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**MEASUREMENT OF SOLUBILITY OF PLUTONIUM TRIFLUORIDE
AND RARE-EARTH FLUORIDES IN MOLTEN $\text{LiF} - \text{BeF}_2 - \text{ZrF}_4$**

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INTRODUCTION

Data on behaviour of plutonium fluoride and fission products (FP) dissolved in fuel composition are needed to calculate the duration of an operating cycle of the ADTT facility (Accelerator-Driver Transmutation Technologies) and to determine the effect of their equilibrium concentrations on nuclear-physical characteristics of reactor operation [1-2].

The data on the FP fluoride solubility in the molten salts are of great important for some industrial processes (electrolytical metal deposition, development of physical-chemical means for processes of chemical technology, etc.) [3-8].

As noted above [9], some information on this question is given in monography [2-4] and articles [5-8]. Data concerning fluoride salts are given in reports [10-11]. However, it was impossible to make the substantial analysis of mutual solubility of fluoride melts.

The primary investigation of CeF_3 and neodymium, samarium and lanthanum fluorides showed that the solubility of the melt $LiF-BeF_2$ and $LiF-BeF_2-ThF_4$ was a linear function of reverse temperature [11] and increases from lanthanum to samarium in the row of rare-earth elements. Of the fluoride systems investigated the more reliable data are available on CeF_3 only, but data on LaF_3 solubility are restricted to the temperature range of 500-600°C [11].

There are no literature data concerning the solubility of the salts UF_4 and ThF_4 in the double and triple compositions based on $LiF-BeF_2$. At the same time there is information on the PuF_3 solubility in the fluoride compositions studied by methods of physical-chemical and radiometric analysis [10, 13-15]. Based on data of PuF_3 solubility in the compositions $LiF-BeF_2$ and $LiF-ThF_4$ it was possible to calculate the PuF_3 solubility in the different molten salts as a function of salt composition.

Comparison between the solubility of PuF_3 in $LiF-BeF_2-ThF_4(75-20-5 \text{ mole}\%)$ and $LiF-ThF_4$ melts at temperatures 600 and 700°C showed that increase in temperature resulted in its increasing from 2.88 and 3.68 mole% to 4.75 and 5.92 mole%, respectively [13-14]. However, in one of the primary Barton's work [10] the smaller value of PuF_3 solubility was obtained in $LiF-BeF_2$ melt. Depending on the beryllium fluoride content in melt, it was 1.33 mole% (28.7 mole% BeF_2) and 0.65 mole% (37.0 mole% BeF_2) at 650°Ct.

Disagreement in estimation of plutonium trifluoride solubility and incomplete data on the solubility of rare-earth elements resulted in necessity of the additional data for these compositions. Therefore, the object of the present investigation was to study the solubility of some FP fluorides (La, Ce, Nd) and plutonium trifluoride in the solvent-melt based on the composition 0.65 $LiF-0.29BeF_2-$

0.05ZrF_4 proposed as a fuel for the ADTT facility. Based on the common approach we have studied the solubility of LaF_3 , CeF_3 , CeF_4 , NdF_3 and PuF_3 in the $\text{LiF}-\text{BeF}_2-\text{ZrF}_4$ melt within the temperature range of $500-700^\circ\text{C}$.

1. SALT PREPARATION AND CHEMICAL REAGENT DECONTAMINATION

1.1. Preparation of high-purity lithium fluoride

At first the traces of moisture were removed from lithium fluoride of the pure grade. For this aim the glass-carbon crucible equipped with lithium fluoride sample was placed into the test tube closed at the top by a rubber plug provided with thermal reflecting nickel screens and located in the shaft furnace. The cell was evacuated and kept at residual pressure 10^{-2} mm Hg during 4-6 hrs. In this case the temperature of furnace was slowly elevated from room one to 700°C . Upon attaining the specified temperature, an inert gas was entered the gas space of the test tube, while the temperature of furnace was elevated to 900°C . Following salt melting, the cell was taken from the furnace and cooled at the air.

The lithium fluoride of special pure grade was prepared by zone melting method [12]. The boat made of nickel and preliminarily loaded by remelted salt was placed into the quartz tube which was then evacuated and filled by an inert gas. The zone melting was carried out in the shaft furnace moved along the quartz tube and inclined at an angle of 10° to horizon to prevent the molten salt overflow from the boat edge after several runs of furnace. The molten zone was $10\text{--}20$ mm wide and the boat was 20 mm wide and 200-300 mm long.

The zone melting was carried out in the current of argon purified from the traces of oxygen and moisture. The optimum rate of furnace movement along the quartz tube was $2.5 \cdot 10^{-2}$ mm/s. At high rate the central part of the quartz tube was strongly elongated in the direction opposite to its movement that adversely affected the extent of purification. A nickel foil screen was placed above the boat to prevent damage of the quartz tube after its interaction with the salt vapour.

The application of zone melting method allowed the production of colourless crystals of fluoride lithium practically free from oxides, hydroxides of alkaline metals and insoluble impurities. It was sufficient 2-3 cycles of zone melting for the good purification of salt.

