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Reactions Induced by Slow Neutron Irradiation of Europium

RICHARD J. HAYDEN, JOHN H. REYNOLDS,* AND MARK G. INGHRAM
Argonne National Laboratory and University of Chicago, Chicago, Illinois

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A sample of europium which had been subjected to slow neutron irradiation was analyzed using mass spectrometric techniques. By using a modified isotopic dilution technique, i.e., adding a standard containing known relative abundances of normal samarium, europium, and gadolinium the complete chemical and isotopic analysis of the irradiated sample was obtained. Combining this nuclide composition and certain radioactivity measurements the thermal neutron cross sections, half-lives and branching ratios of five of the europium isotopes were calculated. The results were $\sigma_{151}=5200$ barns, $\sigma_{152}=3200$ barns, $\sigma_{153}=240$ barns, $\sigma_{154}=880$ barns, $\sigma_{155}=7900$ barns, $T_{\frac{1}{2}}(152)=5.3$ years, $T_{\frac{1}{2}}(154)=5.4$ years, $T_{\frac{1}{2}}(155)=1.7$ years, K/β^- ratio for 9.2-hr. $\mathrm{Eu}^{152}=0.22$, K/β^- ratio for 5.3-year $\mathrm{Eu}^{152}=2.8$, and K/β^- ratio for 5.4-year $\mathrm{Eu}^{154}<0.05$. All these results are believed to be good to ±15 percent.

INTRODUCTION

SLOW neutron induced activities in europium have been studied by several investigators.[1-6] However, because of the complex nature of the radiations induced a more complete analysis of the problem is desirable. Of special interest are the following problems: (1) The determination of the ratio of K-capture to beta-decay for the (n, γ) induced activities of europium. Previous investigation of these branching ratios have been based on a determination of x-ray counter efficiencies. (2) The determination of the characteristic radiations of the long lived Eu^{152} and long lived Eu^{154}. This problem exists since up to this time all studies of these radiations have been made on a mixture of these two long lived activities. (3) The determination of the half-lives of Eu^{152} and Eu^{154}. These measurements have been heretofore uncertain because of the length of both half-lives and because of the intimate mixture of the two isotopes. (4) The determination of the neutron absorption cross sections of the stable and active europium isotopes. This investigation was undertaken to provide answers to these problems.

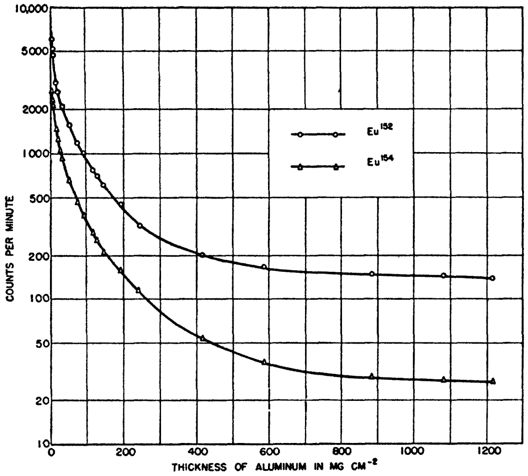

FIG. 1. Aluminum absorption curves taken from mass spectrographically separated samples of 5.3-year Eu^{152} and 5.4-year Eu^{154}.

I. SAMPLE AND IRRADIATIONS

The europium used in this analysis was an $\mathrm{Eu_2O_3}$ sample prepared by Dr. J. K. Marsh of Oxford. It was obtained from Johnson, Matthey and Company who distribute it as Laboratory Number 11,158, Catalog Number F.606. The spectrographic analysis supplied with the material stated that ytterbium was the only rare earth impurity detectable. A mass spectrometric analysis showed additional impurities of the order of one part in 10,000 of samarium, gadolinium, dysprosium, lanthanum, and praseodymium. With the exception of the lanthanum, the effects of which will be later discussed, these impurities did not effect the accuracy of measurement. Two separate neutron irradiations of portions of this material were made. The first of these was a short bombardment in the thermal neutron flux of the Argonne heavy water pile. From this bombardment the mass assignment of the 9.2-hr. activity was obtained. The second sample was subjected to long slow neutron irradiation in a graphite moderated pile. All the other results reported in this paper were obtained from this sample.

II. MASS ASSIGNMENT OF EUROPIUM ACTIVITIES

The mass of radioactive isotopes may be determined by separating the isotopes of an active sample by means of a mass spectrograph and investigating the activity corresponding to each

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* National Research Council Fellow.

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

[2] W. Z. Bothe, Zeits. f. Naturforschung, **1**, 179 (1946).

[3] R. J. Hayden and M. G. Inghram, Phys. Rev. **70**, 89 (1946).

[4] M. G. Inghram and R. J. Hayden, Phys. Rev. **71**, 130 (1947).

[5] J. M. Cook, R. G. Shreffler, and C. M. Fowler, Phys. Rev. **72**, 1209 (1947); **73**, 78 (1948).

