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The Europium Isotopes ^{142}Eu, ^{143}Eu and ^{144}Eu

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Summary

Two short-lived activities, with half-lives 2.61 ± 0.03 min and 1.2 ± 0.2 min, were produced by (*d*, *xn*) reactions on ^{144}Sm. On the basis of chemical separations and their genetic relationships these activities were assigned to ^{143}Eu and ^{142}Eu, respectively. Gamma rays of 1.11 and 1.55 MeV are associated with the decay of ^{143}Eu, and those of 0.77 and 1.03 MeV with the decay of ^{142}Eu. Reported data on ^{144}Eu could not be confirmed.

The half-life of ^{143}Sm has been re-determined as 8.84 ± 0.02 min. Analysis of the gamma ray spectra yielded a new gamma ray of 1.52 MeV.

Zusammenfassung

Bei der Bestrahlung von ^{144}Sm wurden durch (*d*, *xn*) Reaktionen zwei kurzlebige Radionuklide mit Halbwertszeiten von 2,61 ± 0,03 min und 1,2 ± 0,2 min gebildet. Aufgrund des chemischen Verhaltens und der genetischen Beziehungen wurden diese Aktivitäten dem ^{143}Eu bzw. ^{142}Eu zugeordnet. Bei ihrem Zerfall treten Gammalinien von 1,11 und 1,55 MeV bzw. 0,77 und 1,03 MeV auf. Die früher veröffentlichten Daten über den Zerfall von ^{144}Eu konnten nicht bestätigt werden.

Die Halbwertszeit von ^{143}Sm beträgt 8,84 ± 0,02 min. Eine Analyse der Gammaspektren ergab eine neue Gammalinie von 1,52 MeV.

Résumé

Les réactions (*d*, *xn*) du ^{144}Sm fournissent deux activités à vie courte, de périodes 2,61 ± 0,03 et 1,2 ± 0,2 min. Sur la base de séparations chimiques et des relations génétiques, elles sont attribuées respectivement à ^{143}Eu et ^{142}Eu. La désintégration du ^{143}Eu est accompagnée par des raies γ d'énergies 1,11 et 1,55 MeV, celle du ^{142}Eu par des raies γ de 0,77 et de 1,03 MeV. Nous n'avons pas pu confirmer les valeurs indiquées pour la désintégration de ^{144}Eu dans la littérature. La période du ^{143}Sm est 8,84 ± 0,02 min. Une analyse des spectres γ a montré une raie nouvelle de 1,52 MeV.

As indicated in Fig. 1 little is known about the neutron-deficient isotopes of europium, samarium and promethium which contain fewer than 82 neutrons. At the beginning of this investigation ^{142}Eu and ^{143}Eu were unknown and the data reported for ^{144}Eu and ^{143m}Sm were conflicting. After the completion of our investigation KOTAJIMA et al. [5] have published some data which they have tentatively assigned to ^{143}Eu. These and earlier data concerning the isotopes of interest in this work and their mode of production are summarised in Table 1 [2–16].

In the present experiment europium isotopes were prepared by (*d*, *xn*) reactions on ^{144}Sm. Calculated *Q*-values for these reactions, based on the mass-energy tables compiled by CAMERON [17] and by WAPSTRA et al. [18], are given in Table 2. Various samarium and promethium isotopes are also formed by (*d*, αxn) and (*d*, *pxn*) reactions, respectively.

From beta-decay systematics, the expected half-lives of the europium isotopes concerned were estimated to be less than a few minutes. It was obvious that rapid separation of the europium nuclides from irradiated targets would greatly facilitate their study.

Experimental

Highly purified Sm_2O_3, enriched to 94.54% in ^{144}Sm, was used as the target material for the production of the europium isotopes 142–144. Samples of 20 μg to 250 μg were enclosed in thin, high-purity aluminium foil (5.5 mg Al/cm^2) envelopes and irradiated for two to ten minutes in the internal beam of the Karlsruhe Cyclotron. Deuteron energies ranging from 15 to 40 MeV were used at beam currents of 2 to 10 μA.

