less

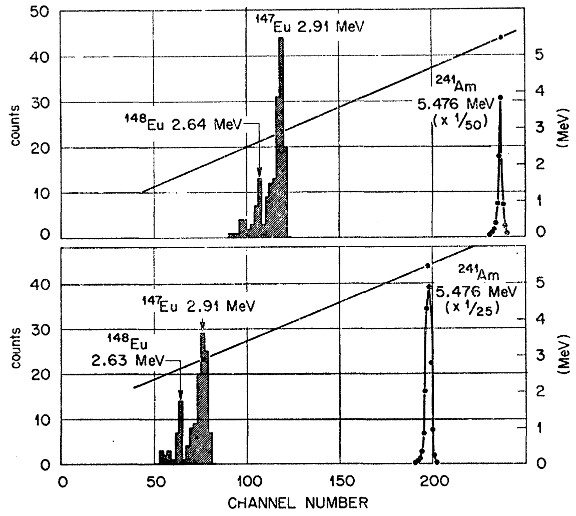

FIG. 2. Spectra showing the energy determination of the lower energy alpha peak.

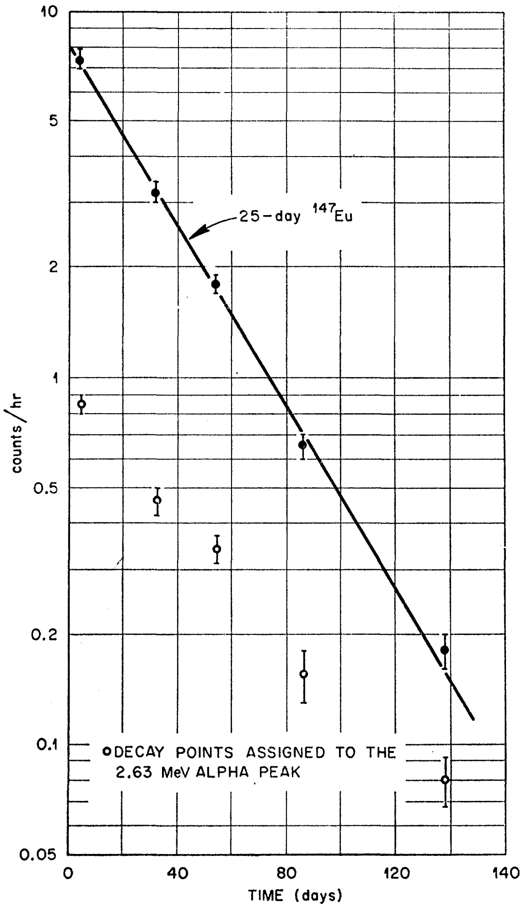

FIG. 3. Decay of the two alpha peaks observed in Fig. 1. The 2.91-MeV alphas decay with the accepted 25-d ^{147}Eu half-life. The lower energy peak displays a half-life which is longer than that of ^{147}Eu.

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[4] A. Siivola, Ann. Acad. Sci. Fennicae, Ser. A, **VI**, 109 (1962).

[5] C. F. Schwerdtfeger, H. J. Prask, and J. W. Mihelich, Nucl. Phys. **35**, 168 (1962).

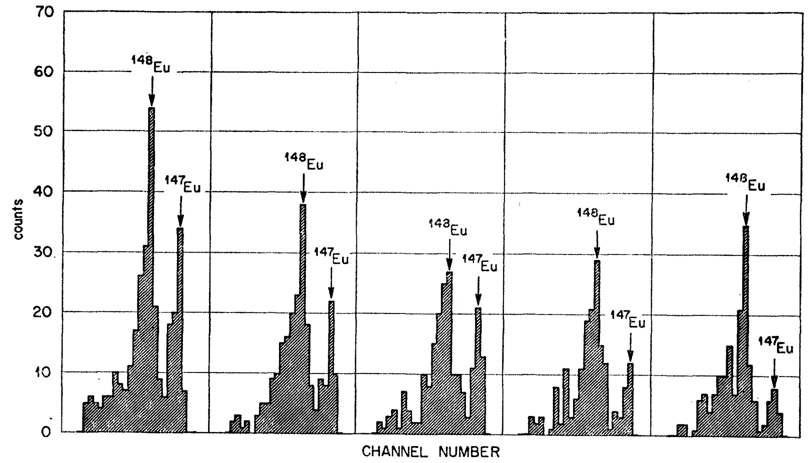

FIG. 4. Portions of alpha-particle spectra taken with the thick ^{148}Eu source seven months after its preparation. Five consecutive 8-day counts were taken over a period of 6 weeks. The lower energy 2.63-MeV peak is now the dominant activity.

intense peak was measured to be 2.63±0.03 MeV. Two of the energy determinations are illustrated in Fig. 2. The apparent full width at half-maximum of the ^{241}Am peak is ~40 keV; that of the europium peaks, as plotted in Figs. 1 and 2, is ~100 keV.