[6] Franklin B. Shull, Phys. Rev. **74**, 917 (1948).

mass position. This method was employed in our investigation of the europium activities. The mass spectrograph and techniques employed have been described by Lewis and Hayden.[7,8] Briefly the method is as follows: A portion of the material to be investigated is irradiated with neutrons to obtain the desired activity. The sample is then dissolved in nitric acid and an aliquot placed on the filament source of the mass spectrograph. By operation of the spectrograph the isotopes are separated according to mass and deposited on a photographic plate called the original plate. After removal from the spectrograph this plate is placed face to face with a second photographic plate called the transfer plate. With the passage of time the radioactive decay particles emitted from the active isotopes on the original plate give rise to developable images on both plates. Upon development the original plate shows the active lines and the normal mass spectrum of the irradiated element, while the transfer plate shows only the radioactive lines. By comparison of these two plates and, if necessary, by observation of the radioactive decay at the various mass positions, the masses of the active isotopes produced by the irradiation can be deduced.

In order to determine the mass number of the 9.2-hr. europium, first reported by Pool and Quill,[1] a sample of europium was irradiated with slow neutrons for eight hours in the Argonne heavy water pile.[3] A longer irradiation would have brought up the long lived europium activities in the sample without appreciably increasing the amount of 9.2-hr. activity. A portion of this irradiated material was run through the mass spectrograph and original and transfer plates were obtained. A decay curve from another aliquot of the irradiated sample obtained while the transfer was taking place, showed that at least 95 percent of the activity of the sample was of 9.2-hr. half-life. A comparison of the two plates showed that the activity deposited is of mass number 152. The simple decay curve showed that the activity in the sample and hence the activity deposited was of 9.2-hr. half-life. The ratio of ion intensity in the oxide to the metal position proved that the 9.2-hr. element was europium. Thus we conclude that the 9.2-hr. activity is associated with a europium isotope of mass number 152.

In an attempt to assign the 5–8-year europium activity first reported by Scheichenberger to a specific isotope it was discovered that there were two long lived (n, γ) induced activities in europium, one at mass number 152 and one at mass number 154. This preliminary work has been previously reported by the authors.[4] For the present work the europium sample which had been subjected to long slow neutron bombardment was separated on a photographic plate by the mass spectrograph and two transfer plates were obtained six months apart. The transfer plate obtained shortly after the end of the irradiation showed activity at masses 152, 154, 155, and 156. The later transfer showed lines only at masses 152, 154, and 155. The fact that the 156 line does not appear in the later transfers proves that this activity has a half-life of less than one month. The next paragraph will describe further work on this isotope. The fact that the 152, 154, and 155 lines did not change between the two exposures proves that the half-lives of these isotopes is considerably greater than one month. The Eu^{155} is therefore quite probably the two-year europium first discovered in fission by Winsberg[9] and shown to be a result of Eu^{155} by one of the authors.[8] In this case, however, the activity was produced by a second-order (n, γ) reaction on Eu^{153}. Previous estimates of the half-lives of Eu^{152} and Eu^{154} were Fajans and Voigt's value of 5–8 years[10] and Krisberg, Pool, and Hibdon's value of greater than 20 years.[11] Both of these measurements, however, were made on a mixture of Eu^{152} and Eu^{154} of unknown relative concentration. Values for the half-lives of these isotopes and for Eu^{155} are deduced later in this paper.

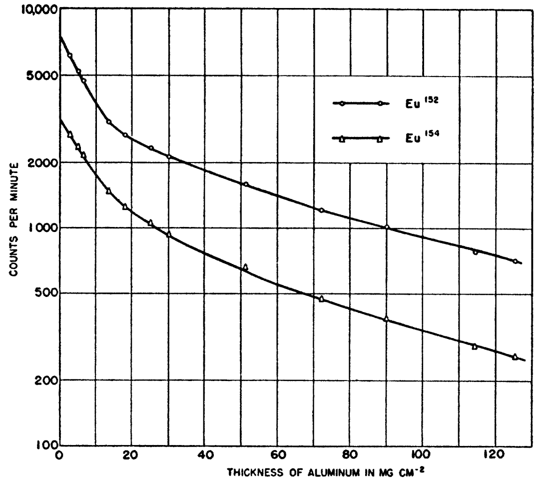

FIG. 2. Aluminum absorption curves taken from mass spectrographically separated samples of 5.3-year Eu^{152} and 5.4-year Eu^{154}. Expanded scale.

Winsberg,[12,13] working with fission europium found an activity of 15.4-day half-life which he

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[7] L. G. Lewis and R. J. Hayden, Rev. Sci. Inst. **19**, 599 (1948).

[8] Richard J. Hayden, Phys. Rev. **74**, 650 (1948).

[9] L. Winsberg, Manhattan Project Report CC-2000 (August, 1944).

[10] K. Fajans and A. F. Voigt, Phys. Rev. **60**, 533 (1941).

[11] N. L. Krisberg, M. L. Pool, and C. T. Hibdon, Phys. Rev. **74**, 44 (1948).

[12] L. Winsberg, Manhattan Project Report CN-2126 (September, 1944).