Europium was separated chemically from the irradiated targets in about one minute by a procedure [19] which gives a separation factor of $> 10^3$ with respect to samarium, and $> 10^4$ with respect to other lanthanides. The separated fractions contained small, variable amounts of ^{24}Na produced by a (*d*, *p*α) reaction in the aluminium envelopes. The activity measurements were started within four to five minutes after irradiation. Beta activities were measured with methane flow counters. Gamma activities were measured by placing

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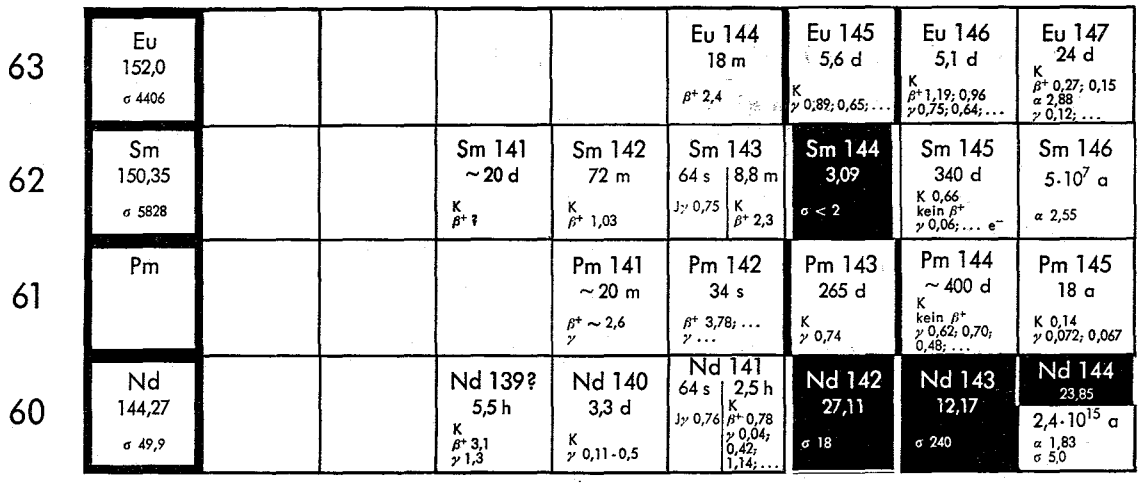

Fig. 1. Section of the chart of the nuclides [1]

Table 1. *Nuclear data on europium isotopes 142–144 and their decay products*

Nuclide	Half-life (min)	$E_{\beta\max}$ (MeV)	E_γ (MeV)	Reaction	Ref.
^{144}Eu	18	2.4			[2]
	Not detected			$^{144}Sm_2O_3$, α up to 44 MeV	[3]
	Not detected			$^{144}Sm_2O_3$, p 11 MeV	[3]
(?)	17			Eu_2O_3, p 660 MeV	[4]
	19	2.4, 3.15		Sm_2O_3, d 26 MeV	[5]
^{143}Eu	2.3 ± 0.2	4.0 ± 0.2	no strong gamma	Sm_2O_3, d 26 MeV	[5]
^{143g}Sm	8.3 ± 0.3	2.3	<0.15	Sm_2O_3, n (fast)	[6]
	9.03	2.6			[7]
	8.6 ± 0.1		no gamma	$^{142}Nd_2O_3$, α 38 MeV	[8]
	8.8 ± 0.2		1.05 weak	Sm_2O_3, γ 25 MeV	[9]
	8.5 ± 0.3			Sm_2O_3, n 14.8 MeV	[10]
	8.95			Sm_2O_3, γ 20 MeV	[11]
^{143m}Sm	2.28 ± 0.04		0.68	$^{144}Sm_2O_3$, p 20.6 MeV	[12]
	1.07 ± 0.05		0.748	Sm_2O_3, γ 25 MeV	[9]
	1.08 ± 0.05		0.75	$^{144}Sm_2O_3$, n 14 MeV	[13]
^{142}Sm	72	~ 1		$^{142}Nd_2O_3$, α 46 MeV	[14]
	72 ± 2	1.03		$^{142}Nd_2O_3$, α 48 MeV	[15]
	72 ± 2	1.03		Cs_2SO_4, ^{14}N ions	[15]
	74			Sm_2O_3, γ 30.5 MeV	[16]
	73 ± 2	no β^+	0.15–0.35 very weak	$^{142}Nd_2O_3$, α 44 MeV	[8]
^{142}Pm	0.5	3.78	1.6		[14]
	0.66 ± 0.1	3.78			[8]
	0.56 ± 0.05	3.80	1.59		[15]

the separated europium fractions, packed between 1 cm thick graphite absorbers, at a distance of 5 to 10 cm from $2'' \times 2''$ and $3'' \times 3''$ NaJ (Tl) scintillation detectors inside a $24'' \times 24'' \times 24''$ shield constructed of lead bricks 4″ thick and lined with thin sheets of cadmium and copper [20]. The detectors were used in connection with either a multiscaler arrangement, or multichannel pulse height analysers.

