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EXPERIMENTS ON CHEMICAL PROPERTIES
OF ELEMENT 102

1. Adsorption of a Chloride of Element 102
from the Gaseous Phase

ЛАБОРАТОРИЯ ЯДЕРНЫХ РЕАКЦИЙ

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Опыты по химии элемента 102. 1. Адсорбция хлорида 102 элемента из газовой фазы.

Впервые изучались химические свойства элемента 102. Проведено параллельное сравнение адсорбционных свойств хлоридов Tb, Cf, Fm и $^{256}102$ в условиях, использовавшихся при химической идентификации 104 элемента. Показано подобие адсорбционных свойств хлорида 102 элемента и хлоридов элементов III группы. Получено дополнительное подтверждение резких различий в летучести хлоридов 102 и 104 элементов.

**Препринт Объединенного института ядерных исследований,
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Experiments on Chemical Properties of Element 102.

1. Adsorption of a Chloride of Element 102 from Gaseous Phase

Chemical properties of element 102 have been first investigated. The adsorption properties of Tb, Cf, Fm and $^{256}102$ chlorides have been compared under conditions used while chemical identification of element 104. The adsorption properties of the element 102 chloride are shown to be similar to that of the chlorides of elements of group III. A sharp difference in volatility between the chlorides of elements 102 and 104 has been confirmed once more.

**Preprint, Joint Institute for Nuclear Research,
Dubna, 1967.**

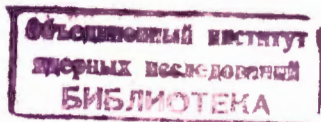
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EXPERIMENTS ON CHEMICAL PROPERTIES
OF ELEMENT 102

1. Adsorption of a Chloride of Element 102
from the Gaseous Phase



First information about chemical properties of element 102 was probably obtained in paper^{1/x/}. The paper dealt with the study of chemical properties of element 104—Kurchatovium. In spontaneously fissioning products obtained from the bombardment of ^{242}Pu with ^{22}Ne along with $^{260}_{104}$ there is an isotope of element 102 with mass number 256, $T_{1/2} = 6+2 \text{ sec}$ ^{3/}. (This nuclide is mainly alpha-active, however, in about 1/150 of events it undergoes spontaneous fission^{4/}). The counting rate of fission fragments of nuclides $^{260}_{104}$ and $^{256}_{102}$ is approximately the same in the interaction products: one event per several hours of bombardment. In experiments on chemical properties of kurchatovium the molecules of the chloride of $^{260}_{104}$ were transported by a gas stream to the fission fragment detectors through a 4-meter-long tube and an adsorbing filter, which adsorbed, as was shown, chlorides of elements of group III (curium and californium).

Fission fragments of $^{256}_{102}$ nuclei were not registered by these detectors. The chloride of element 102 was supposed to be adsorbed as well, since according to predictions available this element should be an "actinide"^{5/} or a "curide"^{6/}.

In principle, the absence of the tracks in the detector could be due to a great volatility of the chloride of element 102 (half-life is about 6 sec, while the counting time is about 0.7 sec). One could not make final conclusions about chemical properties of the new element without excluding this possibility.

Hence, the question whether element 102 is analogous to the elements of group III or is significantly different from them was not unambiguously answered.

x/ Paper^{13/} informing about the discovery and the study of chemical properties of nobelium appeared erroneous^{14,3,9,15,16,17/}.

Therefore we have performed special experiments on $^{286}102$ with the aim of checking the concepts concerning chemical properties of element 102 and the structure of period VII of the Mendeleev periodic table. The purpose of the present paper is to compare the behaviour of element 102 with those of Tb, Cf and Fm during chlorination and adsorption.

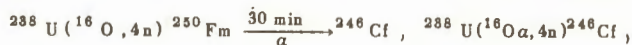
Experiment

The experiments were performed in the JINR Heavy Ion Cyclotron (U-300) by means of an apparatus for a fast continuous separation of nuclear reaction products which was placed in the chamber of the accelerator^{7/}. A block-diagram of the apparatus is shown in Fig. 1.

