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**SUBLIMATION  
OF VOLATILES AS A METHOD  
OF RADIOCHEMICAL SEARCH  
FOR SUPERHEAVY ELEMENTS  
IN A URANIUM TARGET BOMBARDED  
WITH XENON IONS**

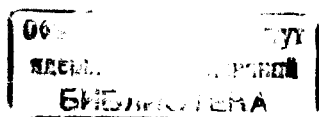
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*Submitted to "Радиохимия".*



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Возгонка летучих элементов как способ радиохимического поиска сверхтяжелых элементов в урановой мишени, облученной ионами ксенона

Металлическая урановая мишень нагревалась в кварцевой трубке последовательно в токе азота, водорода и воздуха при температуре 950-1100°C. Продукты возгонки осаждались на охлаждаемых сборниках. За 3 часа было собрано более 90% радиоактивных атомов Tl, Pb, Cd, Zn, Xe, J и Os, первоначально присутствующих в облученной мишени.

Коэффициент очистки от актиноидов составил больше  $10^6$ .

Работа выполнена в Лаборатории ядерных проблем ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна 1978

Reetz T. et al.

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Sublimation of Volatiles as a Method of Radiochemical Search for Superheavy Elements in a Uranium Target Bombarded with Xenon Ions

A metallic uranium target was heated in a quartz tube successively in the flows of nitrogen, hydrogen and air at a temperature of 950-1100°C. The volatilized products were deposited on cooled collectors. After 3 hours more than 90% of the radioactive atoms of Tl, Pb, Cd, Zn, Xe, J, and Os contained initially in the irradiated target were collected. The separation coefficient of actinides was more than  $10^6$ .

The investigation has been performed at the Laboratory of Nuclear Problems, JINR.

Preprint of the Joint Institute for Nuclear Research. Dubna 1978

Attempts to produce the predicted superheavy elements (SHE) by nuclear reactions are presently being made in several laboratories of the world. As early as in 1964 Flerov<sup>/1/</sup> pointed out that acceleration of very heavy ions such as xenon might open up wide possibilities for producing new nuclei. The bombardment of a uranium target with such ions may lead to the formation of not only new isotopes of known elements, but also of new elements<sup>/2/</sup>.

In 1971-1975 an extensive work on the search for SHE in the products of bombardment of uranium with xenon<sup>/3,4/</sup> was performed at the JINR Laboratory of Nuclear Reactions. Some attempts were made to radiochemically separate the comparatively long-lived isotopes of SHE from the target substances. In the present paper one of the techniques used is described, which is based on the expectation that superheavy elements 112-118 should be relatively volatile and even more volatile than their proposed homologues Hg to Rn. In particular, all of them might be expected to be more volatile than thallium<sup>/5/</sup>. Therefore if thallium and other volatile elements are vaporized from the target material, the conditions of the experiments seem to be favourable for the separation of SHE. The technique should foresee the effective destruction of the crystalline structure of uranium as this considerably enhances the diffusion of the elements through the

matrix of the material<sup>6/</sup>. In addition, it is necessary to provide the efficient separation of volatile elements from actinides since SHE are searched for by detecting their spontaneous fission.

The experiments described below were carried out in the first half of 1975.

