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E6 - 7780

1979/2-74

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OF RADIOACTIVE ISOTOPES
OF RARE-EARTH ELEMENTS DIRECTLY
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1974

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Introduction

The method of obtaining the sources of rare-earth elements (REE) for spectroscopic investigations with application of the deep spallation process used so far included the following operations:

1) irradiation of the target by the proton beam;

2) chemical extraction of elements;

3) preparing of samples for the following separation by isotopes;

4) electromagnetic separation of samples according to isotopes.

The total time of operations 2)-4) was equal to around 30 minutes. This limited the possibility of the spectroscopic investigations of isotopes with the half-life $T_{1/2}$ · · 10 minutes.

In work [1] the possibility of the complete diffusion of REE from the irradiated tantalum during the time of the order of 1 minute was proved. The application of the ion source with the surface ionization (ISSI) in the electromagnetic mass-separator provides the possibility of using the diffusion process what considerably shortens the time of obtaining the radioactive sources due to exclusion of chemical operations. The irradiated target is dipped straight into the evaporator of the ion source. The REE atoms diffuse from the target material into the volume of ionozator and then after ionization and electromagnetic separation on the collector of the separator, the separate isobars and gatheres. The possibility of obtaining the REE isotopes from the tantalum target in this way was propos-

ed in work[2] where the activities of the separated elements have been calculated allowing for the fact that the coefficients of diffusion from tantalum are the same as those from graphite[3].

The present paper is devoted to the experimental determination of conditions of diffusion and of electromagnetic separation of REE from tantalum, rhenium and tungsten targets and to the application of the method to spectroscopic investigations.

In the elaborated method of the fast extraction of REE isobars the process of preparing the spectroscopic samples is divided into the following stages:

1. irradiation of a metallic target with the proton beam;

2. transportation of the irradiated target into the ion source, in our experiments the time of transportation was equal to one to two minutes;

3. the process of separation includes:

   i) diffusion of elements from the target material;

   ii) ionization on the surface of ionizator of the ion source;

   iii) extraction of ions from ionizator with the lens system of the mass-separator and transportation of the separated elements on the collector.

The time of electromagnetic separation is determined mainly by the speed of diffusion. Efficiency of the separation process η is determined by the efficiency of the diffusion η_D of separate elements and their ionization coefficients β, by the efficiency of the mass-separator η_S and also by the coefficient K allowing for the distortion of equilibrium state in the given construction of the ion source:

$$\eta = \eta_D \cdot \beta \cdot \eta_S \cdot K , \qquad (1)$$

where the measured ionization efficiency is $\eta_i = \beta \cdot \eta_S \cdot K$. The efficiency η_S takes into account ion losses in the extraction processes, transportation and precipitation of the separated elements on the collector of the mass-separator ($\eta_S \approx 0.9$).

4

Efficiency of the diffusion process η_D can be determined from the equation of the one-dimensional diffusion (Fick law). However, one has to know for this purpose the values of the diffusion coefficient D .

No information does exist about values of the coefficient D for REE in tantalum, rhenium and tungsten. Therefore we measured the efficiency of diffusion in special working conditions of ISSI depending on temperature, time and thickness of the target.

The coefficient of the surface ionization β of the given element is determined by the Saha-Langmuira formula obtained for conditions close to those existing in ISSI:

$$
\begin{aligned}
\beta &= \frac{n^+}{n^0 + n^+} = \frac{\alpha}{1 + \alpha} , \\
\alpha &= \frac{n^+}{n^0} = A \; e^{\left(\frac{\phi - V_i}{kT}\right)} , \\
A &= \frac{2 J^+ + 1}{2 J^0 + 1} ,
\end{aligned}
\tag{2}
$$

where n^+ is the number of ions, n^0 is the number of neutrals, A is a constant for the given element defined in the first approximation by quantum numbers of total moment of the ion (J^+)   and atomic (J^0) states[1] , ϕ is the work function of the ionizator meterial, V_i is the first potential of ionization for the given element, T the temperature of ionizator.

