

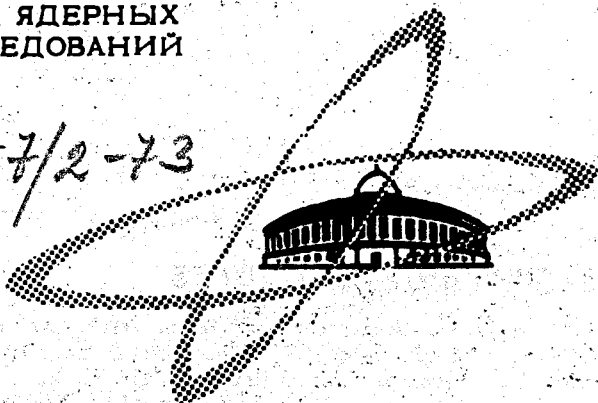
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HIGH EFFECTIVE METHOD  
FOR ACTINIDE ISOTOPE SEPARATION

ЛАБОРАТОРИЯ ЯДЕРНЫХ ПРОБЛЕМ

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**HIGH EFFECTIVE METHOD  
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Submitted to Radiochemical  
and Radioanalytical Letters

Объединенный институт  
ядерных исследований  
БИБЛИОТЕКА

## Introduction

A wide application of the electromagnetic isotope separators for the actinide element investigation (isotopic analysis and enrichment of isotopes, preparation of sources for nuclear spectroscopic investigations, etc.) was restricted by a large loss of the sample material because of the low efficiency of the usual gas discharge ion sources with internal chlorination (or fluorination) of the sample.

The pipe-type surface ionization ion source developed at the Laboratory of Nuclear Problems of the Joint Institute for Nuclear Research /1/ is characterized by a high separation efficiency and a short separation time for small amounts of rare earth isotopes. It was found that the efficiency of isotope separation,  $\eta$ , is close to 100% at the atomic ionization potential values of  $V_i \leq 5$  eV, and decreases to ~1% for  $V_i = 7$  eV. In accordance with the calculated /2/ and experimentally obtained /3,4/ ionization potential values of the actinide elements, high effectiveness is also expected for the separation of the lanthanide analogous actinide isotopes (see Table I).

The aim of this work was to investigate the efficiency of the ion source mentioned above for the preparation of monoisotopic samples of some actinide elements. This was important in the experiments on /6/ the search for transplutonium elements in the uranium targets irradiated with 70 GeV protons.

## Experimental

The experiments have been carried out with the electromagnetic isotope separator of the "YASNAPP" facility /7/. The separation efficiency,  $\eta = N^+ / N^0$ , was determined by measuring (with a bell-type proportional GM counter, VAZ-520) the alpha or beta activity,  $N^0$ , of the sample before separation and the corresponding activity,  $N^+$  on the collector foil, after separation. Taking into account the transmission coefficient of the isotope

separator ( $\sim 0.9$ ) and the remaining part of the sample in the ion source ( $< 10\%$ ) one can roughly estimate the surface ionization coefficients of  $\beta \sim 1.2 \eta$ .

In all experiments for mass scaling a few tenths of a microgram of  $U_3O_8$  mass marker were added to the samples. A rough estimate of the sample temperatures in the zone of the ionizator and evaporator, as a function of the electron bombardment power  $P$ , can be done from Fig.1. The cathode powers of the ionizator and evaporator were generally 250 and 150 W, respectively. Varying the cathode power it is possible to realize a temperature distribution which differs from that shown in Fig.1.