1.2. Preparation of high-purity beryllium difluoride

Since salt mixtures based on lithium fluoride were used in investigation, the preliminarily prepared composition Li_2BeF_4 contained beryllium difluoride were purified by zone melting method. To obtain the salt mixture of suitable composition, its correction was made by the formerly obtained lithium fluoride of high purity.

The glass-carbon crucible provided with the calculated samples LiF and BeF_2 was placed in the cell equipped by the nickel screens. The test tube was closed by the rubber plug with the nickel screens, placed in the shaft furnace, evacuated and slowly heated up to 400°C during 4-6 hrs. Then the gas space of the test tube was filled by inert gas, whereas the furnace temperature was elevated up to 900°C . The obtained composition Li_2BeF_4 was identified by X-ray and chemical methods.

As above show then the composition Li_2BeF_4 was purified by zone melting method.

The use of this method allowed the production of Li_2BeF_4 crystals practically free from oxides and hydroxides of alkali metals and insoluble impurities. It was sufficient 3-4 boat runs for good purification of salt.

1.3. Preparation of high-purity zirconium tetrafluoride.

Zirconium tetrafluoride purified from traces of oxygen, moisture and impurity was prepared by sublimation method in the current of argon in the horizontal type sublimator. It was constructed as follows. The quartz tube of 50 mm diameter and 1200 mm long was inserted in the shaft furnace involved about 1/3 part of the sublimator where the boat made of inert material (glass-carbon, nickel) and filled with zirconium tetrafluoride was located. Then the tube was closed by the rubber plugs, evacuated and filled with argon. Zirconium tetrafluoride was sublimated in the current of purified argon at temperature $650-700^\circ\text{C}$. The purified product was condensed in the cold part of the quartz tube and then it was identified by chemical and X-ray methods.

1.4. Synthesis of neodymium and lanthanum trifluorides and cerium tetrafluoride.

Neodymium and cerium trifluorides were tested with using available anhydrous salts specified as "chemically pure", X-ray analysis of which proved availability of reagents NdF_3 and CeF_3 only.

The reagents LaF_3 and CeF_4 were prepared using neodymium, lanthanum and cerium oxides and hydrofluoric acid. Lanthanum and cerium oxides placed in the teflon cup were filled with the excess of hydrofluoric acid and boiled during 6-8 hours. Then the precipitates were filtered, washed by distilled water and dried. The obtained precipitate was identified by X-ray method as LaF_3 and $\text{CeF}_4 \cdot 0.5\text{H}_2\text{O}$. To remove crystallized water the salt was treated by the mixture of helium and hydrogen during 12 hours at 300°C .

All the reagents were kept in polyethylene pots placed in the dry box under the atmosphere of argon. Before experiment the above reagents were evacuated at 700°C during 6-8 hours.

1.5. Synthesis of plutonium trifluoride

To produce PuF_3 the starting Pu(IV) was reduced to Pu(III) by heating with ascorbine acid.

The extent of plutonium reduction was measured by spectrophotometric method against absorption bands at wave length of 470 nm and 600 nm for Pu(IV) and Pu(III) , respectively.

After cooling the mixture contained HF and HNO_3 in ratio $\text{HNO}_3/\text{HF}=6$ was added in the solution by agitating. The precipitate was sublimated, filtered, washed by diluted solutions HF and HNO_3 and dried by acetone. Plutonium trifluoride was dehydrated in the current of He + 4.98% H_2 saturated by NH_4F vapours by double successive thermal treatment at 300°C during 20 minutes. To complete dehydration once more analogous treatment was made at 400°C during 20-25 min.

The phase content of product was measured by X-ray method and presence of water was determined by infrared spectroscopy. Absence of crystallographic water was found by analysis.

2. INTERACTION OF NEODYMIUM, LANTHANUM AND CERIUM FLUORIDES WITH $\text{LiF-BE}_2\text{-ZrF}_4$ MELT

First stage of work was in preparation of the molten salt of the specified composition and the pellets of neodymium, lanthanum and cerium fluorides. The pellets were prepared at the standard press and no bonding materials were used.

To prepare the solvent composed of 0.65 LiF - 0.29 BeF_2 - 0.05 ZrF_4 the calculated samples of the previously prepared reagents LiF , Li_2BeF_4 and ZrF_4 were loaded in the glass-carbon crucible. Then the crucible was placed in the cell, evacuated and heated to melting temperature of salt composition in the atmosphere of argon. After melting of reagents, the test tube was cooled but the

prepared melt of lithium, beryllium and zirconium fluorides was used directly in experiments to determine the solubility of NdF_3 , LaF_3 and CeF_4 .

Experiments were carried out in cells made of transparent quartz glass in the atmosphere of argon purified from oxygen-contained impurities. The cell was equipped by a lock mechanism allowed to take samples without tightness failure. It contained also a light device provided to observe the behavior of liquid and solid phases in the process of sampling. The experimental cell is shown in Fig.1.