According to eq. (2) in the temperature range of 3000 to $3500°K$ the sufficiently high ionization rate occurs for elements for which $V_i < 7$ eV. This condition is fulfilled for all REE. The values of β for REE ionizator ($\phi = 5.43$ eV) at the temperature $T = 3100°K$ calculated according to eq. (2) are represented in Fig. 6. The values of A are calculated from the data of ref.[5].

Since $\eta_i \neq \beta$ the dependence of η_i on the ionizator temperature and separation time was also a subject of our investigations.

Measurement Procedure

Targets with the thickness of 0.05mm and 0.1mm were irradiated by the proton beam with energy 660 MeV on the JINR synchrocyclotron. The irradiation time and the time of measurement were selected for each isotope dependening on its half-life.

The separation was carried out on the electromagnetic separator working in the program YaSNAPP-1 [6] using ISSI [7]. The temperatures of evaporator and ionizator of the ISSI were determined according to the curve of dependence of temperature on the power of the electron current bombarding evaporator and ionizator (Fig. 1). For construction of this curve a special experiment was performed in which the melting points of some pure metals were used. The target was placed in the ion source as shown schematically in Fig. 1. The conditions under which the values of η_D and η_i were measured were varied in the following ranges:

temperature T	$2000 \leq$ T $\leq 3300^\circ$ K
time of separation t	t $\leq$ 15 minutes
thickness of target a	0.05 and 0.1 mm
atomic number of investigated elements Z	$57 \leq$ Z ≤ 71 (except for Z = 61 and Z = 62)
target materials	tantalum, rhenium, tungsten.

The technical parameters of separator allowed to obtain simultaneously on the collector isobars of all neutron-deficient REE, i.e., isotopes with masses $126 \leq A \leq 173$.

The diffusion efficiency η_D and ionization efficiency η_i were determined according to the formulas:

Fig. 1. Temperatures (I and II) of the ionizator (b) and evaporator (a) as a function of the electron impact power (P) with the ionizator (P_{K1}) and evaporator (P_{K2}). Notations: c - rhenium foil, d - extraction slit, K_1 and K_2 - electron heaters, t - target.

$P_{K_2} = 0.25 P_{K_1}$

7
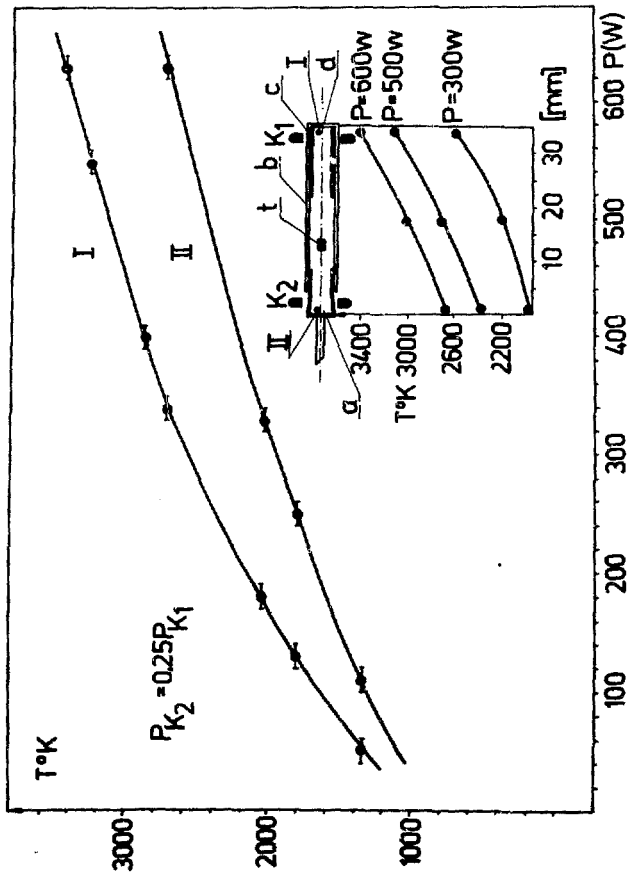
$$\eta_{D} = 100 \left(\frac{J_M - J'_M}{J_M} \right) \% \quad ; \tag{3}$$