A thin target ($900 \mu\text{g} / \text{cm}^2$) (1) was bombarded with a heavy ion beam (2). The recoil atoms were slowed down in the stream of nitrogen continuously flown behind the target (volume-3) and then chlorinated by means of $\text{NbCl}_5 + \text{ZrCl}_4$ or NbCl_5 vapors.

The chlorides are transported by a flowing gas with different speeds and, as a result, distributed between subsequent parts of the apparatus. The experimental conditions (temperature, partial pressure of chloride vapors, the material of the gas conduit walls) are given in Table 1.

Alpha-active isotopes ^{140}Tb , $T_{1/2} = 4.1\text{h}$ (10% alpha-decay); ^{246}Cf , $T_{1/2} = 37\text{h}$; ^{250}Fm ; $T_{1/2} = 30\text{min}$; ^{252}Fm , $T_{1/2} = 23\text{h}$ and $^{286}102$, $T_{1/2} = 6\text{sec}$ were used in the experiments. These isotopes were produced from the bombardment of a complex target consisting of the oxides of ^{238}U and ^{144}Sm (95% enrichment), the weight ratio being 9:1. Oxygen-16 and neon-22 were used as projectiles (at energies given in Table 1). Californium, fermium and element 102 were obtained as a result of the following reactions and processes:^{8,9/}



^{148}Gd and ^{149}Tb were formed by nucleon transfer reactions in the bombardment of ^{144}Sm .

Data on the effective cross sections for the above reactions^{6/} indicate that in the prolonged bombardment with 110 MeV ^{22}Ne ions about 40% of ^{282}Fm was produced as a result of $^{286}\text{102}$ decay. Similarly, a significant part of ^{246}Cf was produced as a result of ^{260}Fm decay after the bombardment with ^{16}O ions.

The amounts of ^{149}Tb , ^{246}Cf and ^{282}Fm were measured in a 4-meter long tube (4), in an inert filter (5) and in chemical filter (6) or in a trap (7) where the chlorinating agent was condensed. For this purpose the apparatus was dismantled after the bombardment and the nuclear reaction products were washed from each part separately. The washing solutions contained (of La (1mg), of Ce (10mg) as carriers and ^{188}Eu used for determining chemical yield. Then LaF_3 and CeF_3 were precipitated, oxydation by KBrO_3 was performed and Ce was separated through the precipitation of iodate. For further purification a partition reversible chromatographic column, 80x3 mm, with di-/2-ethyl-hexyl/-phosphoric acid on teflon (0,25 ml/g) was used. Lanthanum was eluted at the room temperature with 0,5 HNO_3 californium, fermium, terbium and europium were eluted with 3N HNO_3 simultaneously and the solution was evaporated on the platinum disk.

The isotopes obtained were identified on the basis of their alpha-particle energies and half-lives.

Alpha-activities of the samples were measured spectrometrically by means of 2,5 cm² semi-conductor detectors and a 100-channel amplitude analyser. The detector efficiency was about 20 per cent, the equipment stability made it possible to perform measurements for few days. The background in an energy range of > 6 MeV was much less than one event per measurement time. Alpha-activities of the samples were measured in the energy range of 2,5-9 MeV so that alpha-particles from all the studied elements and possible impurities could be simultaneously detected. The measurements were performed continuously for several days.

In Fig. 2 the parts of spectrum near the α -particle energies of the ^{149}Tb , ^{246}Cf and ^{282}Fm obtained in a 35-hour-long measurement are shown. For that time no alpha-particles with energies above those of ^{282}Fm were detected.

Results and Discussion

The distribution of the given isotope along the gas duct which is established in a prolonged experiment is resulted from a number of processes. It depends upon the kinetics of the chlorination reaction, mass transfer from the gas flow to the surface of the walls, adsorption and desorption of chloride molecules, the formation and precipitation of aerosol particles, chemical interaction between molecules and the surface of the walls and so on.