The solubility of NdF_3 , LaF_3 and CeF_4 in the molten salts of the specified composition was determined by isothermal saturation method. For this purpose the pellet prepared from neodymium or lanthanum fluoride (or other RE fluoride) weighing 5-15 g and the melt composed of 0.65 LiF - 0.29 BeF_2 - 0.05 ZrF_4 weighing 50-80 g were placed in the glass-carbon crucible. Then the crucible was placed in the quartz test tube, held at residual pressure 10^{-2} mm Hg during 4-6 hours and heated up to the given temperature in the atmosphere of argon. To remove the possible oxygen traces before melting of salts a portion of powder composed of 45-50% Ce, 22-25% La, 15-17% Nd melts and 8-10% of other RE elements were placed on the bottom of quartz cell.

A few tens milligrammes of melt were periodically sampled and placed in the quartz tubes. In this case the contact time of the quartz glass with the molten salt did not exceed 15-20 sec. The mass content of the investigated component was determined by chemical analysis using spectrophotometer with induced plasma.

The investigation results of interaction kinetics of solid phase with the molten salt showed that the attained time of equilibrium between fission product fluorides and LiF - BeF_2 - ZrF_4 molten composition depended on the nature of material and it does not exceed up to 20 hours at 550°C. It has been found that increasing test temperature resulted in increasing the rate of phase interactions and decreasing the time sufficient to establish the equilibrium under the given conditions. Optical observations on behaviour of FP fluoride pellets showed their damage in fine fragments in the course of experiments. It is unlikely that particles of solid phase will be captured, because the samples were taken under visual inspection of melt state. Results of chemical analysis of double samples taken under the same conditions showed good reproducibility of data especially at temperatures 625 and 700°C and confirmed our assumption about absence of undissolved fluoride particles of fission products therein.