$$\eta_{i} = 100 \left(\frac{J_K}{J_M - J'_M} \right) \% \quad , \tag{4}$$

here: I_M is the intensity of the gamma-line of the given isotope in the target before separation, I'_M is the same after separation, I_K is the intensity of the gamma-line of the given isotope on the separator collector. The quantities I_M, I'_M, I_K had been taken from measurements of the corresponding gamma-spectra. Measurements were performed on the $Ge(Li)$ detector of the volume 50 cm^3 with the resolution 2.8 keV for energy 1300 keV. Processing of gamma-spectra was carried out on the computer "Minsk-2" using the program KATOK [8].

Discussion of the Results
a) D i f f u s i o n

Dependence of the diffusion efficiency η_D on the target temperature for different diffusion times is shown in Fig. 2 for Eu and Tb as the examples. The values of η_D for REE diffusing from tantalum with the thickness 0.1 mm is presented in Fig. 3. The absence of values of η_D for Sm and Pm can be explained by the lack of appropriate for measurements isotopes of these elements. The values of η_D for REE at different periods of diffusion time are presented in Fig. 4. As the example the dependences of η_D on time for Eu and Tb are given in Fig. 5. The sharp dependence of the values of η_D on temperature and diffusion time is observed what is in accordance with the Fick law. In agreement with the work [1] nearly 100% of the diffusion efficiency for all REE is obtained at the temperature $\approx 3000°K$ during the time t $\approx$ 3 minutes. Some correlation between the values of (100 - η_D) of the given element and its boiling point is seen in Fig. 3.

In Tables 1 and 2 the values of diffusion efficiency for some REE are presented depending on material and
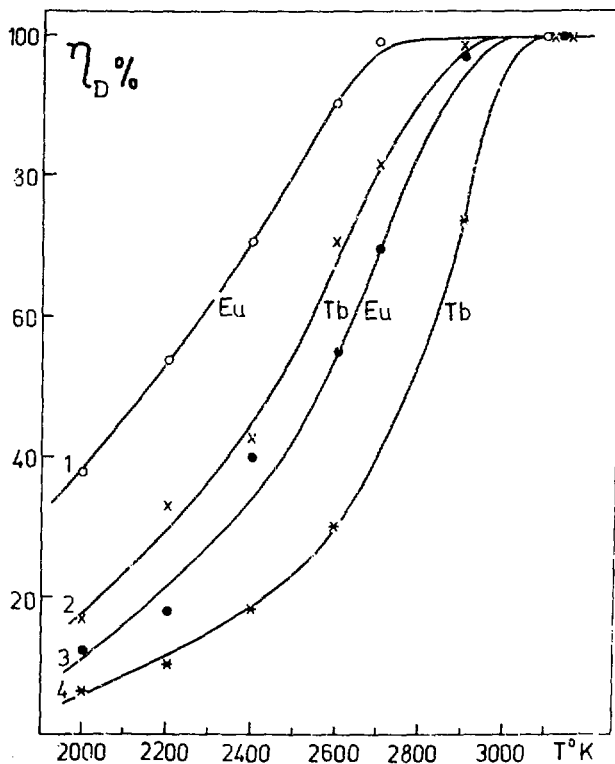
Fig. 2. Dependence of the efficiency of diffusion η_D (%) on temperature T for Eu (curves 1 and 3) and Tb (curves 2 and 4) at the time period 3 minutes (curves 3 and 4) and 15 minutes (curves 1 and 2).

thickness of targets. It is seen that efficiency is larger for targets with smaller melting point T_m. However, targets with low T_m cannot be employed directly in the ISSI construction under consideration. One can shorten

Table 1
Values of η_D % for some REE in different metallic targets.
$T = 2200°K$, $t = 15$ min, $a = 0.1$ mm, T_m - melting point.