The main role probably belongs to the adsorption of molecules on the surface of the conduit walls^[7,10]. It slows down the speed of transportation along the duct of the molecules of the component adsorbed as compared with that of the inert gas. This phenomenon is known to be used in gas chromatography.

It took about 0,2 sec for the gas stream to pass from the target to the chemical filter or to the condensing trap. Under the conditions in the case (Table 1) isotopes of elements forming comparatively volatile chlorides (Sn, Nb, Zr, Hf, kurchatovium) pass this distance for 0,4 sec, i.e. without any significant delay, and are accumulated only in the chemically active filter (6) or in the trap (7) (Fig. 1). (Chemical filtration was based on chemical sorption with sodium chloride). At the same time isotopes of elements forming less volatile chlorides (Na, Sc, lanthanides, Cm, Cf) were by 98-99% adsorbed on the surface of the tube and in the filter^[1,7] in up to 70-hour-long experiments.

The study of the distribution of the chlorides of this group between the tube and the filter indicated that their behaviour is still somewhat different. (c.f. Table 2).

Condensation and adsorption processes occur due to the same molecular forces. Therefore the relationship between the adsorption heats for different compounds is in the first approximation close to that between heats of evaporation or boiling temperatures. This is the reason for the correspondence between adsorption behaviour and volatility of chlorides.

The results obtained in the study of element 102 are given in Table 1. The number of atoms of each isotope found in the tube is taken

as 100, the number of atoms found in the filters is given as a fraction. The standard deviations evaluated from the total number of the detected decay events of the isotope are given. When the values given in Table 1 are based on the detection of a very small or zero number of decays, they are chosen to be the most probable ones. The fiducial limits given in the table are taken from the Poisson distribution^[11,12]. As was noted above, the behaviour of the atoms of ²⁵⁶102 was defined by means of the daughter isotope ²⁵²Fm. If the properties of the chloride of element 102 differ from those of the chlorides of group III, then ²⁵²Fm should be distributed between the parts of the apparatus according to the following inequality:

$$\frac{\text{The amount of } ^{246}\text{Cf}(^{149}\text{Tb}) \text{ atoms in the chemical filter}}{\text{The amount of } ^{246}\text{Cf}(^{149}\text{Tb}) \text{ atoms obtained in the experiment.}} \neq \frac{\text{The amount of } ^{252}\text{Fm} \text{ atoms on the chemical filter.}}{\text{The amount of } ^{252}\text{Fm} \text{ atoms obtained in the experiment.}}$$

As one can see from Table 1 the ratio between the number of atoms of ²⁵²Fm in the chemical and inert filters and the whole number of atoms obtained in the experiment is not higher than that for ²⁴⁶Cf and ¹⁴⁹Tb. If the chloride of element 102 had been significantly volatile, the distribution of ²⁵²Fm should have been displaced in the direction of the gas flow. Since the fraction of ²⁴⁶Cf, ¹⁴⁹Tb in the filter and the subsequent parts of the apparatus is not above several per cent, the sensitivity of detection of this displacement which can be achieved in principle is sufficiently high. One should stress that the behaviour of ²⁴⁶Cf is to a great extent due to the chemical properties of its parent isotope ²⁵⁰Fm (see above).

The above-mentioned results and concepts indicate that the chloride of element 102 undergoes strong adsorption and therefore is not very volatile. It was directly shown that the properties of the chloride of element 102 and of chlorides of fermium, californium and lanthanides are similar. This fact unambiguously indicates that the chemical properties of element 102 are close to those of lighter transplutonic and lanthanide elements.

Conclusions

1) Studies of the chemical properties of element 102 were conducted for the first time.

2) The chloride of element 102 is close to chlorides of Tb, Cf and F_m as far as its volatility (adsorbability on the surface of solids) is concerned.

3) There was a high degree of separation of element 104 from spontaneously fissioning ²⁶⁶102 in the experiments on chemical identification of ²⁶⁰104 / 1/ due to a sharp difference in the volatility of their chlorides.