ACTINIUM-225 was isolated from a thorium target irradiated with 660 MeV protons. The nuclear reaction products, including the actinium, were separated from the target material by anion exchange chromatography. The target was dissolved in nitric acid solution containing one drop of hydrogen fluoride. Then methanol was added to get a solution which contains about 70% of methyl alcohol. This solution was transferred to a column filled with anion exchanger, Dowex 1x8, in the nitrate form. A solution of 65%  $CH_3OH$ -0.5 M  $HNO_3$  was used as an eluent. Results of the model experiment for the separation of actinium and radium from a macroamount of thorium are shown in Fig.2. For further purification from spallation and fission products, actinium, together with rare earth and alkaline earth isotopes, was coprecipitated with one milligram of calcium by adding hydrofluoric acid. The fluoride was dissolved in 0.15 M  $HCl$  saturated with boric acid. The chromatographic resolution of the components of this mixture was made on a 2 mm x 60 mm cation exchange column filled with Aminex A5 in the ammonium form. After sorption the individual elements were eluted with ammonium  $\alpha$ -hydroxyisobutyrate; the actinium elutes after lanthanum and lead (see Fig.3). If the target processing is carried out 10-15 days after the irradiation the actinium fraction contains practically  $^{225}Ac$  only.

THORIUM-234 was isolated from natural uranium by the well known method of coprecipitation with ferric hydroxide. Separation from the iron was made on an anion exchange column.

URANIUM-233 and PLUTONIUM-239 were commercially available and used after further purification. The purification of  $^{233}U$  and  $^{239}Pu$  has been provided by preliminary isotope separation of the initial material.

CURIUM-240 was produced by the nuclear reaction:  $^{232}Th ( ^{12}C, 4n ) ^{240}Cm$ . The upper layer of the thorium target was dissolved and the solution was processed in the manner described above for the actinium. The curium elutes from the cation exchange column together with the promethium, but the latter is beta-active only and does not disturb the measurement of the curium alpha-activity.

CALIFORNIUM-246 was produced by bombardment of  $U_3O_8$  target with  $^{12}C$  ions:  
 $^{238}U ( ^{12}C, 4n ) ^{246}Cf$ .

The target was dissolved in hydrochloric acid and the solution was transferred to an anion exchange resin bed in the chloride form. The nuclear reaction products were eluted with 6 M  $HCl$ . The separation of the californium was made in the above described way. It is seen from Fig.3 that the californium elutes between the terbium and gadolinium.

For loading into the ion source of the isotope separator the actinide elements were deposited as hydroxides by electrolysis onto 2 mm x 5 mm tungsten foil. The hydroxides were transformed to the corresponding oxides by heating the foil up to 500 °C. The radiochemical purity of the samples was checked via alpha spectrum measurements with a surface-barrier detector.

## Results and Discussion

Our experimental results are summarized in Table II. It was previously shown that the maximum efficiency for the rare earth isotope separation was obtained at a three minutes duration of separation. As for the actinide elements, the dependence of the separation efficiency on the separation time was investigated for the actinium, plutonium and californium only. It can be seen from Fig.4 that the separation efficiency for the actinium depends considerably on the temperature of the ion source and reaches a saturation value in about twenty minutes. The californium entirely evaporates from the tungsten foil at a temperature of 1150 °C. Consequently, it may be supposed that after putting on the ion source, a part of the californium is evaporated as early as the ion source reaches a temperature needed for an effective surface ionization.

Probably in the case of a suitable temperature gradient over the ion source, the efficiency for the californium should be higher. For the other actinide elements the time for isotope separation was arbitrarily selected.

Fig.5 shows the differential Pu, <sup>210</sup>Ac<sup>+</sup> and Cf<sup>+</sup> yields as a function of the separation time which give the possibility to estimate the total efficiency for these elements. For this purpose one has to account the heat-up time of the ion source being equal to 1.5 min. (see e.g. the curve for Cf). The slope of the straight line depends on the thermal characteristics of the investigated elements and on the ion hold-up time in the source:

$$r = 4\Omega / SV,$$

where  $\Omega$  is the internal volume of the ion source;  $S$  is the outlet hole area;  $V$  is the ion velocity in the source. The hold-up time for the oxide ions turned out to be higher than for the metallic ions. The ratio  $X^+/XO^+$  changes in a wide range and depends on the ion source temperature <sup>18/</sup>.