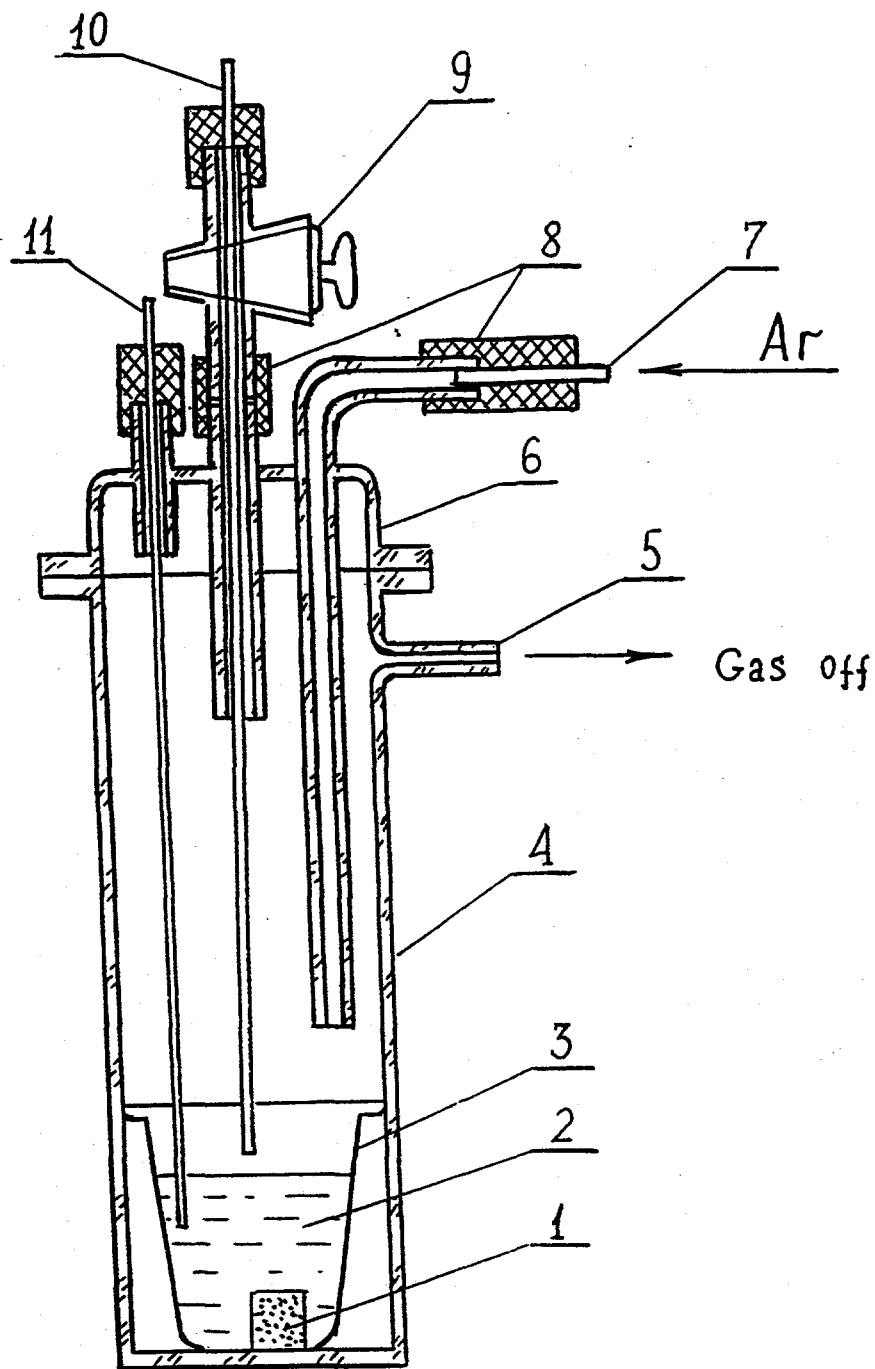


Fig.1. Apparatus to study solubility of rare-earth and plutonium fluorides in the $\text{LiF-BeF}_2\text{-ZrF}_4$ melt

- 1 - pellet; 2 - molten salt; 3 - crucible (glass-carbon);
- 4 - quartz cell; 5 - gas release (through hydrolock);
- 6 - quartz cap; 7 - inert gas feed; 8 - coupling; 9 - crane;
- 10 - quartz tube for sampling; 11 - thermocouple.

3. INVESTIGATED RESULTS OF RE FLUORIDE SOLUBILITY

Averaged data on the solubility of NdF_3 , LaF_3 and CeF_4 in $0.65 \text{ LiF} - 0.29 \text{ BeF}_2 - 0.05 \text{ ZrF}_4$ obtained at 550, 625 and 700°C in the atmosphere of argon are given in Tab.1.

Experimental data on the solubility of NdF_3 , LaF_3 and CeF_4 in $0.65 \text{ LiF} - 0.29 \text{ BeF}_2 - 0.05 \text{ ZrF}_4$ molten salt presented in coordinates $\log S - 1/T$ are given in Fig. 2.

Table 1.

Solubility of neodymium and lanthanum trifluorides and cerium tetrafluoride depending on temperature in $\text{LiF}-\text{BeF}_2-\text{ZrF}_4$ melt in the atmosphere of argon.

Temperature, °C	Solubility, mole %			
	NdF_3	LaF_3	CeF_4	CeF_3
550	0.68 ± 0.07	0.52 ± 0.05	0.62 ± 0.06	1.01 ± 0.10
625	1.62 ± 0.16	1.60 ± 0.16	1.1 ± 0.10	1.98 ± 0.18
700	3.75 ± 0.32	2.72 ± 0.25	2.5 ± 0.22	3.04 ± 0.28

It was found that in the given coordinates they were approximated with the straight lines and may be expressed by equations:

$$\log S = 2.05 - 3984/T \pm 0.06; \quad (\text{for } \text{LaF}_3)$$

$$\log S = 0.46 - 2605/T \pm 0.04; \quad (\text{for } \text{CeF}_3)$$

$$\log S = 1.36 - 3406/T \pm 0.03; \quad (\text{for } \text{CeF}_4)$$

$$\log S = 5.48 - 7042/T \pm 0.02; \quad (\text{for } \text{NdF}_3)$$

Comparison of our results with the primary data on the solubility of CeF_3 in the different salt compositions are given in Fig.3.