[13] L. Winsberg, Manhattan Project Report CC-2310, pp. 231–244 (January, 1945).

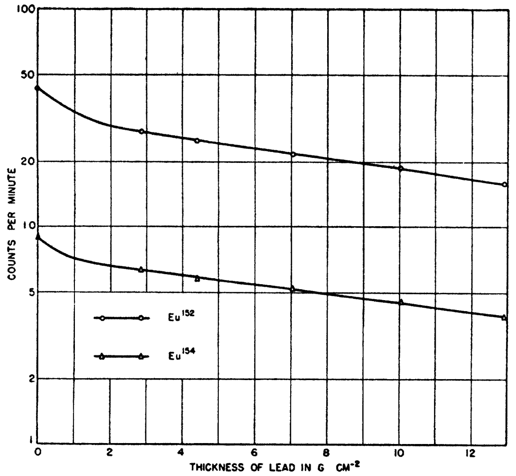

FIG. 3. Lead absorption curves taken from mass spectrographically separated samples of 5.3-year Eu^{152} and 5.4-year Eu^{154}. Zero thickness corresponds to 1000 mg cm^{-2} of aluminum shielding at the counter.

ascribed to Eu^{156}. To verify that the Eu^{156} formed in neutron bombardment of normal europium has a half-life of 15.4 days, a series of transfers were taken from one original plate. The duration of these successive exposures were such as to give a constant number of disintegrations at the mass 156 position if the half-life were 15.4 days. The fact that the intensities at masses 152, 154, and 155 increased while that at 156 remained constant proved that half-life of the europium deposited at mass 156 on the plate is 15.4±4 days. This is, therefore, the same activity reported by Winsberg as a fission product, and proves conclusively that the 15.4-day activity is associated with a europium isotope of mass number 156.

III. CHARACTERISTIC RADIATIONS OF LONG LIVED Eu^{152} AND LONG LIVED Eu^{154}

The characteristic radiations of the long lived activities formed by slow neutron irradiation of europium have been the subject of several investigations.[2,5,6] In all cases the sample studied contained appreciable amounts of both long lived Eu^{152} and long lived Eu^{154}, so that correlation of a particular radiation with a specific isotope was difficult. Shull, in the most extensive of these investigations,[6] used a magnetic two-directional focusing beta-ray spectrometer to obtain the beta-spectrum, the conversion line spectrum, and the photoelectron spectrum. He reported ten conversion lines in the combined activities with the strongest intensities at 73.1, 74.4, 114.0, and 119.6 kev and nine γ-transitions, including γ-rays at energies 0.243, 0.342, 0.959, 1.082, and 1.402 Mev. A Fermi plot of the continuous beta-ray spectrum indicated the presence of two beta-groups with energies of 0.75 and 1.6 Mev. Shull concluded that "The low energy conversion lines observed in 5–8-year europium, which are probably caused by two transitions of approximately equal energy must definitely be associated with Eu^{152}, not Eu^{154}," and "The 0.751-Mev β^--spectrum of the 5–8-year europium must also be associated with Eu^{152}." It is the purpose of this section to describe measurements made of the characteristic radiations of separated isotopes of Eu^{152} and Eu^{154}.

For the study, the isotopes of a portion of the heavily irradiated europium sample were separated with the 60° mass spectrograph in a manner similar to that used for determining the mass assignments. However, instead of the separated isotopes being deposited on a photographic plate, they were deposited on thin aluminum. The locations of the active deposits on this aluminum were then obtained by means of the photographic transfer technique, and the activities completely isolated by mechanically cutting the aluminum between the active deposits. A subsequent photographic transfer served as a check on the accuracy of the cuts. The samples of Eu^{152} and Eu^{154} thus obtained were introduced separately into a vertical lead shield end window counter system of standard design and the absorption curves shown in Figs. 1, 2, and 3 obtained. In the course of this work, three such isotope separations were made; the results were identical in all three cases.

In order to determine the degree of separation, for example, the amount of Eu^{152} activity scattered to the Eu^{154} position, a section of the original aluminum plate two mass units below the Eu^{152} mass position was isolated and counted; the counting rate was less than one percent of the total Eu^{152} activity so that for counting purposes the isolated deposits were "pure."

The aluminum absorption curves shown in Fig. 1, and in expanded scale in Fig. 2 were obtained under identical conditions in an end window counter of 2.6 mg thickness and a calculated geometry of about 16 percent. The lead absorption curves shown in Fig. 3 were obtained under identical conditions in an end window counter of 2.6 mg thickness and a calculated geometry of 5 percent. In order to suppress all betas the counter window was here shielded with 1000 mg/cm^2 of aluminum.

The first conclusion which may be drawn from these curves is that both radiations are complex and that not all the γ-rays are associated with the Eu^{152} as was suggested by Shull.

