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INVESTIGATION OF THE DIFFUSION PROCESS AT ELECTROMAGNETIC SEPARATION OF THE RADIOACTIVE ISOTOPES OF RARE-EARTH ELEMENTS

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Исследование процесса диффузии при электромагнитном разделении радиоактивных изотопов редкоземельных элементов

На основе результатов измерений эффективности выделения радиоактивных редкоземельных элементов из танталовой мишени были определены значения коэффициентов диффузии при разных температурах мишени. Дается краткое теоретическое рассмотрение процесса диффузии, позволяющее оценить ожидаемую активность короткоживущих изотопов на коллекторе электромагнитного масс-сепаратора при работе в режимах "off – line" и "on – line".

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Latuszynski A.

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Investigation of the Diffusion Process at Electromagnetic Separation of the Radioactive Isotopes of Rare-Earth Elements

Making use of the measurement results for the efficiency of migration of radioactive rare-earth elements out of the tantalum target, the values have been found for the diffusion coefficients at different temperatures of the target. A concise theoretical consideration for the diffusion process is given which makes it possible to estimate the activity expected for the short-lived isotopes on the collector of the electromagnetic mass-separator in "off-line" and "on-line" regimes.

**Preprint. Joint Institute for Nuclear Research.
Dubna, 1974**

Introduction

In nuclear-spectroscopic investigations of the short-lived radioactive nuclei far from the line of the beta-stability it is necessary to employ rather fast and effective production methods. In the case of extraction of the rare-earth element (REE) nuclei, through irradiating the corresponding targets by the high-energy protons, the produced radioactive REE are usually isolated from a target by the radiochemical methods [1] and are separated on the electromagnetic mass-separator. These operations require 20-30 minutes, this, in practice, limits the possibilities for studying isotopes with the half-life less than 10 minutes. As has been shown in papers [2-4], the radioactive atoms can be rather fast isolated from the heated refractory targets due to diffusion. This process may conveniently be carried out in the high-temperature ion source with surface ionization. The radioactive atoms emitted from the target can immediately ionize on the hot surface of the mass-separator ion source. By this method, in paper [4], the separated REE isobars have been obtained in the "off-line" regime. Duration of the whole process was 4-6 minutes what made it possible to investigate the isotopes with half-life of the order of 1 minute.

In the earlier published papers no information exists about values of the diffusion coefficient D of separate REE. The author of this paper aimed to determine these coefficients for tantalum, rhenium and tungsten targets. Knowing the values of D allows one to analyse the

possibilities of using diffusion in the electromagnetic separation of radioactive isotopes by the "off-line" and "on-line" methods.

The Diffusion Process

The diffusion coefficient D can be determined from the Fick equation relating the concentration of the diffusing matter C, distance x and time t, under the assumption that D does not depend on C:

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial x^2} . \qquad (1)$$

As is accepted usually, after irradiating comparatively thin targets with the high-energy protons, the diffusing matter in the target is distributed uniformly at the initial time that corresponds to the following initial conditions:

$$c(x,0) = c(0) \qquad 0 < x < a , \qquad (2)$$

$$c(x,t) = 0 \quad \text{at} \quad x \leq 0,\ x \geq a , \qquad (3)$$

where a is the target thickness.

The condition (3) means that the period time for the diffusing matter on the target surface is considerably less than the diffusion time /5/.

Under the above initial and boundary conditions, the solution to equation (1) for a thin plate target is as follows:

$$\bar{c}(t) = \frac{8}{\pi^2} c(0) \sum_{n=0}^{\infty} (2n+1)^{-2} \exp[-(2n+1)^2 K], \qquad (4)$$

where $K = (\pi^2 D t)/a^2$, $n = 0,1,2,3...$, $\bar{c}(t)$ is the average concentration of the diffusing matter in a target at time t, c (0) the same at $t = 0$. Since the concentration c is equal to the amount of diffusing matter per unit volume, the efficiency of diffusion after time t equals

$$\eta_D = \frac{c(0) - \bar{c}(t)}{c(0)} = 1 - \frac{8}{\pi^2} \sum_{n=0}^{\infty} (2n+1)^{-2} \exp[-(2n+1)^2 K]. \qquad (5)$$

In Figure 1 the dependence is shown of the value of $(1-\eta_D)$ on the parameter K calculated by eq. (5) to within the 10^{-3} accuracy. For the given values of η_D, t and a, the diffusion coefficient D can be determined from this graph.