Results and Discussion

As anticipated from the reaction thresholds (Table 2) variation in the deuteron energy gives rise to different mixtures of reaction products. The gamma spectra of three europium fractions are shown in Fig. 2. These spectra were recorded immediately after the separation of europium from targets irradiated with 15, 25, and 40 MeV deuterons. These spectra as well as the analysis of the decay curves clearly demonstrate an increase in the number of nuclides produced as the deuteron energy increases. Changes in the gamma spectrum of a separated europium sample with time, as a result of the decay and growth of various components, are illustrated in Fig. 3. The results of our investigations are discussed below for individual nuclides.

20. R. L. HEATH, „Scintillation Spectrometry", Vol. I, p. 14, TID-4500 (1964).

Table 2. *Estimated reaction threshold values for the production of europium isotopes 142–144 by (d, xn) reactions on* ^{144}Sm

Nuclide	Reaction	Approx. threshold (MeV)
^{144}Eu	*d*, 2 *n*	9
^{143}Eu	*d*, 3 *n*	19
^{142}Eu	*d*, 4 *n*	31
^{141}Eu	*d*, 5 *n*	41

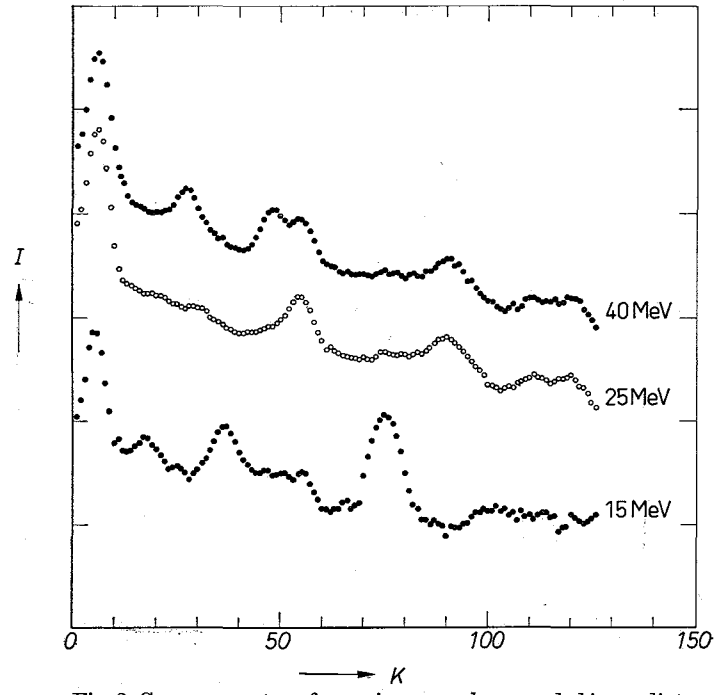

Fig. 2. Gamma spectra of europium samples recorded immediately after chemical separation from targets irradiated with 15, 25, and 40 MeV deuterons. The intensity I is plotted in arbitrary logarithmic units. The abscissa K represents channel number

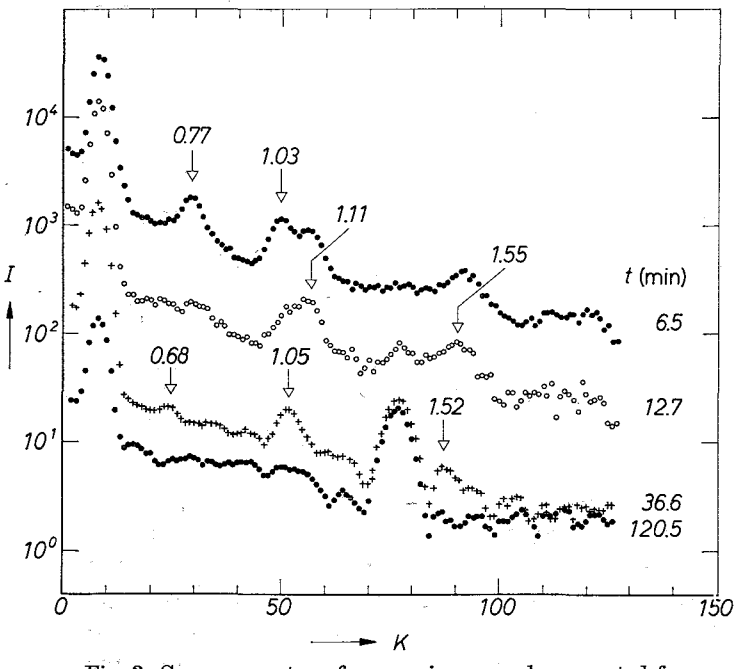

Fig. 3. Gamma spectra of a europium sample separated from a target irradiated with 40 MeV deuterons. The times shown represent the mean counting time after the end of bombardment. The numbers given in the figure show the energy of the different gamma peaks obtained in the analysis of the spectra. The peak at 0.68 MeV appears to be the sum-peak always observed in presence of annihilation radiation