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TABLE 1

Exp. No	Ion energy MeV		Carrier	Part of the apparatus	T /C°/	Material	Distribution of isotopes along the apparatus*		
	^{16}O	^{22}Ne					^{149}Tb ***	^{246}Cf	^{252}Fm
							$^{250}\text{Fm} + ^{246}\text{Cf}$	$^{256}\text{102} + ^{252}\text{Fm}$	
1	90	110	NbCl ₅ 0.2mm Hg	tube	250-	teflon	100 ± 7	100 ± 18	100 ± 10
				inert filter	300	glass	8.7 ^{+2.1} _{-1.7}	5.4 ^{+7.4} _{-3.9}	1.2 ^{+2.2} _{-1.0}
				chem. filter	300	NaCl	1.1 ^{+0.5} _{-1.4}	1.4 ^{+2.3} _{-1.4} **	0.7 ^{+1.4} _{-0.6}
2	90	110	NbCl ₅ 0.2mm Hg	tube	300-	glass	100 ± 6	100 ± 11	100 ± 15
				inert filter	350	glass	7.5 ^{+2.2} _{-1.1}	+10 16 -6.7	5.5 ^{+8.2} _{-3.9}
				chem. filter	300	NaCl	6.5 ^{+2.2} _{-1.7}	2.5 ^{+4.8} _{-2.1}	2.0 ^{+3.3} _{-2.0} **
3	90	110	NbCl ₅ 0.2mm Hg	tube	300-	glass	100 ± 0.5	100 ± 12	100 ± 22
				inert filter	350	glass	1.3 ± 0.05	0.5 ^{+0.9} _{-0.5} **	1.4 ^{+2.3} _{-1.4} **
				trap	30	condens. NbCl ₅ +ZrCl ₄	1.3 ± 0.1	0.8 ^{+1.3} _{-0.8} **	2.4 ^{+4.0} _{-2.4} **

* The amount of isotopes adsorbed on the surface of the tube in all the experiments is taken 100.

** Not a single pulse was detected.

*** The distribution of ^{148}Gd is not given separately.

TABLE 2

Chloride	T boiling (C°)	T exp. (C°)	Notes
SnCl ₄	113	200	Not adsorbed in the tube and inert filter
HfCl ₄	317 subl.	250-350	" - " -
ZrCl ₄	331 subl.	250-350	" - " -
InCl ₃	498 subl.	250	Adsorbed in the inert filter
		350	Not adsorbed in the inert filter
ScCl ₃	1077	250	Adsorbed in the tube and in the inert filter
		350	>20% passes through the inert filter
LnCl ₃	1700	250-350	>90% is adsorbed in the tube
AmCl ₃	1752	250-350	>90% of curium is adsorbed in the tube.