Secondly, from Fig. 2 it is evident that both Eu^{152} and Eu^{154} have soft components with ranges of less than 30 mg/cm^2 corresponding to energies of less than 165 kev. These are undoubtedly the strong conversion lines reported by Shull. The result, however, is contrary to the first of Shull's conclusions, i.e., that the low energy conversion lines must definitely be associated with Eu^{152}. Further, in the case of Eu^{154} where, as will be shown later, the fraction of K-capture is less than 5 percent of the β^--disintegrations, there must be more than one soft conversion electron per disintegration. Otherwise it is difficult to account for the high ratio at zero thickness of total counts to extrapolated higher energy counts. Thus if we assume, as seems reasonable from Shull's work, that the 74.4- and 114.0-kev conversion lines are due to Eu^{152}, it appears probable that the 73.1- and 119.6-kev conversion lines are associated with the decay of Eu^{154}.

Referring to Fig. 1, it is evident that the beta-end points are obscured both in Eu^{152} and Eu^{154} by semi-penetrating γ-rays so as to preclude any beta-energy assignments. From intensity considerations however, using Shull's data for the ratio of the 0.751 to 1.6-Mev betas, it is quite reasonable to assign the former to Eu^{152} and the latter to Eu^{154}.

From the lead absorption curves it is apparent that both radiations have at least two high energy γ-rays. The half-thickness of the high energy Eu^{152} γ-rays is 12.8 ± 0.4 g/cm^2 and that of the Eu^{154} 14.4 ± 1.2 g/cm^2. Thus it appears that the highest energy γ-ray observed by Shull is associated with Eu^{154}.

Unfortunately there is not sufficient activity to justify using these separated samples for beta-ray spectroscopic work. However, using the method of cadmium shielded irradiations, or very long slow neutron bombardment, it should be possible to produce Eu^{154} preferentially for which a complete analysis is more feasible.

Muehlhause[14] has studied the 9.2-hr. Eu^{152} and long lived Eu^{152} and Eu^{154} for K-capture by critical absorption methods. He found K-capture in both activities. His results showed that the branching ratio, i.e., K-capture of β^--emission ratio is 10.22 times greater in long lived Eu^{152} and Eu^{154} than that in 9.2-hr. Eu^{152}. We have rechecked this result and agree with his value within two percent. In the calculations to follow we will assume Muehlhause's value.

IV. NUCLIDE COMPOSITION OF THE IRRADIATED EUROPIUM SAMPLE

As will be shown in Section V, a knowledge of the nuclide composition of an irradiated sample before and after irradiation, together with a knowledge of the conditions of the irradiation, is, in general, sufficient to determine neutron absorption cross sections, nuclide half-lives, and nuclide branching ratios for many of the observed nuclides. The procedures required to make a mass spectrometric determination of nuclide composition, i.e., of both element and isotope abundance are the following. A standard solution is made which contains weighed amounts of all the elements in the original sample for which relative nuclide analysis is to be made. Each element in the standard solution must be of different isotopic composition than the corresponding element in the unknown solution. A portion of this standard solution is mixed with a portion of the unknown solution and the mass spectra of all three samples are observed. As long as only relative nuclide analyses are desired, these data alone are usually sufficient for the analysis. No knowledge of the mixing ratio of the standard and unknown solutions is necessary. Later in this section methods of calculation of the nuclide composition from these data will be discussed; first, however, the europium measurements will be described.

The mass spectrometer used for determining the nuclide composition was the same as that used previously by the authors in measuring the normal isotopic abundances of some of the rare earths.[15,16] Briefly, it consisted of a 60°, 6″ radius of curvature single focusing mass spectrometer using a surface ionization type of source. The resolved ion currents corresponding to the various mass positions were amplified with a vibrating reed electrometer and recorded on a Brown Electronic Strip Chart Recorder, so that permanent records of isotope abundances were obtained.

The analysis of the heavily irradiated europium sample gave currents corresponding to isotopes of mass numbers 151, 152, 153, 154, 155, 167, 168, 169, 170, 171, and 172. The ratio 151/153 was constant throughout the run. Since with a surface ionization type of source the ratio of ion currents due to adjacent rare earths is a marked function of time, it was concluded that these two peaks were due solely to europium, which emits predominately as Eu^+. The lines at masses 152 and 154 were found to increase with time relative to the 153. This effect was attributed to the presence of several elements, europium plus gadolinium or samarium or both, at these positions. The intensity of the 155 line compared to any other position was not constand and relative to the 153 it grew with time. This behavior was attributed to a contribution at 155 as a result of LaO^+. No correction could be made for this and so the 155 peak height was circumvented in the calculations. The peaks at 167 and 169 were always very small. They were caused by the slight europium emission of the form EuO^+. The lines at 170, 171, and 172 were proportional throughout the run. Since the europium contributions here would be negligible all these masses should be attributed to GdO^+. The 168 peak fraction-

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[14] C. Muehlhause, Manhattan Project Report CP-3750 (January, 1947).

[15] M. G. Inghram, R. J. Hayden, and D. C. Hess, Phys. Rev. **72**, 967 (1947).