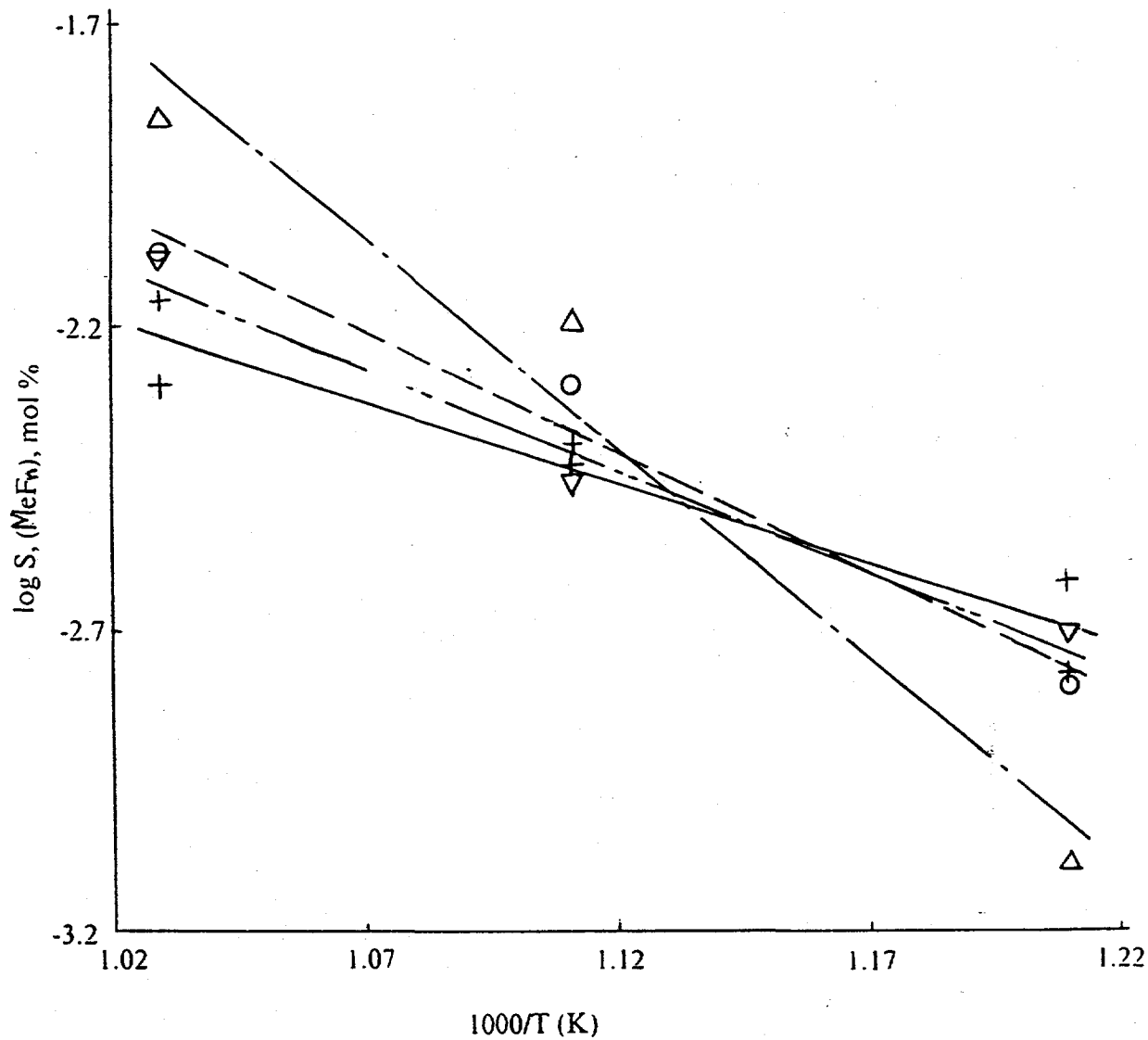


Fig.2. Solubility of rare-earth fluorides as a function of temperature in the $\text{LiF}-\text{BeF}_2-\text{ZrF}_4$ molten salt

- o - solubility of lanthanum trifluoride
- + - solubility of cerium trifluoride
- ∇ - solubility of cerium tetrafluoride
- Δ - solubility of neodymium trifluoride

