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Key Experimental Results of the PYROSMANI Project

Victor V. Ignatiev[a]*, Mikhail V. Kormilitsyn[a], Andrey A. Lizin[b], Alexander V. Zagnitko[a], Sergey A. Konakov[a], Alexander V. Merzlyakov[a], Sergey V. Tomilin[b], Alexander A. Khokhryakov[b], Alexander G. Osipenko[b]

[a]*National Recearch Center "Kurchatov Institute", 1, Kurchatov sq., Moscow, 123182, Russia*
[b]*Joint Stock Company "State Scientific Center – Research Institute of Atomic Reactors", 9, West str., Dimitrovgrad, Ulyanovsk region, 433510, Russia,*

Abstract

The presented key results were obtained in the course of experiments carried out within the PYROSMANI (PYROchemical processes Study for Minor ActiNIdes recycling in molten salt chlorides and fluorides) Project supported by Rosatom. The individual and joint solubility of UF_4, PuF_3 and CeF_3 in ternary LiF–NaF–KF and LiF- ThF_4-UF_4 melts was measured by method of isothermal saturation for the temperature range 550-800°C. The solubility measurement technique based on isothermal saturation was verified in LiF–NaF–KF eutectics for praseodymium trifluoride by the reflectance spectroscopy. The process under investigation was the extraction of lanthanum, neodymium, europium and samarium trifluorides from 73LiF-27BeF_2 melt (mole %) into liquid bismuth at 600-610°C. There were defined temperature dependences of kinematic viscosity and melting temperatures for molten 46.5LiF–11.5NaF–42KF; 73LiF-27BeF_2; 85LiF-15AlF_3 and 43.5LiF–24.3NaF–32.2UF_4 salt mixtures from liquidus temperature to 840°C.

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Keywords: pyrochemical reprocessing, molten salt reactor, actinides and lanthanides fluorides, melting temperature, solubility, kinematic viscosity, reductive extraction, isothermal saturation, reflectance spectroscopy.

* Corresponding author
e-mail address: Ignatev_VV@nrcki.ru

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1. Introduction

Pyrochemical methods have been investigated in Russia for many years as applied to the used fuel reprocessing and molten salt reactors with the use the molten salts as a medium [1] due to following properties of molten salt chlorides and fluorides:

- irradiation resistance allowing the reprocessing of high burnup fuels (> 100GW day/tHM);
- thermal resistance allowing the reprocessing of used fuels with a short cooling times (< 3yrs);
- possibility to have a high concentration of transuranic elements in the melts from the viewpoint of their solubility and nuclear safety;
- separation factors for actinides and rare-earths that allows manage the process with minimal number of stages.

In 2013-2015, in the frame of the ROSATOM-EURATOM cooperation and under coordination with the European Project SACSESS (Safety of Pyrochemical Processes Domain of Safety of ACtinide Separation processes), molten salts properties were studied in frame of the Russian Project PYROSMANI (PYROchemical processes Study for Minor ActiNIdes recycling in molten salt chlorides and fluorides) with goals of the processes and systems justification

2. Joint solubility for actinides and lanthanides fluorides in ternary melts

2.1. LiF–ThF_4–UF_4 melt

The joint solubility of PuF_3 and CeF_3 in 78LiF–7ThF_4–15UF_4 and 72.5LiF–7ThF_4–20.5UF_4 melts (mole %) was studied in the temperature range 550–800°C. Non-aqueous PuF_3 [2] and CeF_3 [2] were synthesized as well as radiolabeled ^{144}Ce and non-aqueous ThF_4 [3]. Figure 1 presents a layout of test section. The isothermal saturation method was used to dissolve pressed pellets PuF_3 and CeF_3 in the solvent system under study with inert atmosphere (argon). The selected compositions of fluoride melts meet specific requirements set to the molten salt fast reactor fuel and can be used as reprocessing medium [4, 5, 6]. Figure 2 presents a phase diagram for a ternary 75LiF–7ThF_4–*x*UF_4 melt [7]. The fraction of heavy metal tetrafluorides in melts under study makes up 0.22 and 0.275, respectively. As it can be seen from Tables 1 and 2, an increase in the molar fraction of uranium tetrafluoride, the one remaining the same for ThF_4, in the ternary melt LiF–ThF_4–UF_4, leads to a decrease in the PuF_3 and CeF_3 solubility. At a temperature of about 600°C, CeF_3 displaces significantly PuF_3 from melts 78LiF-7ThF_4-15UF_4 and 72.5LiF-7ThF_4-20.5UF_4.

Table 1. Weight content of plutonium and cerium and the solubility of the corresponding trifluoride in the 78LiF-7ThF_4-15UF_4 melt [6]

Temperature, ºC	Pu content, wt %	PuF_3 solubility, mole %	Ce content, wt %	CeF_3 solubility, mole %
600	3.66	1.48±0.07	6.6	4.6±0.2
650	12.8	5.8±0.3	8.2	6.3±0.3
700	20.0	9.9±0.5	10.1	8.5±0.4
750	21.6	10.9±0.6	10.3	8.9±0.4

Table 2. Weight content of plutonium and cerium and the solubility of the corresponding trifluoride in the 72.5LiF-7ThF_4-20.5UF_4 melt [6]

Temperature, ºC	Pu content, wt %	PuF_3 solubility, mole %	Ce content, wt %	CeF_3 solubility, mole %
600	0.8	0.36±0.02	3.4	2.6±0.1
650	9.3	4.6±0.2	5.2	4.4±0.2
700	16.1	8.6±0.4	7.1	6.4±0.2
750	17.7	9.6±0.5	7.4	6.8±0.3