[16] David C. Hess, Phys. Rev. **74**, 773 (1948).

ated with time relative to any other peak. Since its fractionation relative to the 170 was too large to be accounted for by a change in the small $Eu^{152}O^+$ current both Gd^{152} and Sm^{152} occur in the sample. That no normal Sm or Gd impurities occurred in the sample is amply shown by the complete lack of the peaks which should be observed from Sm^{147+} and $Gd^{158}O^+$ if normal samarium and gadolinium were present.

A standard solution of europium, gadolinium, and samarium was made by mixing together weighed amounts of normal Eu_2O_3, Gd_2O_3, and Sm_2O_3 and dissolving this mixture in nitric acid. An aliquot of this solution was mixed with an aliquot of a nitric acid solution of the irradiated europium. This mixture was analyzed by the mass spectrometer. The same lines as before and, in addition to these, the previously non-appearing members of the Sm^+, SmO^+, and GdO^+ spectra were observed. The Gd^+ spectrum was not observed except in very weak intensity just before the end of the run. By observation of the new 151/153 ratio the amount of these isotopes in the original sample relative to the amount added was calculated. The ratio of Gd^{154} present to gadolinium added was obtained from the 170 peak. This was found to fractionate very closely with the normal $Gd^{158}O^+$ peak. Utilizing also the $Sm^{147}O^+$ peak and the method of simultaneous equations later described, it was deduced that the greatest amount of Sm^{154} which could be present in the irradiated sample was less than 5 percent of the amount of Gd^{154} in the irradiated sample. In other words the branching ratio of Eu^{154} is less than 0.05. The relative amounts of Gd^{155} and Gd^{156} present were obtained by comparing the 171 and 172 peaks to the 170 peak in the original run. The situation at mass 152 was clarified by observing the change in the 152 line (europium and samarium) and the 168 line (gadolinium and samarium) with time, and so relating the amounts Eu^{152}, Gd^{152}, and Sm^{152} to the standards Eu^{153} (added), Gd^{158}, and Sm^{147}. This gave two separate measurements of the relative amount of Sm^{152} present. These agreed to within 5 percent.

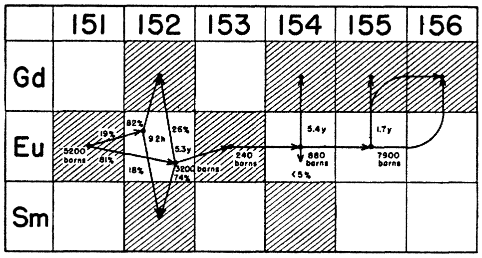

FIG. 4. Summary of reactions induced by slow neutron irradiation of europium, showing cross sections in units of 10^{-24} cm^2, half-lives, and branching ratios. See also footnote to Table I.

The following mathematical analysis summarizes the methods by which the relative nuclide composition of the unknown sample is obtained. Let i and j be two mass numbers where no lines other than those caused by element k occur in the mixture of standard and unknown samples. Let the superscript (1) refer to the unknown sample, (2) refer to the standard sample, and (3) refer to a mixture of the samples. Let a_{ik} and a_{jk} be the fractional abundances of the nuclides i, k and j, k in the element k. Further define $\alpha_{ijk} \equiv a_{ik}/a_{jk}$. Then it is easily shown that, where $N_{ik}^{(1)}$ is the number of atoms of the nuclide characterized by i, k which have been introduced into the mixture by the unknown sample, and where $N_k^{(2)}$ is the total number of atoms of element k introduced into the mixture from the standard sample, that we have

$$N_{ik}^{(1)} = a_{jk}^{(2)} \alpha_{ijk}^{(1)} \frac{(\alpha_{ijk}^{(3)} - \alpha_{ijk}^{(2)})}{(\alpha_{ijk}^{(1)} - \alpha_{ijk}^{(3)})} N_k^{(2)} \equiv b_{ijk} N_k^{(2)}. \quad (1)$$

The quantity b_{ijk} may here be calculated from the observed quantities α and $a_{jk}^{(2)}$. For element s suppose that different masses q and r play the roles of i and j above. Then a similar expression will be obtained for $N_{qs}^{(1)}$. The ratio of these expressions is given by

$$N_{ik}^{(1)}/N_{qs}^{(1)} = b_{ijk}/b_{qrs}(N_k^{(2)}/N_s^{(2)}). \quad (2)$$

Thus the relative abundances of the nuclides characterized by i, k and q, s in the unknown sample depends only on the mass spectrometrically determinable quantities b_{ijk} and b_{qrs} and on the relative numbers of atoms $N_k^{(2)}/N_s^{(2)}$ of elements k and s present in the standard sample. This ratio is determined by weighing the samples used in making the standard solution, and, from the known atomic weights of the compounds used, computing the relative amounts of k and s present. A knowledge of the mixing ratio of unknown and standard solutions is here unnecessary to determine $N_{ik}^{(1)}/N_{qs}^{(1)}$.