### EXPERIMENTAL

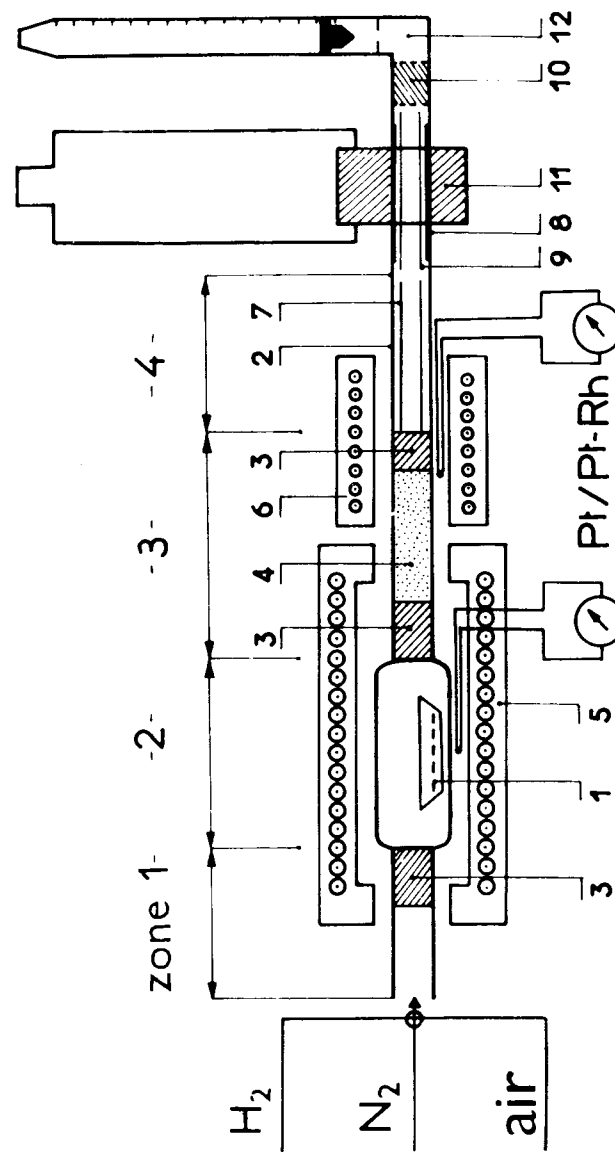
A 50  $\mu\text{m}$  uranium foil glued onto a copper backing with Wood's alloy was bombarded with a xenon ion beam of  $(1.5-10) \cdot 10^9$  ions/sec intensity and 980 MeV energy at a tandem cyclotron for 8 hours. The integral ion flux was equal to  $4.5 \cdot 10^{15}$  ions.

The target was treated using the set-up shown in the figure. It consisted of a quartz thermochromatographic column (2), heated by tubular furnaces (5) and (6), and of two traps - a low-temperature trap (11) and a trap filled with carbon (10) for catching highly volatile products.

Hydrogen was produced in an electrolyzer, dried by concentrated sulphuric acid and purified from oxygen by passing it over a palladium catalyst. The final drying was performed using a liquid nitrogen trap. The commercial nitrogen was purified from oxygen by passing it over incandescent copper and dried in a trap cooled with liquid nitrogen. The air dried by freezing with liquid nitrogen was supplied by membrane pumps.

The quartz column having an inner diameter of 4 mm contained a quartz sand filter (4) supported by quartz wads (3). The metallic foils (7) on which the vaporized products were deposited were also placed in the column.

To collect gaseous elements the quartz column was connected with a teflon tube cooled by liquid nitrogen. The inner walls of the tube were coated with a mylar film (9) which served to detect spontaneous fission events. Trap (10) filled with activated charcoal was placed in front of flowmeter (12).



Experimental set-up. 1 - irradiated uranium target; 2 - quartz tube; 3 - quartz wadding; 4 - quartz powder, 100-15  $\mu\text{m}$ ; 5 - furnace; 6 - furnace; 7 - metallic foil; 8 - teflon tube; 9 - spontaneous fission track detector (mylar); 10 - activated charcoal trap; 11 - liquid nitrogen-cooled copper trap; 12 - flowmeter.

Before the beginning of the experiment the quartz column, sand and wadding were heated at a temperature of  $1100^{\circ}\text{C}$  in the air flow for 30 minutes and in the hydrogen flow for 30 minutes. A  $1.7\mu\text{m}$  foil of copper alloy containing 4.5% Ni, 4.5% Al, 2.5% Mg and 0.7% Cr was placed into the column and annealed at  $700^{\circ}\text{C}$  for 50 minutes in the hydrogen flow.

Before the experiment the irradiated  $20\times 8\text{ mm}^2$  uranium target (3) was refined from most of Wood's alloy by placing it into hot water. After that the dry target was inserted into the quartz column. The target was first heated by furnace (5) at  $950^{\circ}\text{C}$  for 60 minutes in the nitrogen flow at a gas flow of 20 ml/min (different nitrides of uranium, mainly  $\text{U}_2\text{N}_3$ , were expected to be produced during this processing), and then for 60 min in the hydrogen flow at  $1100^{\circ}\text{C}$ . Most of the nitrides were to convert to UN. Then furnace (5) was switched off, furnace (6) was moved to the left and the quartz sand was heated to  $870^{\circ}\text{C}$  in the centre of the furnace ( $800^{\circ}$  at the edge of the copper alloy foil) for 60 minutes in the hydrogen flow.

Then furnace (6) was turned off, the column was cooled to room temperature after disconnecting the teflon tube, the copper alloy foil was removed and its gamma-radiation spectrum was measured. Then the foil was placed into a spontaneous fission counter. The gamma-radiation spectra of the target and of different sections of the tube were measured using a lead collimator.