REE	TARGET		
	$Ta(T_m=3273°K)$	$Re(T_m=3453°K)$	$W(T_m=3653°K)$
Gd	37.8	14.8	7.5
Tm	46.2	29.2	27.8
Yb	48.4	47.4	35.0
Lu	13.9	12.0	11.6

Table 2
Values of η_D % for some REE in tantalum target of different thickness at temperature $T = 2900°K$, the diffusion time - 2 and 4 mins.

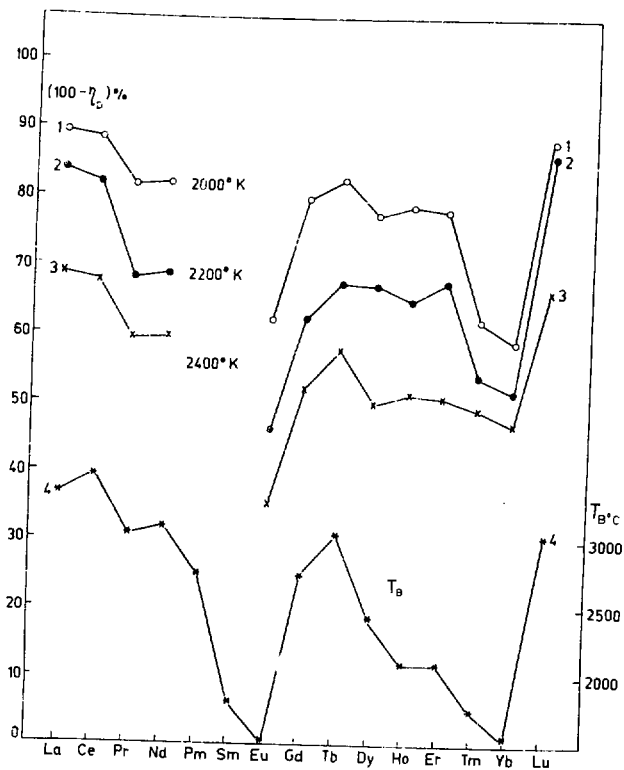| REE	$a = 0.05$ mm		$a = 0.1$ mm	
	2 min	4 min	2 min	4 min
Eu	~100	~100	63	95
Gd	90	~100	50	92
Tb	93	~100	29	90
Yb	93	~100	40	93

=

Fig. 3. The values of $(100 - \eta_D)$, %, for REE at temperatures: curve 1 - T = 2000°K, curve 2 - T = 2200° K, curve 3 - T = 2400°K. Time t = 15 minutes. Curve 4 gives the value of the boiling point of REE.

the time or enlarge the diffusion efficiency decreasing the target thickness. Using the method for off-line spectroscopic investigations the thickness 0.05 mm of the tantalum target was the optimal one.
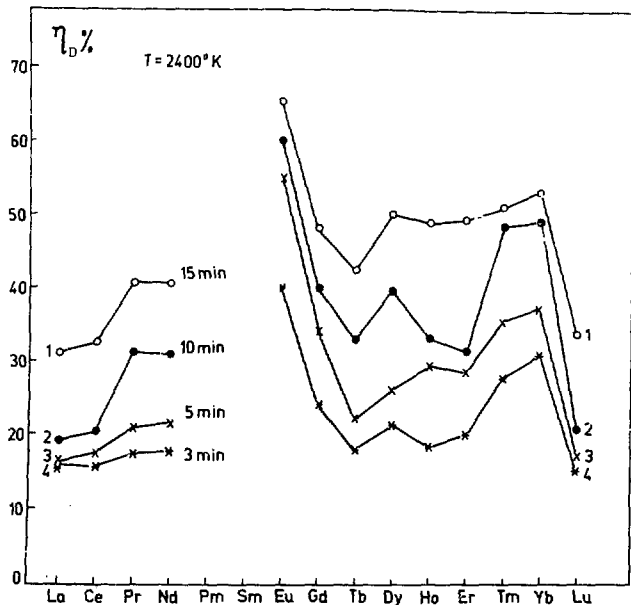

Fig. 4. Values of η_D of REE for different diffusion times: Curve 1 - 15 min, curve 2 - 10 min, curve 3 - 5 min, curve 4 - 3 min. Temperature $T = 2400°K$.