The possibility of the mentioned ion source <sup>11/</sup> to separate macroamounts is shown in Fig.1 at the total Nd ion current  $I_{\Sigma}$  as a function of the electron impact power to the ion source. An ion current density of about 0.3 a/cm<sup>2</sup> has been obtained (exit hole is 0.3 mm in diameter). Ion current of admixtures equals 10% of the total ion current.

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T A B L E I

Known values for the first ionization potential of some actinide elements

ELEMENT		Ac	Th	U	Np	Pu	Am	Cm	Bk	Cf
Ionization potential $V_i$ (eV)	theor. 2/	4.92	5.62	5.4I	5.49	5.II	5.65	5.67	5.49	5.4I
	exper. 3/	-	7.5	6.I9	6.I6	5.7I	6.0			
	exper. 4/	$\begin{matrix} 6.86 \\ \pm 0.6 \end{matrix}$	$\begin{matrix} 6.95 \\ \pm 0.06 \end{matrix}$	$\begin{matrix} 6.03 \\ \pm 0.08 \end{matrix}$						
Melting point °C 5/		1050	1750	1132	637	640	1176	1340	986	

T A B L E 2

Measured efficiencies for the electromagnetic separation of the actinide isotopes

S A M P L E materi- al	atoms	Separation time/min	Electron impact power/w	Separation efficiency $\eta$ /%				$X^+ / XO^+$
				$X^+$	$XO^+$	$XO_2^+$	$X^+ + XO^+ + XO_2^+$	
ACTINIUM	$4 \cdot 10^8$	40 18	415 390	63.6	16.5	-	80.1	3.9
				33.4	10	-	43.4	3.3
THORIUM	$2 \cdot 10^8$	15	480	0.2	5	-	5.2	0.04
URANIUM	$4 \cdot 10^{12}$	20	425	2.2	6.2	0.6	9	0.35
PLUTONIUM	$2 \cdot 10^{12}$	35	430	43.5	7.5	-	51	5.8
CURIUM	$3 \cdot 10^6$	35	480	68	3.6	0.4	72	19
CALIFOR- NIUM	$10^5$	6	430	30	7.5	-	37.5	5