Decay curves (see Fig. 3) for the two alpha peaks were determined from the data shown in Fig. 1. Since, in the earlier spectra, the two peaks are incompletely resolved, the total number of counts in a fixed set of channels are plotted as a function of time. The ^{147}Eu peak decays with the correct 25-day half-life. The decay curve for the less intense peak shows that, to begin with, a large amount of ^{147}Eu activity is present in that particular set of channels; later the decay curve displays a half-life which is longer than that of ^{147}Eu.

Seven months after the preparation of the strong ^{148}Eu source, its alpha spectrum was studied over a period of six weeks to determine the half-life of the 2.63-MeV peak. Two peaks were again visible in the alpha spectrum (see Fig. 4). The decay curve of the lower-energy peak is shown in Fig. 5, together with a least squares fit to the decay points. The best fit to the experimental points was found to be 50 days, with a standard deviation of 11 days.

While the measurements described above were being made, a ^{148}Sm target was irradiated at an energy below the $(p,2n)$ threshold to assure production of ^{148}Eu by the (p,n) reaction. An extensive study of this new source was not undertaken; but it was clear that its alpha spectrum was similar to that observed in the ^{149}Sm$(p,2n)$ irradiation. In particular, the 2.63- and 2.91-MeV peaks were both present and the lower energy peak decayed with a half-life which was longer than that of the ^{147}Eu 2.91-MeV alphas.

The 2.63-MeV alpha particle is assigned to the decay of ^{148}Eu on the basis of the following evidence:

(1) The mass and atomic numbers are established because the alpha emitter was observed in chemically purified europium fractions prepared from two separate bombardments, ^{148}Sm(p,n) and ^{149}Sm$(p,2n)$. In each instance, the γ-ray spectrum showed that ^{148}Eu was by far the predominant activity in the source. In addition a ^{147}Eu sample prepared from a ^{148}Sm$(p,2n)$ bombardment showed, within the apparatus detection limits, no sign of the 2.63-MeV alphas.

(2) The half-life of the 2.63-MeV peak was found to be 50±11 days, in agreement with the accepted 54-day half-life of ^{148}Eu.

(3) The measured alpha-decay energy corresponds closely to the value predicted for ^{148}Eu from rare-earth alpha-decay systematics[2] (see discussion in the following section).

The total number of ^{148}Eu disintegrations in each sample was determined from the counting rates of the three prominent γ-rays (414, 551, and 631 keV) following the electron-capture decay of ^{148}Eu. The ^{148}Eu decay scheme proposed by Schwerdtfeger *et al.*[6] was used to convert the absolute γ-ray counting rates into total distintegration rates. In this manner, the ^{148}Eu α/E.C.

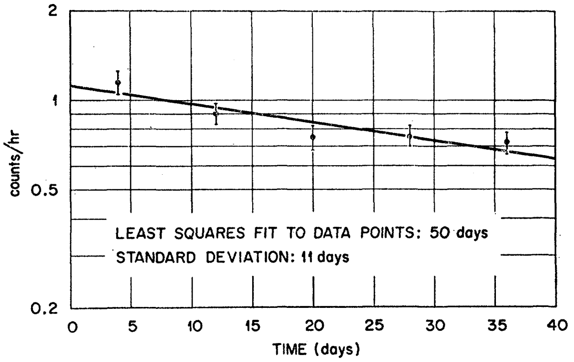

FIG. 5. Decay of the 2.63-MeV peak as derived from the data shown in Fig. 4. The least squares fit to the points is a 50-day half-life with a standard deviation of 11 days.

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[6] C. F. Schwerdtfeger, E. G. Funk, Jr., and J. W. Mihelich, Phys. Rev. **125**, 1641 (1962).

branching ratio was measured to be $(9.4\pm2.8)\times10^{-9}$, corresponding to a partial alpha half-life of $(1.6\pm0.5)\times10^{7}$ years. The experimental uncertainty was estimated on the basis of errors present in the NaI crystal efficiency, the alpha-counter geometry determination, and the statistical fluctuations in the low alpha counting rates that were encountered in this study.