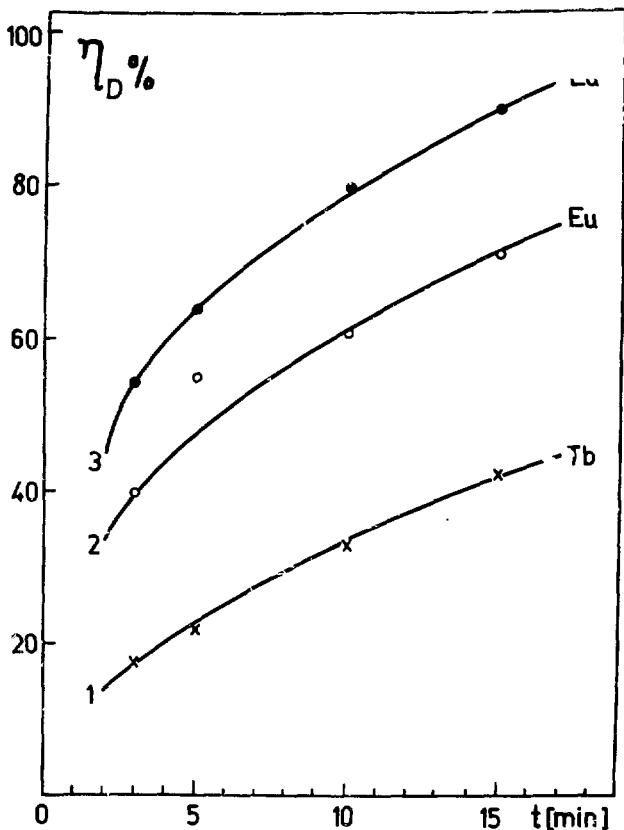Fig. 5. Dependence of the diffusion efficiency η_D (%) for Eu (curves 2 and 3) and Tb (curves 1) on time, for temperatires $T = 2400°K$ (curves 1 and 2) and $T = 2600°K$ (curves 3).

b) Ionization

The obtained values of the ionization efficiency η_i exhibit dependence on temperature of the surface of ionizator of the ion source and sharp dependence on ionization potential of the given element (Fig. 6). The values of η_i turned out to be, as well as in works /7,9 /, significantly larger than the values of β calculated by the Saha-Langmuira formula (2), but the nature of dependence of η_i on Z of elements remained the same (Fig. 6). Due to the fact that in the formula (4) the losses connected with the ion migration through the mass-separator and with their scattering on the collector material were not taken into account, the true values of η_i are even higher.

One can mention the following possible sources of some disagreement between the values of η_i obtained experimentally in the present work, in works /7,9 / and those calculated by the eq. (2) values of β :

i) Geometrical parameters of the ion source and electrical conditions of extraction, due to which conductivity of the exit hole is larger for positive ions than for neutrals of the separated elements. This violates the equilibrium conditions for which eq. (2) was obtained;

ii) The work function of the ionizator surface can be larger than the accepted reference value because of the oxidation of the surface;

iii) In our case due to the relatively slow process of diffusion in the ion source the violent jumps of density of vapors of the separated element do not occur.

The factor K, eq. (1), defining qualitatively this and other processes, under our conditions proves to be equal to about 4.