Consider now the analysis of a composite peak at mass m to which n elements give contributions. We assume that the unknown solution and the available standard solution are such that for every element k in the solutions there exist masses $i \neq m$ and $j \neq m$ which satisfy at least one of the following conditions: Case 1: At mass i only the element k occurs in the standard solution and no element occurs in the unknown solution. Case 2: At mass i only the element k occurs in the standard solution and at i and j only k occurs in the unknown solution. These assumptions are not the most general which might be made but they are sufficient for the europium analysis. An example of Case 1 in this analysis is the $Sm^{152}O^+ - Gd^{152}O^+$ conflict at $m = 168$. The "free position" i for samarium is mass 163 where $Sm^{147}O^+$ occurs in the standard spectrum and nothing is present in the irradiated spectrum. The $Gd^{158}O^+$ peak in the normal spectrum at $i=176$ establishes the free position for gadolinium. In the case of the $Sm^{152+}-Eu^{152+}$ conflict at $m=152$ the samarium satisfies the first condition with $i=147$ while the europium satisfies Case 2 with $i=151$ and $j=153$. For both cases we have for the peak heights P in arbitrary units at various mass positions

$$P_m^{(3)}=\sum_k A_k N_{mk}^{(3)}, \tag{3}$$

$$P_{ik}^{(3)}=A_k N_{ik}^{(3)}, \tag{4}$$

where the quantities A_k are proportionality factors depending only on the element k and the summation in (3) extends over all elements contributing to the peak at mass m. Letting

$$\alpha_{mik}^{(3)}=N_{mk}^{(3)}/N_{ik}^{(3)}$$
$$=(N_{mk}^{(1)}+N_{mk}^{(2)}/N_{ik}^{(1)}+N_{ik}^{(2)}), \tag{5}$$

we have substituting (4) and (5) in (3)

$$P_m^{(3)}=\sum_k \alpha_{mik}^{(3)}P_{ik}^{(3)}. \tag{6}$$

Because of the fractionation effects in the surface ionization source the relative peak heights for the various elements in the mixture change markedly with time. However, though the P's and their ratios are changing, Eq. (6) must be satisfied at any time. Thus if n measurements of the peak heights are made at different times throughout the run, n linear equations of type (6) would result. These may be solved for the n quantities α_{mik}. In case (1) where $N_{ik}=0$ reduction of Eq. (5) gives

$$N_{mk}^{(1)}=a_{ik}^{(2)}[\alpha_{mik}^{(3)}-\alpha_{mik}^{(2)}]N_k^{(2)}. \tag{7}$$

In case (2) a solution of the type described by Eq. (1) is possible at masses i and j. This result substituted into (5) gives

$$N_{mk}^{(1)}=[\alpha_{mik}^{(3)}(b_{ijk}+a_{ik}^{(2)})-a_{mk}^{(2)}]N_k^{(2)}, \tag{8}$$

where b_{ijk} is defined in Eq. (1). From Eqs. (1), (7), and (8) relative nuclide compositions are obtained, as in (2) by taking ratios.

Since the isotopic abundances of samarium, europium, and gadolinium are different in the irradiated and normal sample all the quantities a, α, and b involved are determinate. Thus straightforward application of the methods outlined above leads to a determination of the relative nuclide composition of the irradiated europium sample, complete except for the Eu^{155}.

V. DETERMINATION OF CROSS SECTIONS, HALF-LIVES, AND BRANCHING RATIOS

The values of the nuclide composition make it possible to calculate the thermal neutron absorption cross sections for all the europium isotopes observed, the half-lives of Eu^{152}, Eu^{154}, and Eu^{155}, the ratio of the number of atoms of Eu^{151} going into the long and short lived isomeric states of Eu^{152} upon thermal neutron absorption, and the branching ratio of the europium activities. The decay and absorption scheme which leads to the observed nuclides is shown in Fig. 4.

In order to calculate the unknown quantities it is necessary to set up a mathematical formulation of the changes (indicated in Fig. 4) which are taking place at time t in the pile. The differential equations so obtained must be integrated over the time of irradiation τ and corrected for the decay of certain of the nuclides in the time T since the end of the irradiation. Let $N_1(t)$ be the number of atoms of Eu^{151} present at time t, $N_2(t)$ be the number of Eu^{152} atoms, etc. Primes on the N's will denote gadolinium of similar mass and double primes samarium. σ_1, σ_2, σ_3, σ_4, σ_5, and λ_2, λ_4, λ_5 are the thermal neutron cross sections and the decay constants, respectively, for the corresponding europium isotopes, a is the fraction of Eu^{152} formed which is long lived, b is the fraction of 9.2-hr. Eu^{152} decaying to gadolinium, c is the fraction of long lived Eu^{152} decaying to gadolinium, and Φ is the thermal neutron flux. The equations describing conditions at time t during the irradiation are