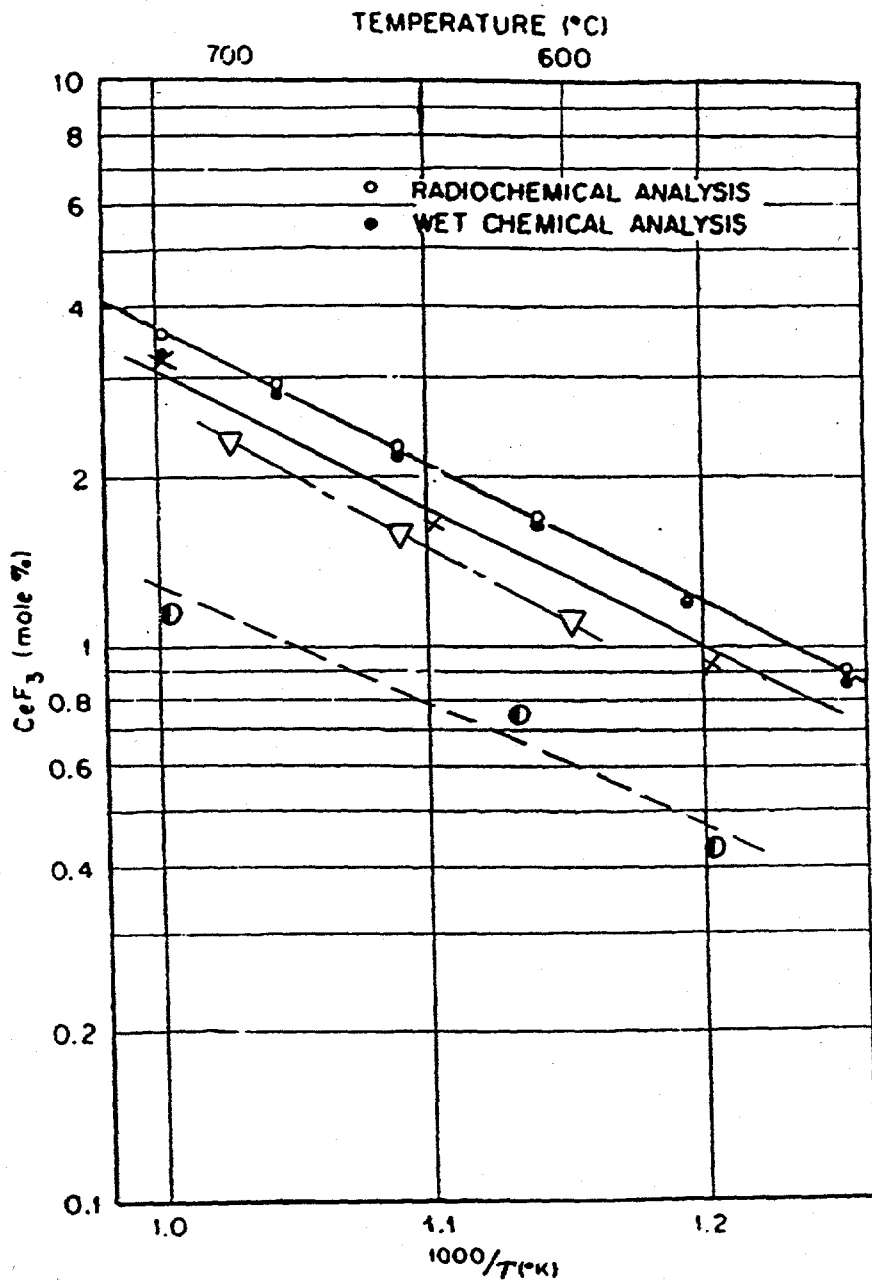


Fig.3. Solubility of cerium trifluoride as a function of temperature and content of heavy metal fluoride

- - solubility of CeF₃ in the melt of 0.71 LiF-0.29 BeF₂ [11];
- ▽ - solubility of CeF₃ in the melt of 0.68 LiF-0.25 BeF₂-0.07-ThF₄ [11];
- , ○ - solubility of CeF₃ in the melt of 0.72 LiF-0.16 BeF₂-0.12-ThF₄ [11];
- × - solubility of CeF₃ in the melt of 0.65 LiF-0.29 BeF₂-0.05 ZrF₄ (our data).

The obtained results allow to note that quantity of heavy metal tetrafluoride (zirconium, thorium, etc.) introduced in melt determines the solubility extent of RE metal fluorides.

In performing experiments the following features were noted. Although the necessary steps related to the purity of prepared salts were taken in the course of experiment the new films of unknown nature were observed on the surface of the solvent-melt. Upon cooling the film and sediment of grey colour were observed on the surface and in the bottom part of melt. The cut of melt took the following appearance: the grey film about 1 mm thick and about 2 mm thick in the bottom layer and contour of dissolved fluoride pellet were observed in the melt volume. The cuts of pellets were grey while the initial salts were white. The other volume of molten salt was white. To study nature of film and grey precipitates as well as colour transformations of pellets the corresponding samples were analyzed. X-ray fluorescence spectrum and data of the grey film are presented in Fig.4 and table 2.

According to X-ray fluorescence and phase analysis there are no carbide metal forms in specimens and oxide content is about 0.1%.

4. INVESTIGATION OF THE PLUTONIUM TRIFLUORIDE SOLUBILITY IN LiF-BeF₂-ZrF₄ MELT

The solubility of plutonium trifluoride was investigated by methods similar those applied above in section 2.

The solubility of PuF₃ was determined by method of isothermal saturation in melts contained different quantity of zirconium tetrafluoride (5 and 7 mole %) at temperature 550, 625 and 700°C.

Two pellets of mass 3.05 and 2.82 g were introduced into the melt 0.65LiF-0.30BeF₂-0.05ZrF₄ and held during 20 hours at temperature 550°C. Following sampling the temperature was elevated up to 625°C and the melt was held at this temperature during four hours and samples were taken again. The same way of measuring was applied at 700°C.

To study the plutonium trifluoride solubility about 1.89 g of PuF₃ was added in the melt 0.64LiF-0.29BeF₂-0.07ZrF₄.

Since PuF₃ is poorly dissolved in water and water solutions of acids the sample was dissolved in 6M HNO₃ with added H₃BO₃ and boiled during 5-6 hours with reverse cooling. The residue of solution was dried and dissolved in the specified volume of water.