The change of the value of η_i for most of the REE depends only negligibly on the separation time.

c) Separation

The values of η_{D} and η_i presented in Figs. 2-6 provide the possibility to calculate by the formula (1) the separation efficiencies η for REE from the irradiated tantalum targets. As an example in Fig. 7 the value of η is given for different conditions of separation.
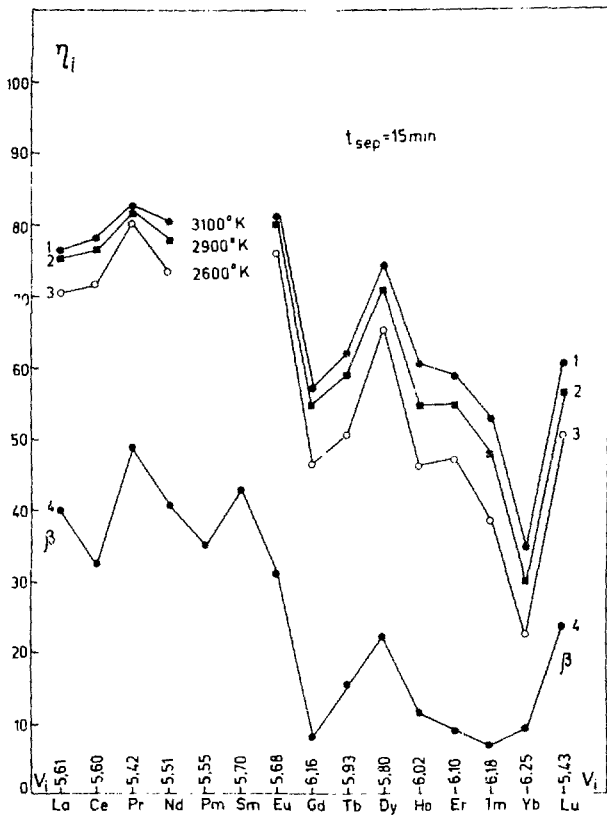
Fig. 6. Values of the ionization efficiency η_i of REE for different temperatures of rhenium ionizator: curves 1 - $T = 3100°K$, curves 2 - $T = 2900°K$, curves 3 - $T = 2600°K$. The separation time $t = 15$ min. The values of β are calculated by Saha-Langmuira formula for $\phi = 5.43$ eV, $T = 3100°K$. A is calculated individually for each element.
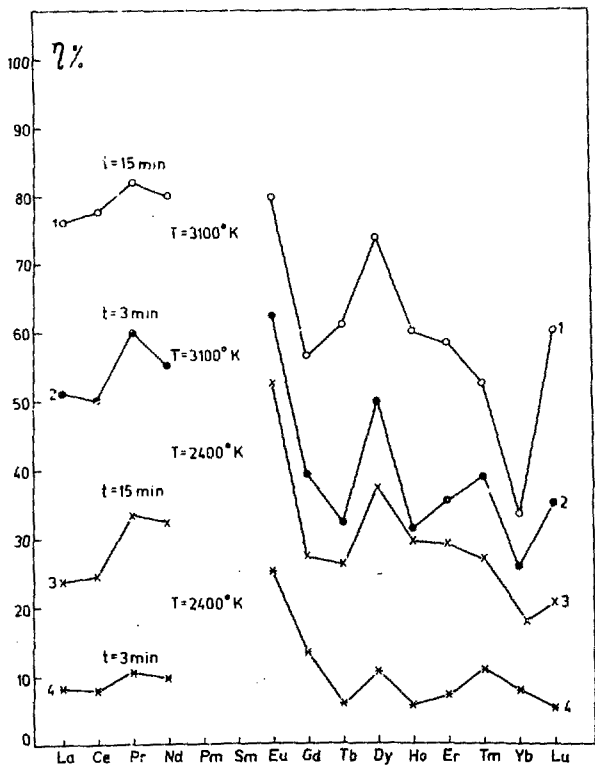
Fig. 7. Values of separation efficiences of REE calculated by the formula (1) for different working conditions of ISSI. Curve 1 - T = 3100°K, t = 15 min, curve 2 - T = 3100°K, t = 3 min, curve 3 - T = 2400°K, t = 15 min, curve 4 - T = 2400°K, t = 3 min. Temperature of ionizator T_{ion} = 3100°K.