The dependence of the diffusion coefficient D on the target temperature T is given via the following expression:

$$D = D_0 \exp(-Q/RT), \qquad (6)$$

where D_0 is a constant dependent on the target material and on properties of the diffusing matter, Q is the activation energy, R - the gas constant, T - the absolute temperature. The quantities D_0 and Q can be found from eq. (6) if the D(T) dependence is known.

Experimental Procedure and Measurement Results

The procedure of measurement of the diffusion efficiency was analogous to that described in /4/. The metallic foils with thickness of 0.1 mm and 0.05 mm and with sizes of about 2x2 mm^2 were used as targets. These targets were irradiated with the external beam of 660 MeV protons of the JINR synchrocyclotron and then were dipped into the ion source with surface ionization /6/. Experiments were carried out on the electromagnetic mass-separator of the YASNAPP facilities /7/.

The diffusion efficiency η_D was defined by the formula:

$$\eta_D = \frac{I_M - I'_M}{I_M} , \qquad (7)$$

where I_M is the intensity of characteristic gamma-transition in the nucleus of an isotope of REE(M) in the target after irradiation, I'_M the same after heating the target for the time period t.

The gamme-spectra (I_M and I'_M) were measured by means of the 50-cm^3 Ge(Li)-detector. The gamma-spectrum processing was carried out on the computer "Minsk-2". To determine η_D, sufficiently long-lived isotopes were used.

The values of η_D were measured at temperatures T = 2000, 2200, 2400, 2600 and at about 3100° K for time $t \leq 15$ min. The values of temperature of heating of the target were measured with the ± 50°K accuracy. Time was counted from the moment of establishing of the given temperature in the ion source. The time of heating of the ion source to this temperature was about 1 minute.

Using expression (5) and the graph on Fig. 1 for the measured values of η_D, taken partly from /4/, we defined the quantities $K=(\pi^2 Dt)/a^2$ from which the values for D were calculated for some REE. It is obvious that under our experimental conditions the values of D can somewhat be influenced by the processes occuring on the surface of the target (see condition (3)).

The first experiments have revealed that the diffusion coefficient of REE from tantalum is larger than that from rhenium and tungsten (Table 1). Therefore the investigation results will be presented only for the tantalum targets.

In Table 2 the values are given for η_D measured for time t = 3, 5, 10 and 15 min. at temperature T = 2400° K and a = 0.1 mm. The values of D were calculated for each value of time t and then the average value of the diffusion coefficient $\bar{D}$ was determined for T = 2400 °K. Using the values $\bar{D}$ thus defined for all the above-mentioned temperatures and taking exp. (6) in the graphical form: $\ln \bar{D} = \ln D_0 - (Q/RT)$, we have obtained the values of $\ln D_0$ and Q (Table 3). These values were computed by the least squares method on the computer "BESM-6". As an illustration, in Fig. 2 the graphs are plotted for europium, cerium and gadolinium.

Table 1. Comparison of the diffusion coefficients D for the tantalum, rhenium and tungsten targets

Element	D x 10^9 (cm^2/sec), T=2200°K		
	tantalum	rhenium	tungsten
Tm	6.2	1.9	1.7
Yb	6.8	4.9	2.7
Lu	0.98	0.32	0.28

Table 2. Measured values of the diffusion efficiency η_D and coefficients $\bar{D}$ for various REE in the tantalum target with thickness a=0.1 mm and for T=2400°K

Element	η_D				$\bar{D} \times 10^9$
	t=3min	t=5min	t=10min	t=15min	(cm^2/sec)
La	0.161	0.163	0.192	0.312	2.0±0.7
Ce	0.158	0.175	0.205	0.325	2.1±0.5
Pr	0.174	0.209	0.312	0.407	3.2±0.3
Nd	0.178	0.215	0.309	0.405	3.2±0.3
Eu	0.400	0.550	0.600	0.652	14.7±4.8
Gd	0.240	0.340	0.399	0.481	6.0±1.1
Tb	0.180	0.220	0.330	0.424	3.5±0.3
Dy	0.212	0.260	0.395	0.502	5.0±0.5
Ho	0.183	0.294	0.332	0.490	4.5±1.1
Er	0.198	0.284	0.312	0.495	4.5±1.0
Tm	0.278	0.355	0.484	0.510	7.5±1.0
Yb	0.310	0.372	0.491	0.532	8.3±1.7
Lu	0.150	0.170	0.208	0.337	2.1±0.5