^{144}Eu

No trace of a short-lived component which could be assigned to ^{144}Eu was found in the decay curves and gamma spectra of the europium fractions separated from targets irradiated with 15, 25, and 40 MeV deuterons. There was also no indication of the presence of a nuclide which decayed with a half-life of ~ 20 minutes. We must, therefore, conclude that the half-life of ^{144}Eu is either considerable shorter than one minute or longer than several hours. Beta decay systematics seems to favour the former.

The half-life of approximately 20 minutes reported for ^{144}Eu by several investigators (Table 1) should probably be assigned to ^{141}Pm [8], produced by (p, α) and $(d, \alpha n)$ reactions on ^{144}Sm. Such an erroneous assignment is understandable, for in some investigations [2, 5] the europium was not separated chemically from the other lanthanides.

^{143}Eu – ^{143}Sm

The beta decay curves (Fig. 4) as well as the gamma decay curves of europium fractions obtained from targets irradiated with 25 MeV deuterons showed the presence of four components, with half-lives 2.5 minutes, 8.8 minutes, 15 hours (^{24}Na), and ~ 6 days (^{145}Eu). Separation on an ion-exchange column [21] showed that the 8.8 min nuclide was a samarium isotope, which was identified as ^{143}Sm from its half-life and the prominent 1.05 MeV peak in its gamma spectrum.

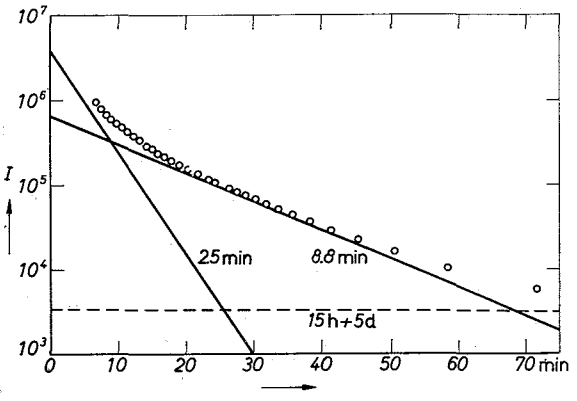

Fig. 4. Beta decay curve of a europium sample separated from a target irradiated with 25 MeV deuterons

By adding a ^{152}/^{154}Eu spike to the target material after irradiation and performing several consecutive europium separations, the 2.5 min component was shown to be a europium isotope. The ratio of the 2.5 min activity to the activity of the spike, referred to some particular time after bombardment, remained unchanged in the various fractions separated. The mass number 143 could be assigned to it on the basis of its genetic relationship with ^{143}Sm. Europium separations were performed either singly or repeatedly on the target material at various intervals of time after irradiation. In each case the ratio of 2.5 min activity to that of ^{143}Sm, referred to the time of separation, was about 2.8. Numerical analysis of the decay curves by the method

21. H. J. BLEYL (in preparation).

of least squares using an IBM 7070 computer [22] gave half-lives of 2.61 $\pm$ 0.03 min for ^{143}Eu and 8.84 $\pm$0.02 min for ^{143}Sm.

Gamma spectra of the europium fractions in the energy range below the annihilation radiation peak contained no characteristic peaks. Therefore only the energy range above 450 keV was investigated further. To follow the decay of the various peaks a series of successive spectra was recorded. By subtracting the spectra of longer-lived nuclides, corrected for decay, from the earlier spectra, the position of prominent peaks in the spectra of ^{143}Sm and of ^{143}Eu could be located. In this way the following gamma ray energies could be assigned:

^{143}Sm: 0.51 (annihilation radiation); 1.05; 1.52 MeV

^{143}Eu: 0.51 (annihilation radiation); 1.11; 1.55 MeV

For both nuclides the intensities of the gamma rays in the energy range above 0.51 MeV are one to two orders of ten smaller than that of the annihilation radiation. In the energy range above 1.6 MeV intensities were too low to permit reliable analysis by subtraction. There is, however, some evidence of low intensity peaks at energies ca. 1.8 MeV and ca. 1.9 MeV in the ^{143}Eu spectrum.

Measurements with aluminium absorbers indicated that the maximum positron energy of ^{143}Eu is greater than 3 MeV.