$$dN_1/dt=-\sigma_1\Phi N_1, \tag{9}$$

$$dN_2/dt=-(\sigma_2\Phi+\lambda_2)N_2+a\sigma_1\Phi N_1, \tag{10}$$

$$dN_3/dt=-\sigma_3\Phi N_3+\sigma_2\Phi N_2, \tag{11}$$

$$dN_4/dt=-(\sigma_4\Phi+\lambda_4)N_4+\sigma_3\Phi N_3, \tag{12}$$

$$dN_5/dt=-(\sigma_5\Phi+\lambda_5)N_5+\sigma_4\Phi N_4, \tag{13}$$

$$dN_2'/dt=c\lambda_2 N_2+(1-a)b\sigma_1\Phi N_1, \tag{14}$$

$$dN_4'/dt=\lambda_4 N_4, \tag{15}$$

$$dN_6'/dt=(\sigma_5\Phi+\lambda_5)N_5, \tag{16}$$

$$dN_2''/dt=(1-a)(1-b)\sigma_1\Phi N_1+(1-c)\lambda_2 N_2. \tag{17}$$

In writing Eq. (16) the simplifications were made that (1) the half-life of Eu^{156} is zero and (2) the cross section of Gd^{155} is infinite. Since Eu^{156} has been observed in fission to have a half-life of 15.4 days,[12] and since measurements were made at a time several times that long after removal of the sample from the pile, and since the use of Eq. (16) is only to calculate $N_6'(\tau+T)$ the first of these simplifications introduces no errors into the calculation. Because σ_5' is so large (57,000 barns),[17] because the contribution to Gd^{156} via the Gd^{155} route turns out a good deal smaller than the contribution via the Eu^{156} route, and because the amount of Gd^{155} formed by decay after the end of irradiation

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[17] R. E. Lapp, J. R. Van Horn, and A. J. Dempster, Phys. Rev. **71**, 745 (1947).

is considerably larger than even the ***total*** Gd^{155} formed (most of which soon goes to Gd^{156}), the errors resulting from the second simplification are also negligible.

To solve the Eqs. (9)–(13) we will derive a general expression for $N_k(\tau+\mathrm{T})$, where τ is the time of irradiation during which the previous equations hold and T is the time after the end of the irradiation during which radioactive decay occurs. During the irradiation the general differential equations are

$$\begin{aligned} \frac{dN_1}{dt} &= -(\sigma_1\Phi+\lambda_1)N_1 \\ \frac{dN_j}{dt} &= -(\sigma_j\Phi+\lambda_j)N_j+\sigma_{j-1}\Phi N_{j-1} \quad j=2,\,3,\,\cdots k. \end{aligned} \tag{18}$$

Applying the Laplace transform

$$\bar{N}_j(s)=\int_0^\infty e^{-st}N_j(t)dt \tag{19}$$

to these equations we have, where N_{j0} is the initial value of N_j,

$$\begin{aligned} (s+\sigma_1\Phi+\lambda_1)\bar{N}_1 &= N_{10} \\ -\sigma_{j-1}\Phi\bar{N}_{j-1}+(s+\sigma_j\Phi+\lambda_j)\bar{N}_j &= N_{j0} \quad j=2,\,3,\,\cdots k. \end{aligned} \tag{20}$$

Solving the linear Eqs. (20) for $\bar{N}_k$ we obtain

$$\begin{aligned} \bar{N}_1 &= \frac{N_{10}}{s+\sigma_1\Phi+\lambda_1} \\ \bar{N}_k &= \frac{N_{k0}}{s+\sigma_k\Phi+\lambda_k}+\sum_{j=1}^{k-1}N_{j0}\Phi^{k-j}\left[\prod_{i=j}^{k-1}\sigma_i\right]\prod_{i=j}^{k}\frac{1}{s+\sigma_i\Phi+\lambda_i} \quad k=2,\,3,\,\cdots. \end{aligned} \tag{21}$$

Expanding the last product in the expression for $\bar{N}_k$ by partial fractions we have, assuming $(\sigma\Phi+\lambda)$ is not exactly the same for any two isotopes,

$$\begin{aligned} \bar{N}_1 &= \frac{N_{10}}{s+\sigma_1\Phi+\lambda_1} \\ \bar{N}_k &= \frac{N_{k0}}{s+\sigma_k\Phi+\lambda_k}+\sum_{j=1}^{k-1}N_{j0}\Phi^{k-j}\left[\prod_{i=j}^{k-1}\sigma_i\right]\sum_{m=j}^{k}\prod_{\substack{i=j\\ i\neq m}}^{k}\frac{1}{\sigma_i\Phi+\lambda_i-\sigma_m\Phi-\lambda_m}\left(\frac{1}{s+\sigma_m\Phi+\lambda_m}\right). \end{aligned} \tag{22}$$