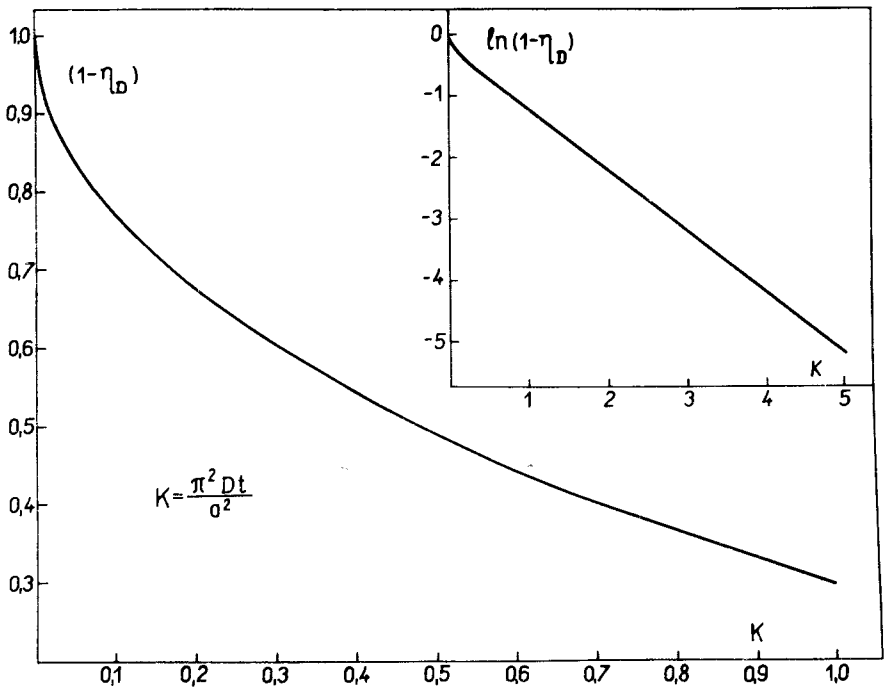

Fig. 1. Dependence of the diffusion efficiency η_D on the parameter K.

Employing the values found for D_0 and Q from eq. (6) one can calculate the values of D, for any temperature, within the accuracy defined by our experimental errors (not worse than 25%). The values of D thus calculated for T = 2000, 2500 and 3000°K are presented in Table 3. The data are absent for Sm and Pm because these elements have no isotopes suitable for our conditions of measurements of values of η_D.

To make easier the practical applications of the obtained data, in Fig. 3 the dependences $\eta_D = f(t/a^2)$, see formula (5), are given for europium, lutecium and holmium at T = 2000, 2500 and 3000°K. The curves for other REE are lying between the curves for europium and

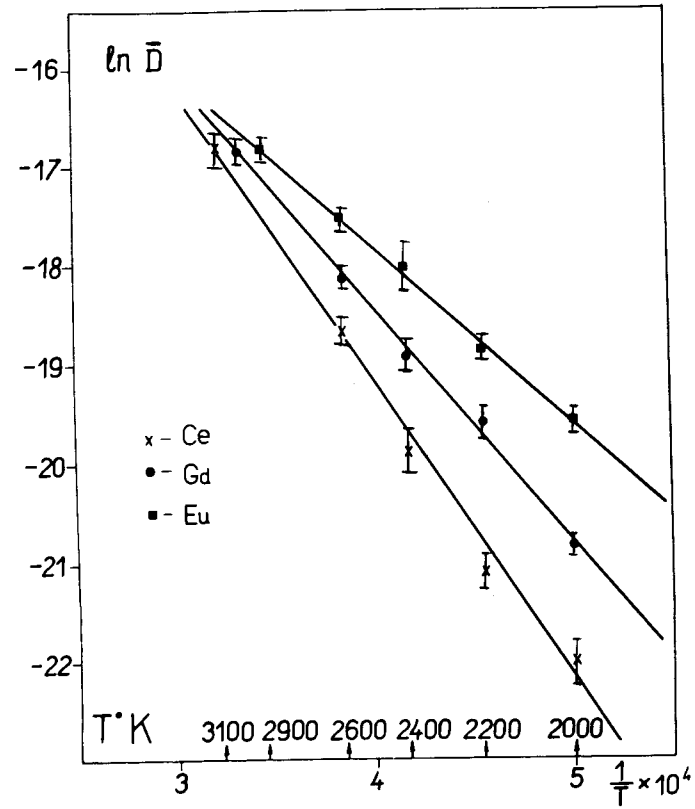

Fig. 2. The graph for determining the constants D_0 and Q in the diffusion of REE from tantalum.

lutecium. From these graphs one can easily estimate the magnitude of the efficiency of diffusion of REE from the tantalum target under different experimental conditions, as well as for products of heavy-ion nuclear reactions /8/.

