EXPERIMENTAL JUSTIFICATION OF PYROCHEMICAL TECHNOLOGY FOR REPROCESSING OF MIXED NITRIDE URANIUM-PLUTONIUM SPENT NUCLEAR FUEL

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Processing of spent nuclear fuel (SNF) including plutonium being the material enabling fuel reprocessing via multiple fuel materials recycling is one of the main elements of closed nuclear fuel cycle [1]. Development and implementation of the SNF processing technology is a technological step for closing the nuclear fuel cycle [2]. Pyrochemical technology is considered among the most promising ones, as its enables processing of spent nuclear fuel with a minimum exposure and high combustion, which is economically efficient for the closed nuclear fuel cycle.

To date pyrochemical technology is developed within the framework of the "Proryv" project ("Breakthrough" project) [4, 5]. Alkali metal chlorides serve as radiation resistant technological media. The developed scheme of the pyrochemical processing of mixed nitride uranium-plutonium fuel includes the following basic operations:

• High temperature processing via the UN oxidation to remove the fuel element cladding, removing of the light volatile fission products and transfer of nitrides into the oxide phase.

$$UN \rightarrow UN_{15} \rightarrow UN \rightarrow cladding removal \rightarrow U_3O_8 \rightarrow UO_2$$
.

• Compacting for pellets production from the products of high temperature processing. Zinc stearate is added into the obtained powder of oxidized spent nuclear fuel as a bonding agent. This mixture is used to press pellets that are further sintered in argon at 1700°C.

• Electrochemical reduction of actinide oxides in the LiCl-Li₂O melt (650°C) up to metals using inert anodes [6]. Oxides of U, Pu, Np, Am and Cm reduce at the cathode following the reaction:

$$UO_2 + 4Li \rightarrow 2Li_2O + U,$$

$$PuO_2 + 4Li \rightarrow 2Li_2O + Pu.$$

• High temperature vacuum distillation to purify the reduced pellets from the electrolyte. The reduced actinide pellets are purified from Li_2O via rinsing in the pure LiCl melt and from LiCl via the method of high temperature vacuum distillation at 800-900 °C. It is important to elucidate that during reduction and purification processes nuclear materials do not transfer to the electrolyte.

• Remelting for preparation of consumable metallic anodes in the form of compact ingots. This operation is aimed at the formation of anodes suitable for the electrolytic refining. Regimes of the induction remelting of the reduced UO_2 pellets and UO_2 pellets containing model fuel with additions of Pd, Ru and LA, Nd, Ce oxides imitators in the laboratory glove box. Compact ingots have been obtained.

• Electrolytic refining of actinides in chloride melts for purification of rare-earth and noble metals. A LiCl-KCl eutectic was used as working media. An alloy of actinides containing noble and rare-earth metals served as an anode. First rare-earth metals then uranium and plutonium dissolve at the anode, whereas noble metals under chosen electrolysis regimes do not dissolve and form an anode residue. Uranium and plutonium are extracted at the cathode and rare-earth metals accumulate in the electrolyte.

• Purification of cathode deposit from the electrolyte. The electrolyte is removed using the method of high temperature vacuum distillation at 800–900°C.

Such technological scheme allows purifying nitride SNF from the basic fission products and to use repeatedly uranium and plutonium for fuel fabrication, hence closing nuclear fuel cycle.

To realize a pyrochemical processing technology, radiation resistant chambers with an inert atmosphere are being developed. A pilot sample of radiation-resistant chamber for installation system fine-tuning, creation and maintaining inert high purity atmosphere in large technological volumes and pilot tests has been produced. The pyrochemical processing devices, assembled inside the chamber, operate in a distance mode via robotization, using manipulators, technical vision system and operator virtual media.

References

1. Adamov, E. O. Spent Nuclear Fuel Reprocessing and Nuclear Materials Recycling in Two-Component Nuclear Energy [Text] / E. O. Adamov, Yu. S. Mochalov, V. I. Rachkov et al. // Atomic Energy. – 2021. – Vol. 130. – P. 29–35.

2. Shadrin, A. Yu. Processing methods of mixed U-Pu SNF of fast neutron reactors with increased combustion and small exposure [Text] / A. Yu. Shadrin, V. A. Kasheev, K. N. Dvoyeglazov et al. // Voprosy atomnoy nauki y tekhniki. Ser. "Materialovedeniye i noviye materialy". – 2016. – Vol. 87, No. 4. – P. 48–60. (in Russian).

3. White Book of Nuclear Energy. Closed Nuclear Fuel Cycle with Fast Reactors / ed. E.O. Adamov. – M. : JSC NIKIET Publ, 2020. – 495 p. (in Russian).

4. **Kizub, P. A.** Criticality analysis of pyrochemical reprocessing apparatuses for mixed uranium-plutonium nitride spent nuclear fuel using the MCUFR and MCNP program codes [Text] / P. A. Kizub, A. I. Blokhin, P. A. Blokhin et al. // Nuclear Engineering and Technology. – 2023. – Vol. 55, No. 3. – P. 1097–1104.

5. **Zaikov, Yu. P.** Research and development of the pyrochemical processing for the mixed nitride uraniumplutonium fuel [Text] / Yu. P. Zaikov, V. Yu. Shishkin, A. M. Potapov et al. // J. Phys.: Conf. Ser. – 2020. – Vol. 1475. – P. 012027.

6. Zaikov, Yu. P. Prospects for use of oxide composites as nonsacrificial anodes in high-temperature electrolysis of oxide-halide salt melts [Text] / Yu. P. Zaikov, A. P. Khramov, L. E. Ivanovskii // Russian Journal of Electrochemistry. – 1997. – Vol. 33. – P. 1306–1310.