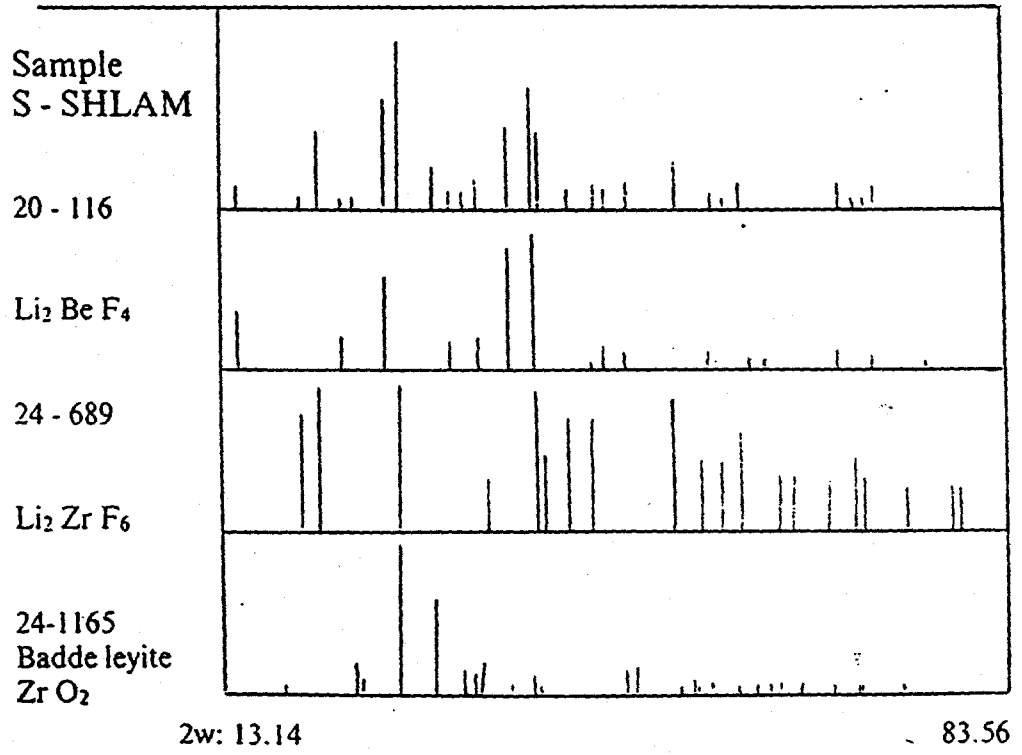
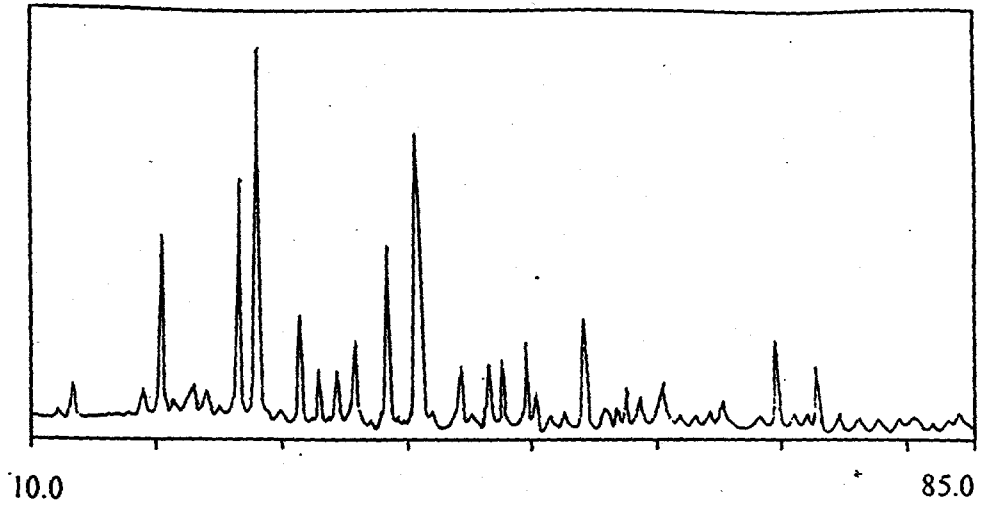


Fig.4. Spectra of X-ray fluorescent analysis

Comparison Table

Sample	20 - 116 95	24 - 689 90	24 - 1165 93
	Li ₂ Be F ₄	Li ₂ Zr O ₂	Baddeleyite Zr O ₂
6.740 6*	-	-	-
6.635 10	6.623 38	-	-
4.660 8	-	4.630 80	-
4.310 44	-	4.310 100	-
4.149 5	4.149 9	-	-
3.906 8	3.906 23	-	-
3.836 9	3.846 20	-	-
3.695 7	-	-	3.690 18
3.632 5	-	-	3.636 12
3.325 62	3.322 56	-	-
3.208 9*	-	-	-
3.163 100	-	3.165 100	3.165 100
2.838 27	-	-	2.841 66
2.710 13	2.710 31	-	-
2.613 12	-	-	2.618 20
2.538 6	-	-	2.538 15
2.509 20	2.513 23	-	2.500 3
2.489 5*	-	-	-
2.346 45	2.347 100	-	2.331 5
2.218 74	2.217 95	-	2.212 12
2.192 44	2.188 3	2.193 100	2.188 5
2.154 8	-	2.155 50	-
2.047 16	2.045 2	2.049 80	-
2.017 4	-	-	2.020 6
1.990 3	-	-	1.988 6
1.955 16	1.953 2	1.953 80	-
1.916 15	1.919 15	-	-
1.842 22	1.842 8	-	1.848 14
1.818 11	1.828 1	-	1.818 18
1.802 5	-	-	1.802 11
1.782 3	1.776 2	-	1.783 5
1.740 5	1.742 2	-	-