The Expected Activity on the Collector of Mass-Separator

Making use of the diffusion process for obtaining the short-lived isotopes of REE to estimate the activity expected on the collector of mass-separator, one should

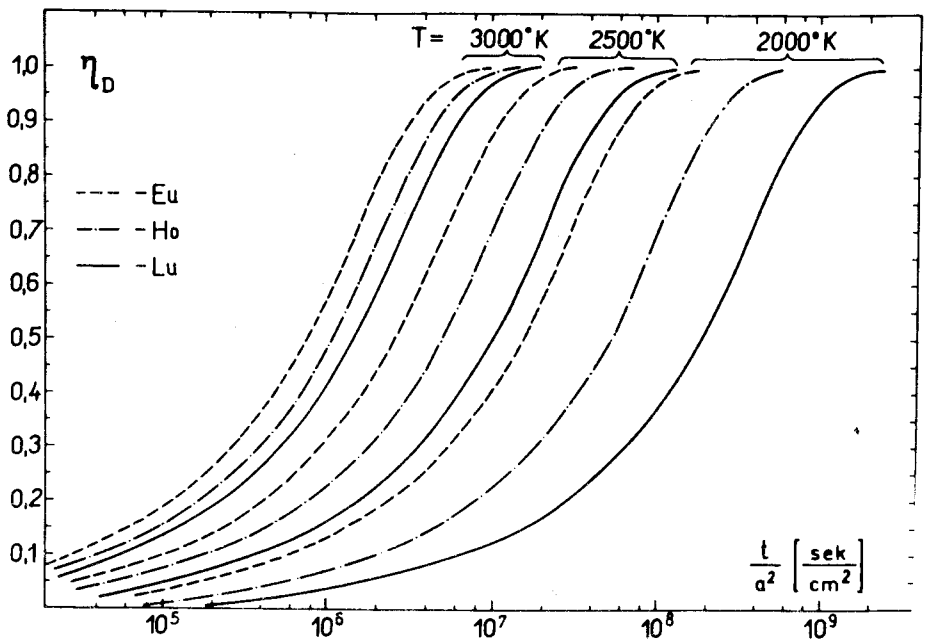

Fig. 3. Dependence of η_D on the diffusion time and target thickness a for europium, holmium and lutecium at temperatures T = 2000°K, 2500°K, 3000°K.

take into account the conditions of irradiation of a target and the regime of separation process. When the diffusion of short-lived radioactive atoms proceeds simultaneously with the target irradiation ("on-line" regime) it is necessary to allow for the rate of production of the radioactive atoms and their decay. Then the Fick equation takes the form

$$\frac{\partial c}{\partial t} = D\,\frac{\partial^2 c}{\partial x^2} - \frac{c}{\tau} + \delta\rho I, \qquad (8)$$

where τ is the mean life of the radioactive atoms, σ - the production cross section for these atoms, I - the density of the proton beam, ρ - the target density (number of atoms per 1 cm^3 of the target).

Under the condition of established equilibrium in the target when $\partial c/\partial t = 0$ and with (3), the solution of equation (8) has the form (see, e.g., ref. [9]):

$$c = \sigma\rho I\tau\left[1 - \frac{\cosh\left[\left(x - \frac{a}{2}\right)(D\tau)^{-1/2}\right]}{\cosh\left[\frac{a}{2}(D\tau)^{-1/2}\right]}\right]. \qquad (9)$$

The number of atoms of a given isotope diffusing per unit time from the target with surface S is equal to:

$$\frac{dN}{dt} = -DS\left(\frac{\partial c}{\partial x}\right)_{x=0} - DS\left(\frac{\partial c}{\partial x}\right)_{x=a}. \qquad (10)$$

Making use of (9) we obtain

$$\frac{dN}{dt} = \frac{2\sigma I}{a} N_0 (D\tau)^{1/2} \tanh\left[\frac{a}{2}(D\tau)^{-1/2}\right]. \qquad (11)$$

(N_0 is the total number of atoms of the target).