Activity of the separated radioactive samples depends on the irradiation conditions, cross section of the isotope

production σ , half-life of the isotope under investigation, time of preparation of the source for separation and its efficiency. In our experiments it turned out that the separation time from two to three minutes and $T = 3000°K$ (see Figs. 7 and 8) were enough for obtaining the samples, reliable for spectroscopic measurements. Allowing for
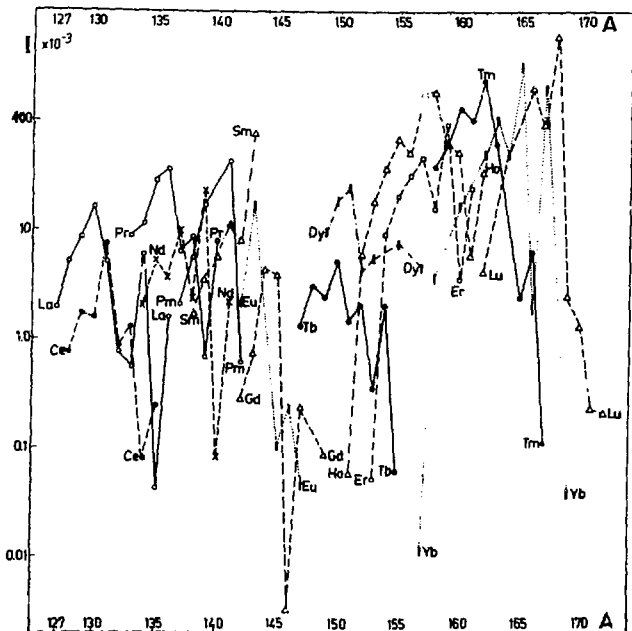

Fig. 8. Values of quantities I evaluated by formula (5) for separate isotopes of REE under the following conditions: the mass of the target 0.5 g, $\phi = 10^{11}$ protons/sec cm^2 the irradiation time $t_0 = 5$ min, the preparation of radioactive sources $t_n = 5$ min, the values of η are taken for temperature $T = 3100°K$ and the separation time $t = 3$ min.

the transportation time of the target and preparation of the source of mass-separator, we obtain the summary time from the end of irradiation to the end of separation not longer than $t_n \approx 4-6$ min. This summary time limits the application of the method for investigations of isotopes with the half-life $T_{\frac{1}{2}} < 1$ minute.

Activity of obtained on the collector of the mass-separator isotopes with the decay constant λ from target (N_0 atoms), irradiated during the time t_0 with the proton beam with intensity ϕ after the time t_n is determined by the formula:

$$I = N_0 \, \sigma \, \phi \, \eta \, e^{-\lambda t_n} \, (1 - e^{-\lambda t_9}) . \qquad (5)$$

On the basis of the values of η measured in the present work the values I for the short-living neutron-deficient REE are calculated according to formula (5). The results of the calculations for concrete experimental conditions are presented in Fig. 8. The values of η for Sm and Pm are calculated from the data indicated in works [3,9]. The cross sections of the isotope production σ are taken from work [10].

Figures 7 and 8 provide the possibility of selecting the most suitable conditions for obtaining any of indicated isotopes depending on the physical problem. By means of the data from Fig. 8 one can determine the isotopic structure in each of the separated isobars, facilitating by this the composition of the program of investigations and interpretation of the results obtained. In constructing Fig. 8 there was not taken into account the possibility of collection on the collector of the mass-separator, aside from elementary ions of the given element, also the ions of its oxides MO^+ contaminating the isobar with mass A by isobar with mass $A-16$. However, the measurements showed that under our conditions of separation only the oxides La and Ce (for these elements $MO^+/M^+ + MO^+ \approx 0.2$) are produced in significant amounts. Already for Pr and Nd the ratio $MO^+/M^+ + MO^+$ is of the order of 0.1 and for remaining REE it is even smaller. Such an amount of oxides does not change the interpretation of the spectra significantly.