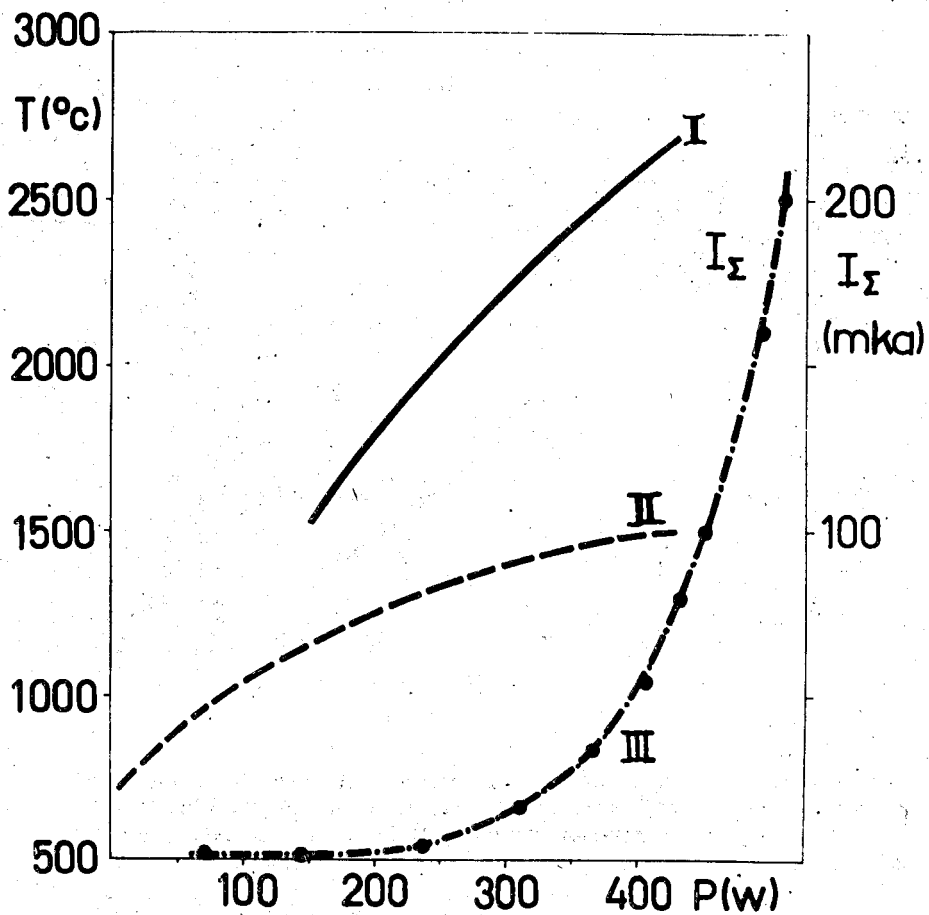


Fig.1. Ionizer temperature (I) and evaporator temperature (II) as a function of the electron impact power to the ionizer. Curve III: current of Nd ions as a function of the power to the ion source.

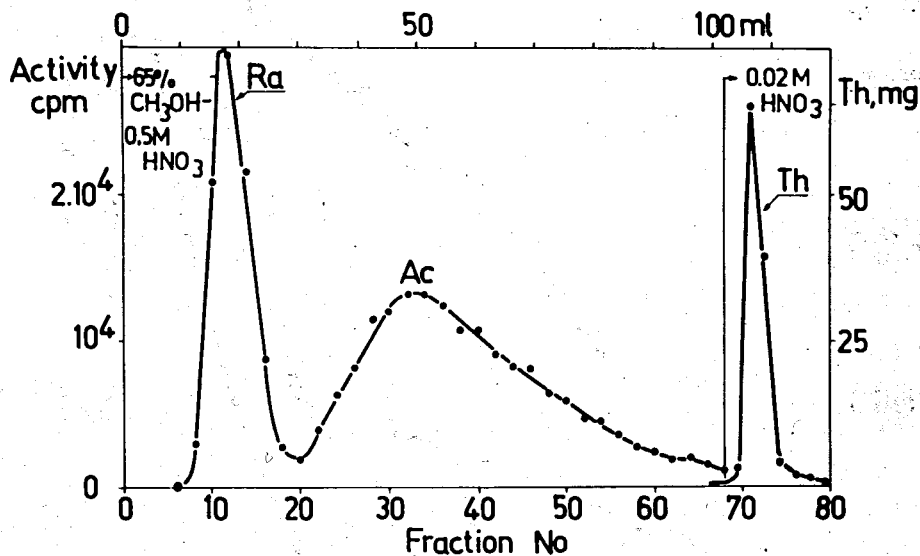


Fig.2. Chromatographic separation of radium and actinium from 100 mg of thorium. Column: 6 mm x 235 mm Dowex: 1x8(200-400 mesh,  $NO_3^-$  form). Eluents: 65%  $CH_3OH$ -0.5 M  $HNO_3$  for radium and actinium, and 0.02 M  $HNO_3$  for thorium. Flow rate 1 ml/cm<sup>2</sup> min,  $t = 22^\circ C$ .