The results of JAMES and BINGHAM [12], who report a gamma energy of 0.68 MeV and a half-life of 2.28 minutes for ^{143m}Sm, and which completely disagree with the results of other investigators (Table 1), can now be explained in the light of the results of the present investigation. JAMES and BINGHAM irradiated $^{144}Sm_2O_3$ with 20.6 MeV protons and recorded gamma spectra of the irradiated target material without previously performing chemical separations. From the conditions of irradiation it is obvious that the irradiated targets contained ^{143}Eu, produced by a $(p, 2n)$ reaction on ^{144}Sm, and its ^{143}Sm daughter. The gamma spectrum of the 2.28 min activity obtained by these authors, and erroneously attributed to ^{143m}Sm, actually indicates the presence of weak 1.05 MeV (^{143}Sm) and 1.11 MeV (^{143}Eu) gamma rays.

^{142}Eu – ^{142}Sm

In addition to the nuclides mentioned above, the europium fractions separated from targets irradiated with 40 MeV deuterons contained two more components with half-lives of 72 minutes and $\sim$1 minute (Fig. 5). The short-lived component was difficult to detect in gross beta decay and gamma decay curves. It could be identified in beta decay curves measured through aluminium absorbers thick enough to suppress the contribution of ^{143}Sm and ^{24}Na, and in gamma decay curves measured with an energy window set at 0.65 to 1.2 MeV.

By methods similar to those described under ^{143}Eu – ^{143}Sm it could be shown that the 72 min component was ^{142}Sm. Its genetic relationship to the one-minute component was demonstrated by analysing beta decay curves of the europium fractions separated from aliquots of the irradiated target material at various time intervals after irradiation. The amount of ^{142}Sm present

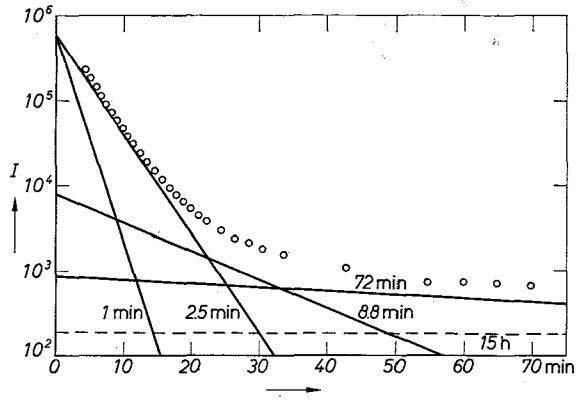

Fig. 5. Beta decay curve of a europium sample separated from a target irradiated with 40 MeV deuterons. The curve was measured with an Al-Absorber of 1080 mg/cm²

diminished rapidly with increasing length of time between termination of irradiation and time of europium separation. In the decay curves of europium fractions separated later than about ten minutes after irradiation, the 72 minute component could no longer be detected. Both for chemical and genetic reasons the one-minute component is therefore identified as ^{142}Eu. Calculation of the half-lives by means of the IBM computer gave 1.2 $\pm$ 0.2 min for ^{142}Eu and 72.49 $\pm$ 0.05 min for ^{142}Sm.

Measurements with aluminium absorbers indicated that the maximum positron energy of ^{142}Eu is greater than 3 MeV.

The gamma spectra of the europium fractions separated from targets irradiated with 40 MeV deuterons were likewise investigated in the energy range above 450 keV. The analysis was done by the subtraction method described above. In this way the following gamma energies could be assigned:

^{142}Sm: 0.51 (annihilation radiation); 1.59 MeV (^{142}Pm) very weak.

^{142}Eu: 0.51 (annihilation radiation); 0.77; 1.03 MeV. The intensities for the two gamma rays of 0.77 and 1.03 MeV are nearly equal, where the intensity of the annihilation radiation is rather small. This indicates that ^{142}Eu decays mainly by electron capture.

The assignment of the mass number 142 to the one-minute europium isotope is further supported by the fact the energies of these two gamma rays are very

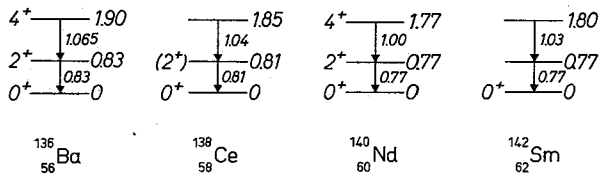

Fig. 6. The first two excited levels of the isotonic nuclei $N = 80$

22. H. MÜNZEL (in preparation).

similar to the energies corresponding to the first two excited levels in even-mass isotones of $N = 80$. The energy levels of the isotone nuclei ^{136}Ba [23], ^{138}Ce [23], and ^{140}Nd [24] are shown in Fig. 6.

Acknowledgement

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