The activity obtained on the collector of mass-separator will depend not only on the diffusion process but also on the efficiency of ionization of atoms in the source (β) and on the efficiency of the transportation of the ion beam (η_t), i.e., on the efficiency of mass-separation $\eta_i = \beta\eta_t$. The ionization time for atoms in the ion source and the time of transportation of ions through the mass-separator can be neglected. Then, according to eq. (11), the activity of a given isotope, I_c, in the equilibrium state, obtained on the collector when working in the "on-line" regime is as follows:

$$I_c = \left(\frac{dN}{dt}\right)_c = \frac{2N_0\sigma I}{a}\eta_i (D\tau)^{1/2} \tanh\left[\frac{a}{2}(D\tau)^{-1/2}\right]. \qquad (12)$$

Working in the "off-line" regime, when the equilibrium state is not established in the target ($\partial c/\partial t \neq 0$) because of diffusion and radioactive decay, one should take into account the time of transportation of the target and that of preparation for work of the ion source (t_n) and also the duration of the mass-separation process which, in practice, equals the diffusion time (t). The activity of a given isotope obtained on the collector of mass-separator at the moment of time ($t_n + t$) can in this case be written in the following way:

$$I_c = \left(\frac{dN}{dt}\right)_c = N_0 \sigma I \eta_i \eta_D \exp\left(-\frac{t+t_n}{\tau}\right)\left[1 - \exp\left(-t_0/\tau\right)\right], \quad (13)$$

where t_0 is the time of target irradiation.

These two latter equations make it possible to estimate the activity expected for two regimes of work of the electromagnetic mass-separator, if one knows the values of σ, τ, D and η_i corresponding to a given isotope.

Using the obtained coefficients of diffusion of REE from the tantalum target we have calculated, by formulae (5), (12) and (13), the values of the activity of radioactive isotopes on the collector of mass-separator for two regimes of work of the YASNAPP facilities (Figs. 4 and 5). In Table 4 we give the characteristics of work in the "off-line" regime, accepted in calculations, and those for the "on-line" regime planned in the future. The weight of target chosen (1g) is connected with the present construction of the ion souce [6]. The value accepted for η_i is the average value for REE, the concrete data for various REE are given in refs. [4,6].

For the "off-line" regime, the corresponding values of the activity $I_C = f(D)$ of isotopes with $T_{1/2} \sim 5$ min. for the target thickness a from the interval 0.01 to 1 mm are shown in Fig. 4. The vertical lines restrict the regions corresponding to REE with the minimal (Lu) and maximal (Eu) values of the diffusion coefficient at different temperatures (this case is described in more detail in ref. [4]).

Table 3. The values obtained for constants $\ln D_o$ and Q and calculated coefficients of diffusion of REE from tantalum

Element	$-\ln D_o$ (cm^2/sec)	$Q \pm \Delta Q$ (kcal/g-atom)	$D \times 10^9$ (cm^2/sec) 2000°K	2500°K	3000°K
La	6.78	62.6±4.5	0.18	4.2	33.5
Ce	7.15	60.1±3.2	0.23	4.7	34.9
Pr	8.53	50.4±2.1	0.67	8.3	44.4
Nd	8.38	51.0±0.9	0.67	8.6	47.1
Eu	10.60	36.0±1.0	3.2	18.6	63.6
Gd	9.02	47.2±1.8	0.92	9.8	47.1
Tb	8.35	51.0±1.3	0.67	8.6	47.6
Dy	9.33	45.6±2.7	0.98	9.6	43.9
Ho	9.25	46.0±1.8	0.96	9.6	44.8
Er	9.17	46.6±2.6	0.92	9.4	44.4
Tm	10.33	37.8±4.0	2.6	17.3	61.1
Yb	10.65	36.0±3.8	3.0	18.0	59.3
Lu	7.56	58.0±4.2	0.26	4.8	32.9