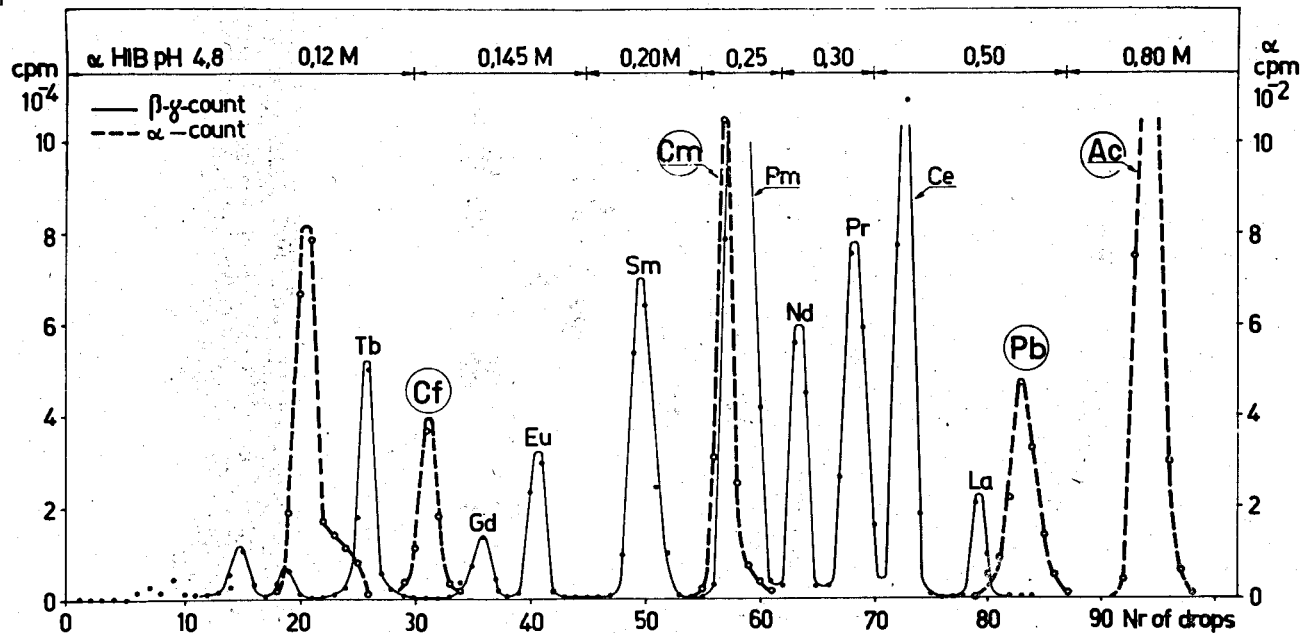


Fig.3. Chromatographic resolution of some lanthanide and actinide elements produced in the nuclear reactions. Column: 2 mm x 60 mm Aminex A5 (-400 mesh,  $NH_4^+$  form). Eluent: ammonium  $\alpha$ -hydroxyisobutyrate, pH = 4.7. Flow rate: 2 drops/min.  $t = 22^\circ C$ .

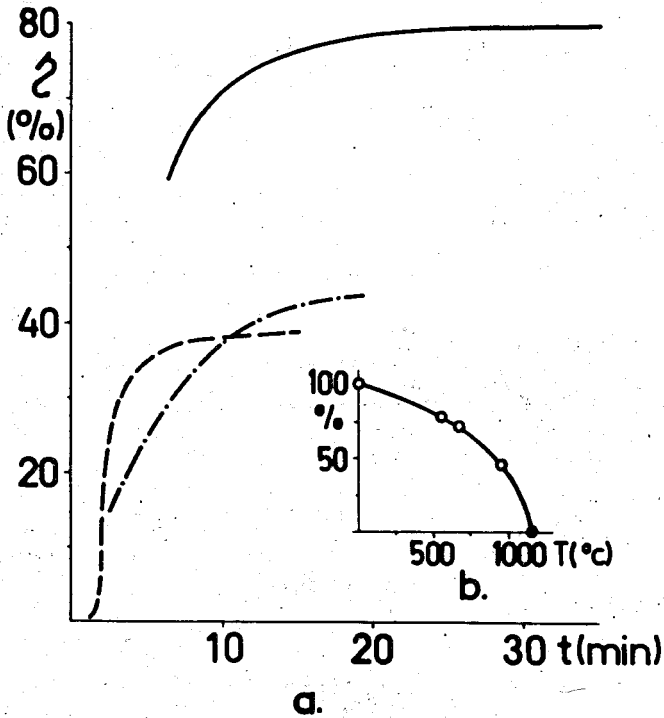


Fig.4. a) Separation efficiency  $\eta$  as a function of the separation time for different source powers and charge materials. Full curve is charge material Ac, power 415 W; dash-dotted curve is charge material Ac, power 390 W; Dashed curve is charge material Cf, power 450 W. b) Part of Cf sample remaining in the ion source after a 15 min separation for different evaporation temperatures.