A platinum foil was placed into the column and the column was connected to teflon tube (8) again. The target was heated by furnace (5) to  $1100^{\circ}$  in the hydrogen flow. After several minutes the nitrogen was replaced by air and the target was heated in the air flow for 60 min at  $1100^{\circ}\text{C}$ . During this treatment uranium nitrides were expected to yield  $\text{U}_3\text{O}_8$ . Then furnace (5) was switched off, furnace (6) was again moved to the left, and the

sand was heated in the air flow for 60 min at  $940^{\circ}$  in the furnace centre.

After cooling the system the teflon tube was disconnected from the column, the ends of the tube were closed by copper stoppers and the gamma-radiation spectrum of the tube was measured. Thereafter the trap was disconnected and the tube was placed into a Dewar vessel filled with liquid nitrogen.

The platinum foil was removed from the column to measure the gamma-ray spectrum and placed into the apparatus to measure its spontaneous fission activity.

The quartz column was cut to several sections, and the gamma-radiation spectra of each individual section and of the remainder of the uranium target were measured.

## RESULTS

The volatilization yields of some products from the target are shown in table 1. The results were obtained by comparison of the over-all gamma-radiation spectra of the target measured before and after the experiment. The spectrum was measured also after heating the target in nitrogen and hydrogen. Two Ge(Li)  $\gamma$ -ray detectors,  $40\text{ cm}^3$  and  $24\text{ cm}^3$  in volume, were used and the species were treated (the energies and peak areas were measured) using a Minsk-32 and TPA computers. The data on volatilization yields obtained in two series of measurements agree fairly well. After heating in nitrogen and hydrogen most of volatile elements were already extracted. The volatilization rate depends not only on the volatility of the elements, but also on the conditions of their transportation in the target material and on the character of their interaction with uranium. That was probably the reason why only 70% of xenon and a small amount of iodine were removed from the target at the first stage of the treatment.

Table I

Percentage volatilization yields of isotopes from the target

Isotope	Energy (keV)	After heating in N <sub>2</sub> and H <sub>2</sub>	After the experiment
<sup>237</sup> U	208	< 0.01	< 0.01
<sup>239</sup> Np	278	< 0.01	< 0.01
<sup>143</sup> Ce	294	< 0.01	< 0.01
<sup>115m</sup> Cd	492	90	100
<sup>111</sup> In	171	-	100
<sup>69m</sup> Zn	439	85	100
<sup>200</sup> Tl	367	80	100
<sup>131</sup> I	364	8	90
<sup>212</sup> Pb	238	40	90
<sup>132</sup> Te	228	8	60
<sup>135</sup> Xe	250	72	95
<sup>103</sup> Ru	497	0	80
<sup>136</sup> Cs	817	0	50
<sup>99m</sup> Tc	140	0	55

Table 2 shows the distribution of individual products at the end of the experiment.

Such elements as Y, La and lanthanides, Ac and actinides, Zr and Hf, alkali and alkali-earth metals were either not vaporized at all, or were irreversibly adsorbed on quartz. All the uranium compounds, U<sub>2</sub>N<sub>3</sub>, UN and U<sub>3</sub>O<sub>8</sub>, produced during the experiment are involatile. On the contrary, the elements of the main groups of the sixth period, being highly volatile in the elemental state (and in the form of oxides), were precipitated on the copper alloy and platinum foils.

The gaseous Xe and Rn were caught in the liquid-nitrogen cooled teflon tube having the inner mylar coating.

For the purpose of detecting SHE the tube was stored in liquid nitrogen for a long time.

The elements involatile in the elemental state, but which form volatile oxides could be precipitated on the platinum foil. These elements are Tc, Ru, Os, Jr and partly Pt.

Table 3 shows the distribution of some of the products over different zones of the quartz tube. One can see that technetium and ruthenium were not desorbed fully from the quartz sand in air at 900°. Cesium is evaporated from the target material both in the elemental form and as an oxide, but irreversibly adsorbed on quartz. The behaviour of lead is somewhat different from the usual behaviour of the microquantities of this element - about 7% of Pb were deposited on quartz. This may be due to, first, the presence of lead macroquantities coming from the residue of Wood's alloy. Thus usually results in a partial deposition of lead due to its reaction with quartz. In addition, the distribution of lead was measured by tracing the isotope <sup>212</sup>Pb (E = 238 keV) with a half-life of 10.5 hours. It is possible that <sup>212</sup>Pb is in part a daughter product of <sup>224</sup>Ra (T<sub>1/2</sub> = 3.2 d), while the behaviour of radium in the elemental state is similar to that of cesium (see table 3).