Table 4. The main parameters of work of the YASNAPP facilities

Parameters	YASNAPP-1 (off-line)	YASNAPP-2 (on-line)
Density of the proton beam I (cm^{-2} sec^{-1})	10^{11}	10^{13}
Number of atoms of the irradiated target N_o	3×10^{21}	3×10^{21}
Time of separation t (min)	~ 5	continuous
Time of preparation of the ion source t_n (min)	~ 2	0
Half-life of the investigated isotopes $T_{1/2}$	~ 5min	1sec
Time of irradiation t_o	~ $3T_{1/2}$	continuous
Production cross section of radioactive atoms (cm^{-2})	2×10^{-27}	2×10^{-29}
Efficiency of mass-separation	0.5	0.5

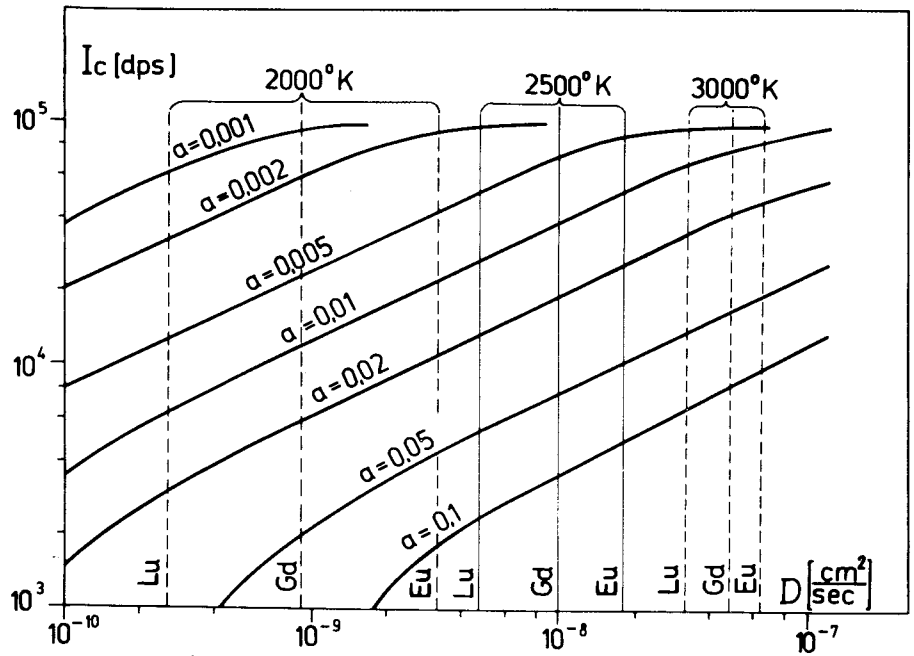

Fig. 4. Dependence of the activity of isotopes of REE with $T_{1/2}$ ~ 5 min. on the collector of mass-separator in the "off-line" regime upon the diffusion coefficient D for different target thickness. (The target mass lg, time of preparation of sources ~ 7 min).

The values of the activity $I_c = f(\sqrt{D\tau})$ for the "on-line" regime, at the same thickness of the target are given in Fig. 5. Here the vertical lines enclose the working region, which is interesting for this regime, restricted by the isotopes with $T_{1/2}$ ~ 0.1 sec. and by the minimal for REE value of D (for lutecium) from the one side, and by those with $T_{1/2}$ ~ 30 sec. and maximal value of D (for europium) from the other side. In these cases the values D are taken for T = 2500 °K (Fig. 6).

The data shown on Figs. 4-6 allow one to choose the optimal conditions of work (temperature and thickness of