DISCUSSION

From a consideration of alpha-decay systematics in rare-earth elements with $Z\leq66$,[2] the ^{148}Eu total alpha-disintegration energy Q_{eff} of the bare nucleus has been predicted to be 2.75 MeV. By subtracting from Q_{eff} the orbital electron screening correction ($\sim$20 keV for the rare-earth nuclides) and the recoil energy, the predicted alpha-decay energy in the laboratory system is found to be 2.65 MeV. This decay energy agrees with the value of 2.63 ± 0.03 MeV measured in the present investigation. Another prediction from the same discussion of alpha-decay systematics[2] has been found to agree well with a subsequent experimental measurement. The Q_{eff} for ^{145}Pm was predicted to be 2.30 MeV.[2] This corresponds to a laboratory energy of 2.22 MeV, which in turn agrees with the experimental value of 2.24 ± 0.04 MeV measured by Nurmia *et al.*[7]

In the heavy element region, half-lives for alpha-decay transitions from even-even nuclides to the ground states of the daughter nuclei have been found to agree well with those calculated from simple barrier penetration theory.[8] Transitions from odd-nucleon isotopes and from even-even nuclides to excited states in their daughter nuclei are ordinarily found to have decay rates which are slower than those calculated from this theory. Hinderance factors can be defined for the slower transitions as being the ratios of experimental to calculated half-lives.

The experimental decay energy, 2.63 ± 0.03 MeV, was used in a simple Coulomb barrier penetration formula (see Ref. 2 for a discussion of the expressions employed in the calculation), and a partial alpha half-life of $(6.2_{-3.3}^{+6.7})\times10^{6}$ years was determined for ^{148}Eu. The rate formula used in the calculation was designed to give perfect agreement with ^{148}Gd[2] and not a general average agreement for all the even-even rare-earth alpha emitters. Within the errors involved in the measurements of the branching ratio and decay energy, this calculated half-life value is not significantly different from the experimental value $(1.6\pm0.5)\times10^{7}$ years. The implication then is that ^{148}Eu alpha decay is not significantly hindered.

The daughter and parent nuclei, ^{144}Pm and ^{148}Eu, each have an odd $f_{7/2}$ neutron and an odd $d_{5/2}$ proton if it is assumed that the order of filling of the shells is that given by Mottelson and Nilsson for the case of zero deformation.[9] While the ground-state spins could therefore be anywhere from 1 to 6 with a negative parity, published results indicate that the spin assignment is $5(-)$ or $6(-)$ for both ^{144}Pm (Refs. 10 and 11) and ^{148}Eu (Ref. 6), although not necessarily the same for the two isotopes. The lack of a large amount of hindrance in ^{148}Eu alpha decay indicates a favorable overlap between the initial- and final-state wave functions; the present results, therefore, suggest that ^{144}Pm and ^{148}Eu have the same ground-state spins and parities.

The only rare-earth isotopes exhibiting alpha-decay hindrances which cannot be accounted for by experimental errors are the two known terbium alpha emitters. The hindrance factors for ^{149}Tb and ^{151}Tb are 7 and 140, respectively.[2] While ^{152}Tb alpha decay has not been found, a lower limit on its alpha half-life has been set, i.e., $>2.2\times10^{4}$ years.[2,12] From systematics, the ^{152}Tb Q_{eff} is estimated to be about 3.2 MeV, corresponding to a half-life of 3.95×10^{3} years. Therefore, the ^{152}Tb hindrance factor is >5.5. Among the remainder of the known alpha emitters in the 82-neutron region, the decay rates of odd-nucleon isotopes are comparable to those of neighboring even nuclides. Qualitatively, the explanation might lie in the fact that the terbium nuclides are beginning to fill in the $h_{11/2}$ proton shell while the daughter europium isotopes are completing the $d_{5/2}$ shell. The overlap of the initial- and final-state wave functions is thus not expected to be favorable. The decay of the other known rare-earth odd-nucleon alpha emitters is such that the initial and final odd nucleons are in the same neutron or proton configurations, with resulting favorable overlap integrals.

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[7] M. Nurmia, P. Kauranen, and A. Siivola, Phys. Rev. **127**, 943 (1962).

[8] I. Perlman and J. O. Rasmussen, *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1957), Vol. 42, p. 143.

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