Applying inverse Laplace transforms to these equations we have at time t

$$\begin{aligned} N_1(t) &= N_{10}\exp[-(\sigma_1\Phi+\lambda_1)t] \\ N_k(t) &= N_{k0}\exp[-(\sigma_k\Phi+\lambda_k)t]+\sum_{j=1}^{k-1}N_{j0}\Phi^{k-j}\left[\prod_{i=j}^{k-1}\sigma_i\right]\sum_{m=j}^{k}\prod_{\substack{i=j\\ i\neq m}}^{k}\frac{\exp[-(\sigma_m\Phi+\lambda_m)t]}{\sigma_i\Phi+\lambda_i-\sigma_m\Phi-\lambda_m}, \end{aligned} \tag{23}$$

and at a time T after irradiation time τ

$$\begin{aligned} N_1(\tau+T) &= N_{10}\exp[-(\sigma_1\Phi+\lambda_1)\tau]\exp(-\lambda_1T) \\ N_k(\tau+T) &= \exp(-\lambda_kT)\left\{N_{k0}\exp[-(\sigma_k\Phi+\lambda_k)]\tau+\sum_{j=1}^{k-1}N_{j0}\Phi^{k-j}\left[\prod_{i=j}^{k-1}\sigma_i\right]\sum_{m=j}^{k}\prod_{\substack{i=j\\ i\neq m}}^{k}\frac{\exp[-(\sigma_m\Phi+\lambda_m)\tau]}{\sigma_i\Phi+\lambda_i-\sigma_m\Phi-\lambda_m}\right\}. \end{aligned} \tag{24}$$

Utilizing the general solutions (24) for the Eqs. (9)–(13) we have five equations, of increasingly complicated algebraic nature for $N_1(\tau+T)$, $N_2(\tau+T)$, $N_3(\tau+T)$, $N_4(\tau+T)$, and $N_5(\tau+T)$. Wherever σ_1 appears in (24) representing contributions to Eu^{152} (long) or beyond a result of the original

Eu^{151} it must be replaced in the solution of the five special equations of $a\sigma_1$. Four more equations are obtained by direct integration of (14)–(17) with the now known N's substituted on the right-hand side. A tenth equation is obtained by integration of the differential equation

$$dN_5'/dt=\lambda_5 N_5, \tag{25}$$

which prevails after the irradiation. Three final equations are obtained from (1) the value (2600) of the cross section for normal europium which was computed from the best estimate of Φ_T as obtained from known absorbers irradiated with the europium; (2) the ratio of the beta-counting rates at masses 152 and 154 as described in Section III; and (3) the ratio of the branching ratios in 9.2-hr. Eu^{152} and long lived $Eu^{152}+Eu^{154}$ as given by Muehlhause. It should be pointed out that this definition is such that the x-ray counting efficiencies cancel. The three additional equations are

$$\frac{N_{10}\sigma_1+N_{30}\sigma_3}{N_{10}+N_{30}}=\sigma_T, \tag{26}$$

$$\frac{\lambda_2}{\lambda_4}=\frac{1}{c}\frac{(dN_2/dt)_\beta N_4(\tau+T)}{(dN_4/dt)_\beta N_2(\tau+T)}, \tag{27}$$

$$\frac{[(dN_2/dt)+(dN_4/dt)]_\beta}{(dN_2/dt)_K}\frac{(dN_2{}^*/dt)_K}{(dN_2{}^*/dt)_\beta}=\frac{c}{1-c}\frac{1-b}{b}\left[1+\frac{N_{30}\sigma_3\lambda_4}{N_{10}\sigma_1 ac\lambda_2}\right]. \tag{28}$$

In these equations subscripts K and β refer to the K-capture and beta-disintegration rates respectively; the superscript * denotes the 9.2-hr. isomer and terms without superscripts refer to the long lived activities.

The foregoing equations allow calculation of all the quantities of interest without involving counter efficiencies or use of $N_5(\tau+T)$. Considering the unknowns in the equations to be σ_1, σ_2, σ_3, σ_4, σ_5, λ_2, λ_4, λ_5, a, b, c, and $N_5(\tau+T)$ a numerical solution of the equations was carried out.

TABLE I. Pile neutron absorption cross sections, half-lives and branching ratios of the europium isotope.

Mass	151	152	153	154	155
$\sigma\times10^{24}$ cm^2 (see note (a))	5200	3200	240	880	7900
$T_{\frac{1}{2}}$ in years	—	5.3	—	5.4	1.7
K/β-ratios	—	0.22 (9.2 hr.) 2.8 (5.3 year)	—	<0.05	—

[a] These values of the pile neutron cross sections are all related linearly to the assumed value of 2600×10^{-24} cm^2 for the absorption cross section of normal europium. Thus an error in this value will enter linearly in all the cross sections. However it will leave the branching ratios and half-lives unchanged.

VI. CONCLUSIONS

The results of this calculation are summarized in Table I and Fig. 4. The values obtained for the constants a, b, and c, respectively, imply that of the Eu^{152} formed 81 percent has a half-life of 5.3 years and 19 percent has a half-life of 9.2 hr. Of the 9.2-hr. Eu^{152} formed 82 percent decays by beta-emission to gadolinium and 18 percent by K-capture to samarium. Of the 5.3-year Eu^{152} formed 26 percent decays by beta-emission and 74 percent by K-capture.

Because of the complexity of the computations necessary to arrive at these values no systematic analysis of error was carried out. However, the following possible sources of error were considered: (1) weighing errors in the preparation of the standard solution; (2) random and systematic errors in the nuclide composition chart as a result of mass spectrometer errors; and (3) different counting efficiencies for the activities deposited at 152 and at 154. We believe the cumulative effect of these errors is such that all the above values are good to 15 percent.

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