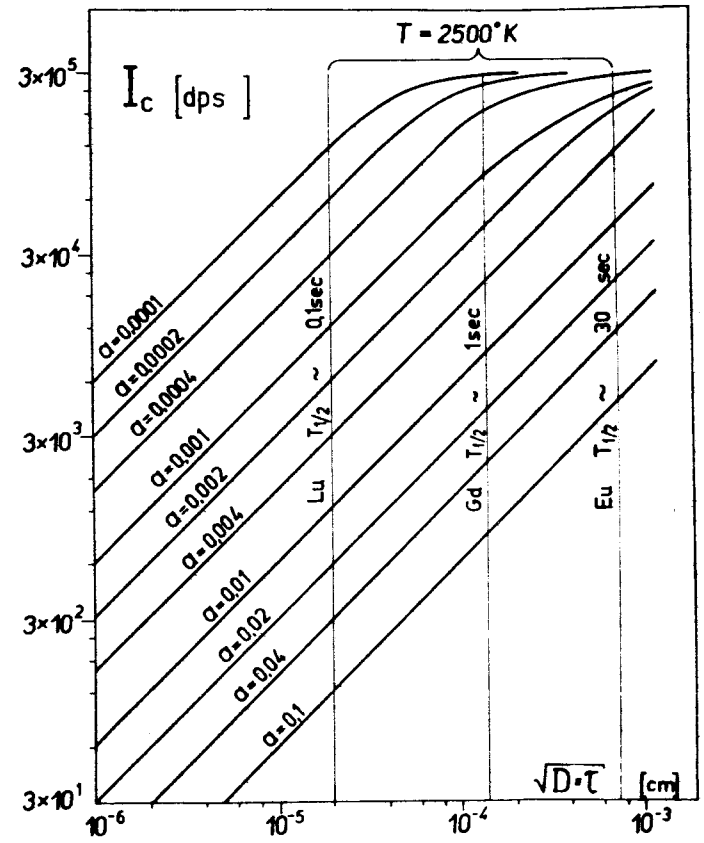

Fig. 5. Dependence of the activity on the collector of mass-separator in the "on-line" regime upon the diffusion coefficient D and mean lifetime of radioactive atoms τ for various target thickness. (The target mass lg).

the target) for each REE. Therefore, one of the possible methods of identification of the obtained elements in an isobar can be based on the difference between their values of D (especially, at low temperatures, Fig. 6) and on the distinction between the ionization efficiency β in the ion source.

As is shown on Figs. 4 and 5, making use of the diffusion process for releasing the radioactive atoms

of REE from tantalum, working both in the "off-line" and in the "on-line" regimes, one can obtain on the collector of mass-separator the isobars of REE with sufficiently large for spectroscopical investigations values of the activity. The real possibility of such an estimation, in the case of the "off-line" regime, is well justified by the contributions on the spectroscopic investigations of REE isotopes (the YASNAPP-1 program) submitted to the Symposium in Kharkov [10]. For the "on-line" regime, the data obtained allow one to assert that for the effective application of the diffusion process it is optimal to use the thin ($a \approx 0.01$ mm) tantalum targets (for instance, in the form of converted foils) and irradiate them directly in the high-temperature ion source with surface ionization (the working temperature $T = 2500 - 3000^\circ$ K). In doing so, it is sufficient to employ

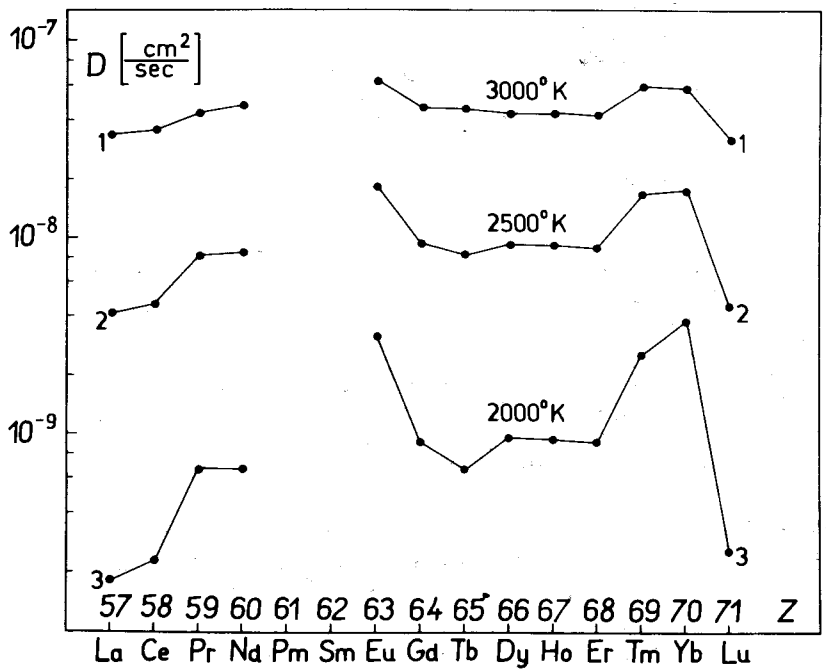

Fig. 6. The diffusion coefficients D for REE diffusing from the tantalum target at various temperatures.

the targets with weight of several gs. Under such conditions it is possible to obtain, with efficiency 10 - 30%, on the collector of mass-separator the REE isotopes with $T_{1/2} \sim 1$ sec produced in the irradiated target.

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