The gamma-radiation spectra of the quartz sand measured after the experiment did not exhibit the lines due to <sup>237</sup>U, <sup>237</sup>Np, <sup>238</sup>Np as well as x-rays of U, Np, Pu and Am, the areas of their peaks being < 10<sup>2</sup> counts. In the target gamma-radiation spectrum the most intensive lines due to uranium and neptunium contained 10<sup>6</sup> counts. Hence it follows that the separation coefficient of volatiles from uranium and neptunium on the section between the target and the quartz sand is not less than 10<sup>4</sup>.

Table 2

Percentage distribution of products

Isotope	$\beta$ -ray energy (keV)	On copper alloy foil	On Pt foil	In traps (I0) and (II)	The isotope residue in the target and on quartz in the high-temperature zone
$^{237}\text{U}$	208	< 0.01	< 0.01	0	100
$^{239}\text{Np}$					
$^{239}\text{Np}$	278	< 0.01	< 0.01	0	100
$^{143}\text{Ce}$	294	< 0.01	< 0.01	0	100
$^{136}\text{Cs}$	817	< 0.01	< 0.01	0	100
$^{200}\text{Tl}$	367	80	20	0	0
$^{212}\text{Pb}$	238	40	40	0	20
$^{203}\text{Pb}$	279	45	55	0	x)
$^{115\text{m}}\text{In}$	336	90	10	0	0
$^{111}\text{In}$	171				
$^{115\text{m}}\text{Cd}$	492	90	10	0	0
$^{69\text{m}}\text{Zn}$	439	85	15	0	0
$^{135}\text{Xe}$	250	0	0	95	5
$^{222}\text{Rn}$	352	0	0	100	x)
$^{103}\text{Ru}$	497	0	80	0	20
$^{99\text{m}}\text{Tc}$	140	0	55	0	45
$^{132}\text{Te}$	228	8	50	0	40
$^{191}\text{Os}$	130	0	100	0	x)
$^{131}\text{I}$	361	8	80	2	10

x) Due to the relatively low activity of the isotope, the lines due to  $\beta$ -radiation are unseen in the target spectrum. Therefore the sum of the  $\beta$ -activities on the collectors is taken to be 100.

Table 3

The distribution of some products in zones I, 2 and 3 of the quartz tube in per cent of the total activity.

Isotope	Energy (keV)	Zone I	Zone 2 without target	Zone 3
$^{99\text{m}}\text{Tc}$	140.5	0.02	0.07	0.1
$^{103}\text{Ru}$	497	0	< 0.03	0.2
$^{136}\text{Cs}$	817	25	8	15
$^{212}\text{Pb}(+^{224}\text{Ra})$	238	4	1	2
$^{200}\text{Tl}, ^{115\text{m}}\text{In},$ $^{115}\text{Cd}, ^{203}\text{Pb},$ $^{69\text{m}}\text{Zn}$		0	0	0

On the quartz sand filter the concentration of actinides is expected to decrease sharply, but no additional experimental data on uranium and neptunium have been obtained. However, if one estimates the lower limit of the separation coefficient from uranium on quartz sand on the basis of the behaviour of cesium, the total separation coefficient for uranium will exceed  $10^6$ . Consequently less than  $1.5 \times 10^{-7}$  g of uranium were precipitated on the metallic foils, which indicates, on the average, one spontaneous fission event per 50 years.

Thus, the sublimation method described permits the efficient extraction of volatile products from a uranium target at practically complete separation from uranium and other actinides. This makes it possible to search with a maximum sensitivity for SHE in the products of nuclear reactions by detecting spontaneous fission events.

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#### REFERENCES

1. Flerov G.N., Karnaukhov V.A. In : C.Rendus du Congrès Intern. de Physique Nucléaire, Paris, 1964, p.
2. Flerov G.N. Atom. Energiya, 1969, 26, p.137.
3. Flerov G.N., Oganessian Yu.Ts. J.Inorg.Nucl.Chem., 1976, Suppl., p.131.
4. Flerov G.N. In: Proc. Intern. Conf. on Reactions Between Complex Nuclei, Nashville, 1974, v.2, p.459.
5. Eichler B. JINR, P12-7767, Dubna, 1974.
6. Eichler B. J.Inorg.Nucl.Chem., 1973, 35, p.4001.

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