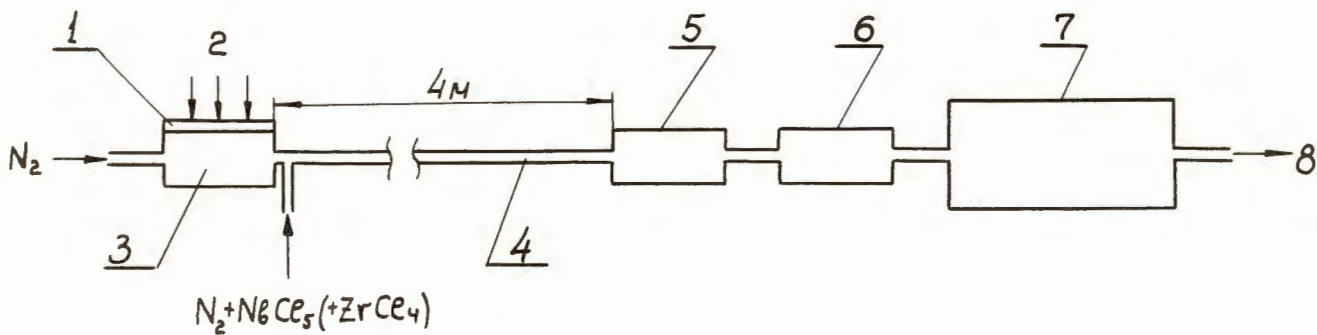


Fig. 1. A block-diagram of the apparatus for chlorination and fast separation of nuclear reaction products, 1, Target, 2, Beam of accelerated particles, 3, Gas volume behind the target, 4, Tube 5, Inert filter, 6, Chemical filter, 7, Condensate trap, 8, Outlet of the gas.

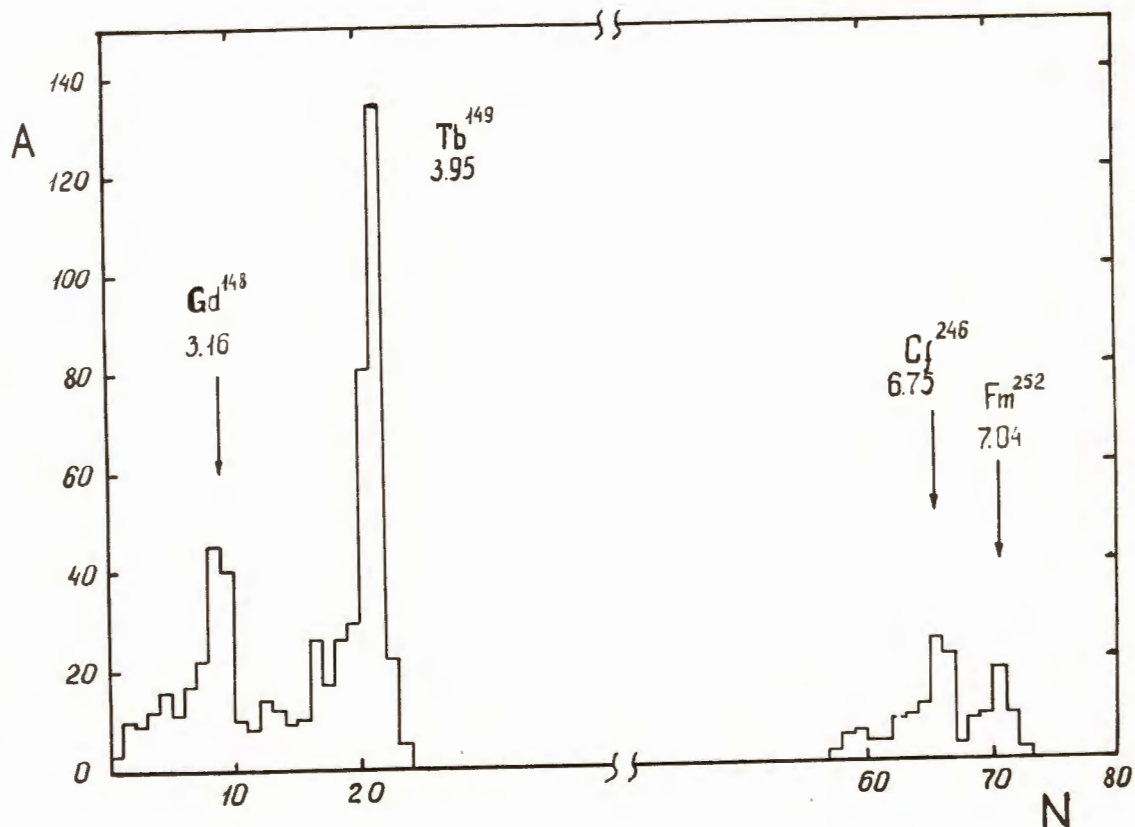


Fig. 2. An example of alpha-spectrum of terbium and heavy transplutonic elements fraction, A-number of alpha-particles detected in for 35-hour measurement, N-channel number. The values of alpha-particle energy (MeV) are given.