1.698 28	1.704 2	1.701 90	1.692 10
1.657 6	-	-	1.656 10
1.644 4	1.650 2	-	1.650 8
1.629 6	-	1.629 50	-
1.610 10	1.610 15	-	1.610 7
1.580 8	-	1.580 50	1.580 3
1.555 3	1.555 4	-	-
1.545 5	-	-	1.546 7
1.538 14	-	1.538 70	1.538 7
1.524 4	1.524 5	-	-
1.509 3	1.515 2	-	1.508 4
1.495 3	-	-	1.495 5
1.484 3	1.486 7	-	-
1.477 5	-	-	1.477 8
1.458 5	1.453 2	1.462 40	-
1.436 7	-	1.437 50	-
1.420 4	-	-	1.427 2
1.356 20	1.357 14	-	1.361 2
1.334 5	-	1.333 50	-
1.318 4	-	1.318 40	1.321 4
1.303 14	1.304 6	-	-
1.279 3	1.279 4	-	-
1.258 3	-	1.259 30	1.264 3
1.213 3	1.214 3	-	-
1.201 3	-	1.202 30	-
1.157 5*	-	-	-
	+ 3.521 2	+ 2.451 40	+ 5.076 7
	+ 3.106 4	+ 1.374 30	+ 2.604 14
	+ 3.012 1	+ 1.195 30	+ 2.179 5
	+ 2.740 2		+ 1.959 2
	+ 2.273 1		+ 1.642 7
	+ 1.721 1		+ 1.490 4
	+ 1.570 2		+ 1.447 3
	+ 1.504 1		+ 1.418 5
	+ 1.350 2		+ 1.414 4
	+ 1.233 4		+ 1.325 2

The experimental data on the solubility of plutonium trifluoride in the above melts are given in Tab.3 and Fig.5. This relation may be described by the following equation:

$$\log S = 0,700 - 1380/T \pm 0,015 \quad \text{ZrF}_4 \text{ content in 5 mole \% melt}$$

$$\log S = 1,056 - 1640/T \pm 0,015 \quad \text{ZrF}_4 \text{ content in 7 mole \% melt}$$

In Fig. 5 there is comparison of our results with the primary ones [10].

Table 3

Solubility of plutonium trifluoride in LiF-BeF₂-ZrF₄ melt against ZrF₄ content and temperature

Temperature, °C	Solubility PuF ₃ , mole %	
	ZrF ₄ content 5 mole %	ZrF ₄ content 7 mole %
550	0.103±0.015	0.116±0.015
625	0.151±0.018	0.169±0.017
700	0.188±0.021	0.235±0.019

The values of the plutonium trifluoride solubility obtained in our experiments are considerably different from those presented in [10,13,14]. Such difference results from the experimental technique. For example, plutonium trifluoride in the form of powder was loaded into the nickel container having the porous bottom of 200 mkm pore in size. The bottom part of the container was loaded in the nickel crucible contained molten salt. After leakage and interaction with PuF₃ the melt was able to leave the container due to diffusive and convective flows and to transfer the finest solid particles of plutonium trifluoride from the container to the crucible. It was able to cause the more high values of solubility in the above works.

Conclusions

The investigation results of the RE fluoride solubility showed that temperature increasing resulted in increase of solubility.

The solubility of rare-earth fluorides was stated to be 0.52-3.75 mole % within 550-700°C and to be described by a straight line in the coordinates $\log S - 1/T$. They may be presented as equation:

$$\log S = 2.05 - 3984/T \pm 0.06 \text{ (LaF}_3\text{)}$$

$$\log S = 0.46 - 2605/T \pm 0.04 \text{ (CeF}_3\text{)}$$

$$\log S = 1.36 - 3406/T \pm 0.03 \text{ (CeF}_4\text{)}$$

$$\log S = 5.48 - 7042/T \pm 0.02 \text{ (NdF}_3\text{)}$$

The solubility of plutonium trifluoride and rare-earth elements in the $\text{LiF-BeF}_2\text{-ZrF}_4$ melt increases with increase in temperature and comes up to 0.11-0.24 mole%. This relation may be described by the following equation:

$$\log S = 0,700 - 1380/T \pm 0,015 \quad \text{ZrF}_4 \text{ content in 5 mole \% melt}$$

$$\log S = 1,056 - 1640/T \pm 0,015 \quad \text{ZrF}_4 \text{ content in 7 mole \% melt}$$

With increasing the ZrF_4 concentration in the molten salt the PuF_3 solubility increases. For example, with increasing the content of zirconium tetrafluoride in the melt from 5 mole % to 7 mole % the solubility of plutonium trifluoride increased from 1.9 mole % to 2.4 mole %.

The values of plutonium solubility found in our experiments are lower than those obtained in literature.

Thus, the experiments performed by the single technique found both coincidence of plutonium trifluoride solubility data with literature ones and their significant difference. Therefore, it is necessary to make further comparative investigations to obtain the proper initial data on conceptual developments of the ADTT facility.

The further systematical investigations are required to study the PuF_3 , PuF_4 , NpF_3 , AmF_3 and Cm_3 solubility in the $\text{LiF-BeF}_2\text{-ZrF}_4$ molten salt against composition of molten salt (content in ZrF_4 melt), the type of dissolved fluoride (dispersive or compact form) and kinetics of such processes. The systematical investigation of rare-earth fluorides is also required.

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