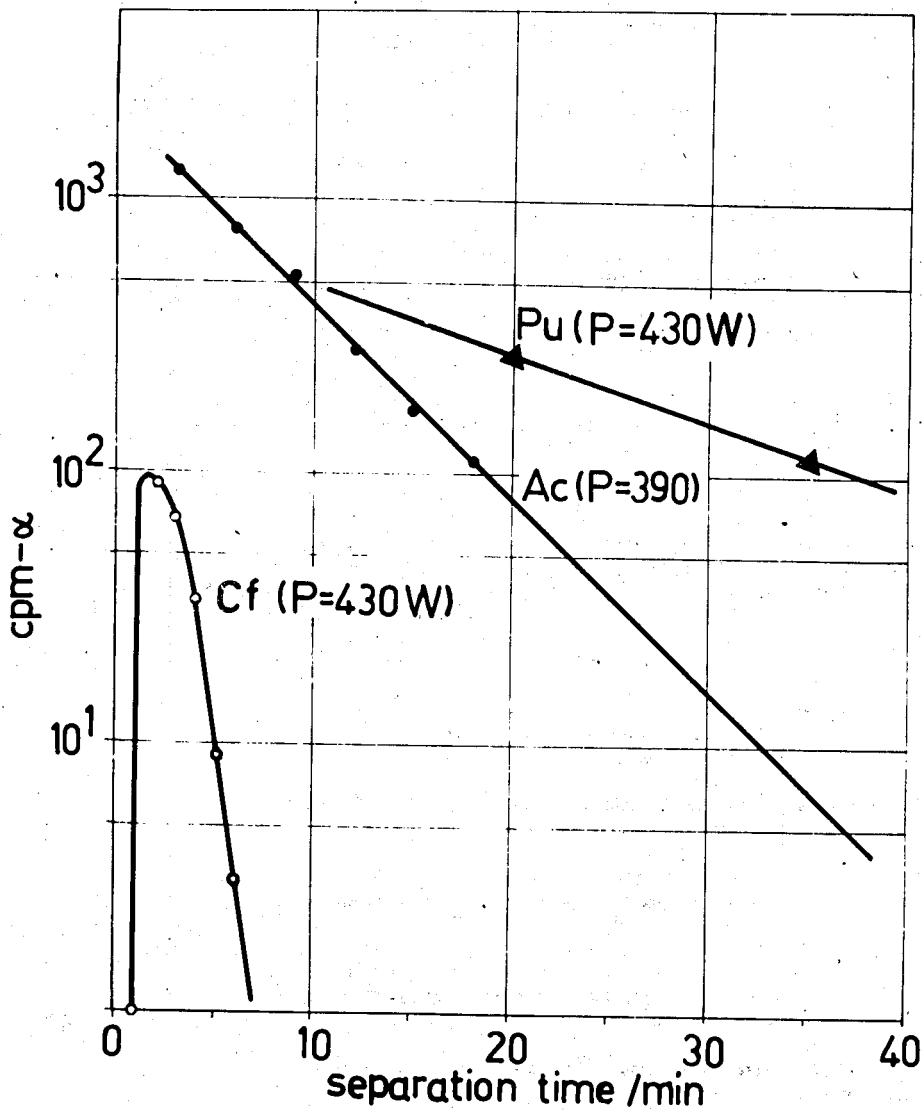


Fig.5. Differential yields from the ion source for Pu, Ac and Cf as a function of the separation time. Initial sample activities has been 880 cpm, 9200 cpm and 547 cpm for Pu, Ac and Cf, respectively.