Examples of Use of the Procedure for Spectroscopic Investigations

The application of the procedure under consideration provided the possibility of identification of new short-living isotopes and isomers, as well as to carry out a number of spectroscopic investigations. The examples of some results of measurements of isobars, obtained from the irradiated during the 5 minute period tantalum target with the thickness 0.05 mm, weight 0.5 g are presented in Table 3.

The attempts were made to use the methods for obtaining the short-living isotopes of other elements. The domain of our investigations which are going on is presented in Table 4.

The data from Table 4 indicate the possibility of using the methods for obtaining radioactive sources of the wide range of elements. This is shown by an example of obtaining the isotopes of In from stannum target. In this case during the irradiation by the protons of the stannum target the recoil nuclei are slowed down in tantalum foil. Further, this foil, as in the case of REE, is put into ISSI. The diffusion efficiency of the indium isotope from tantalum reached almost 100%. Due to the fact that ionization potentials of accompanying elements Cd , Ag , Pd , Rh , Ru are larger than 7 eV the pure indium isotopes were obtained on the collector of mass-separator. The obtaining of the indium isotopes and the noble gases are examples of the fact that using some modifications one can employ the procedure for obtaining the isotopes with different V_i and apply the targets with the low melting point.

Using the procedure in physical investigations for some problems one should carry out chemical separation of the obtained isobars after the process of separation. The necessary chemical operations in this case are simpler and significantly shorter than in the case of isolation of element from the irradiated target, because one has not get rid of the carrier [11].

<p style="text-align:center">**Table 3**</p>

Examples of the results obtained by the elaborated procedure.

A	Isotope $T_{1/2}$	New physical results	References
161	^{161}Yb(4.2 min)	new isotope $T_{1/2}=4.2\pm 0.2$ min	12
155	^{155}E (5.3 min)	E_{γ}	15
154	^{154m}Ho(3.2 min) ^{154}Ho(11.8 min)	E_{γ} extra new levels	15
153	^{153}Ho(9.3 min) ^{153m}Ho(2.0 min)	E_{γ}, decay scheme E_{γ}, decay scheme	16
152	^{152}Ho(2.7 min)	E_{γ}	15
151	^{151}Dy(17.0 min)	E_{γ}, decay scheme	16
150	^{150}Dy(7.1 min)	E_{γ}, decay scheme	13
149	^{149}Dy(4.6 min) ^{149m}Tb(4.2 min)	E_{γ}, new levels E_{γ}, decay scheme	16
148	^{148}Dy(3.5 min)	New isotope $T_{1/2}=3.5\pm 0.2$ min $E_{\gamma}=620.1$ keV	13
147	^{147m}Tb(1.8 min)	$E_{\gamma}=1397.2$ keV	17
142	^{142}Pr(1.6 min)	New isotope $T_{1/2}=1.6\pm 0.3$ min	14

Table 4
Examples of the use of the elaborated procedure.

Target	The elements obtained and some their characteristics				Type of the ion source
Material of the target	$T_{melting}$, °K	Elements	$T_{boiling}$, °K	Ionization potential V_i	
Ta	3273				
Re	3453	REE	$1773 \leqslant T_b \leqslant 3741$	$5.42 \leqslant V_i \leqslant 6.25$	ISSI
W	3653				
Ta	3273	Hf	4703	7.0	ISSI
Mo	2883	Zr,Nb,Y	$3573 \leqslant T_b \leqslant 4773$	$6.38 \leqslant V_i \leqslant 6.88$	ISSI
Th	1973	Kr, Xe,Ra	$120 \leqslant T_b \leqslant 211$	$10.75 \leqslant V_i \leqslant 14.0$	plasmic
Sn - Ta (recoil nuclei)	505	In	2373	5.785	ISSI

Conclusion

The elaborated procedure shortening the time of obtaining the radioactive isotopes of the rare-earth elements to about 5 minutes, provides the possibility for the detailed investigation of the decay of the isotopes of the rare-earth elements far from the stripe of beta-stability.

The results of the measurements for the efficiency of the separation process indicate the possibility of the further essential shortening of the time of obtaining the sources when working near the beam or on the beam of an accelerator.

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