Institute of High Temperature Electrochemistry (IHTE), Ekaterinburg, Russia. Current Status of Pyrochemical Research in IHTE

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1. Institute of High Temperature Electrochemistry

The Institute of High Temperature Electrochemistry of the Ural Branch of the Russian Academy of Sciences (IHTE UB RAS) was founded in January 1958 on the base of the Laboratory of Molten Salts Electrochemistry, a worthy part of the Ural electrochemistry research school tradition. Professor Smirnov M.V., Doctor of Science (Chemistry), honored Master of Science and Technology of RSFSR, was the founder and the first head of the Institute. In the following years the Institute was led by such distinguished people as Corresponding Member of the USSR Academy of Sciences Karpachev S.V., Academician Baraboshkin A.N, professor and Doctor of Science (Chemistry) Khokhlov V.A. Since 2006, the Institute was headed by Honored Scientist of the Russian Federation, Doctor of Chemical Sciences, Professor Yu.P. Zaykov. From 2017 to the present time the Director of the Institute is Doctor of Chemical Sciences M.V. Ananyev. Nowadays IHTE is the only institution in Russia that specializes in the field of high temperature physical chemistry and electrochemistry of molten salts and solid state electrolytes. During 60 years IHTE has been engaged in fundamental studies focused on creation, development and application of the following:

– theoretical and experimental foundations of modern physical chemistry and electrochemistry of molten salts and solid state electrolytes;

– principles of electrochemical processes for the production and physical-chemical analysis of new materials for various applications in corrosive environment and high temperatures;

– scientific fundamentals of resource saving, human and environment friendly technologies for electrochemical processes used for the production, refining, and protection of metals and processing inorganic raw materials;

– principles for construction of high temperature molten and solid electrolyte devices which provide the most rational direct conversion of molecular energy into electric power.

For several decades Institute performs research work in the field of physical chemistry and electrochemistry of molten salts for nuclear application.

Properties such as electrical conductivity, density, viscosity as far as structure of melts are studied. Halides of actinides, rare earth metals and other their compounds has been researched. Electrochemical processes and their mechanisms for liquid and solid electrodes has been investigated and modeled, interaction and behavior of different constructive materials in molten salt media were subjects of work aiming to development of fundamentals for the novel pyrochemical technologies of spent nuclear fuel recycling.

2. Progress in study of pyrochemical processing of spent nuclear fuel (SNF) in IHTE

2.1 "Soft chlorination" of nitride SNF

As a first step in the processing of nitride SNF it is proposed to dissolve it in the molten LiCl-KCl eutectic using $CdCl_2$ as a chlorinating agent:

$$UN + 1.5CdCl_2 = UCl_3 + 1.5Cd + 0.5N_2\uparrow$$
$$\Delta G = -58.7 \text{ kJ at } 773 \text{ K} \qquad (1)$$

The results of a typical experiment are shown in Figure 1.

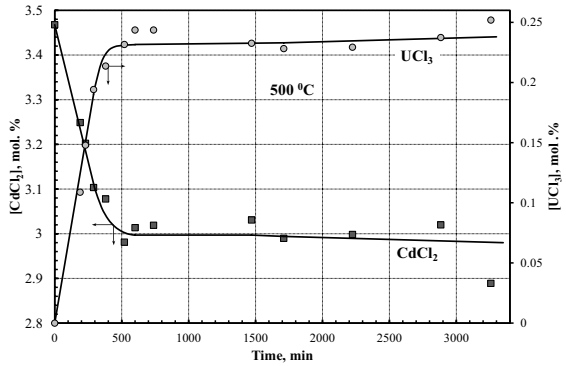

Fig. 1. The $CdCl_2$ and UCl_3 concentration time dependences according to chemical analysis. Initial molar ratio $CdCl_2$ / UN = 4.42.

Based on experimental results and thermodynamic modeling, it was found:

- at 500°C, only ~30% of UN is converted to UCl_3. The remaining uranium passes into a black precipitate consisting of a mixture of UNCl, $UN_{1.50}$, $UN_{1.51}$, $UN_{1.55}$, $UN_{1.59}$, $UN_{1.69}$, $UN_{1.73}$, UN_2.
- at a temperature of 750°C and above, a 100% conversion of UN → UCl_3 is achieved;
- the reaction of UN + $CdCl_2$ proceeds by several parallel reactions and, at least, in two stages.

2.2 Electrolytic reduction of UO_2, rare earthes oxides and their mixtures

The electrolytic reduction of the UO_2 tablets of various porosities in the LiCl + (0.5-2.0)wt.% Li_2O melt at 650°C was studied. The process was monitored by measuring the cathodic potential, with a brief disconnection of the electrolysis current. A complete reduction of UO_2 was observed after passing 165-180% of electricity from the theoretically required amount. Various materials were used as the anode: platinum, graphite, oxide ceramics. The best results were obtained using ceramic non-consumable anodes.

The metallization of the model nuclear fuel UO_2 + La_2O_3, CeO_2, Nd_2O_3 (5-17wt.% in total) was studied. It is found that UO_2 is also completely reduced to metallic uranium, while rare earth metal oxides remain unchanged. Only CeO_2 is reduced to Ce_2O_3.

Individual rare-earth oxides also did not reduced in wide ranges of Li_2O concentrations and cathodic potential.

These results give us a probable way of dividing the fission products. In the electrolytic reduction, alkali and alkali-earth metals will dissolve in the melt, uranium and plutonium will be reduced to the metals, and the lanthanides will remain in the oxide forms. These conditions are favorable for further separation.

2.3 Other research areas

In addition to the above-described works, IHTE performed in a wide range of research aimed at creating a pyrochemical technology for reprocessing spent nuclear fuel. These, for example:

- development of non-consumable anodes;
- thermodynamic modeling of all possible processes;
- physicochemical properties of working media et all.