THE OPTIONS OF SPENT FUEL REPROCESSING AND PARTITION IN THE FRAME OF FUTURE ADVANCED FUEL CYCLES IN RUSSIA

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Abstract

The paper presents the main Russian concepts and projects in the field of spent nuclear fuel and high level liquid waste reprocessing. A new concept of partition was proposed to minimize waste requiring deep geological disposal. The key problems and tasks of the concept are discussed.

1. INTRODUCTION

Currently a number of countries, including Russia, have adopted a strategy of nuclear fuel cycle, including reprocessing of spent nuclear fuel (SNF) and recycling of uranium and plutonium. The technology based on the PUREX process is used in reprocessing plants around the world. Radioactive waste is generated during the reprocessing of SNF. High-level liquid waste (HLLW) is the most dangerous. They contain most of the fission products and also transuranium elements. Vitrified high level waste requires deep geological disposal. Such disposal option must meet high safety and security standards and needs to achieve public confidence. It should be noted that not all countries, operating atomic reactors, have appropriate stabile geological formations. The option of the spent fuel management, which allows minimizing the volume of waste requiring deep geological disposal, seems very attractive. The goal can be achieved by partitioning of most dangerous and long-lived radioactive isotopes and elements (radioactive isotopes of cesium, strontium and iodine, technetium and minor actinides, etc.). Long lived isotopes of Np, Tc and Am can be transmuted in fast reactors. The nuclear cycle based on fast reactors are actively developed in many countries. Group of Cs-Sr after storage in 200-300 years can be dispose in near-surface disposal. Also HLLW contains potentially valuable components, for example, platinum group metals and noble gases, which can be used in radiochemical and other industries.

When the partitioning technology of all dangerous radionuclides implements it is will be possible to reprocess SNF of countries which cannot solve the problem with SNF on their own.

In the last decade researches in partitioning area in Russia are carried out within the framework of two projects — PRORYV (BREAKTHROUGH) and “The experimental demonstration center for the reprocessing of SNF on the basis of innovative technologies”. Both of the projects include involving of transplutonium elements, primarily americium, in the nuclear fuel cycle. The article discusses the achievements in the field of partition in the framework of these projects, and also presents the main terms of a new partitioning program, which is one of the priorities of The State Atomic Energy Corporation ROSATOM today.

2. PARTITIONING WITHIN THE FRAMEWORK OF CURRENT PROJECTS

2.1. Project “The experimental demonstration center for the reprocessing of SNF on the basis of innovative technologies”

The experimental demonstration center (EDC) is under construction at The Mining and Chemical Combine (Zheleznogorsk, Krasnoyarsk region). The advanced technologies of reprocessing of thermal reactors SNF for the large radiochemical plant RT-2 construction will be test at EDC. The basic SNF reprocessing technology of EDC is the simplified PUREX process to produce mixture of U, Pu and Np applicable for fabrication of mixed fuel [1]. EDC technology provides for immobilization of generated during processing radioactive waste by traditional methods – vitrification and cementation. The researches to improve the basic extraction EDC technology is running. The methods of purification U-Pu mixture from Np and Tc have been developed [2-6]. The basic technology of EDC includes voloxidation of SNF, its dissolution in nitrogen dioxide

and then extraction of U, Pu and Np. The dissolution of SNF in low acid solution of salting out agent is alternative option [7]. When salting out agent is used, partition of TPE+REE from PUREX raffinate using PUREX solvent (TBP in hydrocarbon) is possible. A process of TPE+REE extraction with TBP was developed as a part of the modified PUREX process [8]. The dynamic test of the process using PUREX raffinate after reprocessing of a thermal reactor WWER-1000 SNF was carried out. Before extraction step raffinate was evaporated and iron (not less 1 mole/L) was dissolved in it. 50 % TBP was used as exractant, isopar M was used as diluent on the extraction step. Not less 99.99 % TPE (Am + Cm) and REE were recovered from feed solution. In the continuation of these studies the possibility of separation TPE from REE using DTPA and salting out agent was shown.

The main advantage of such mono-solvent approach is complete compatibility of SNF reprocessing with partitioning of TPE, therefore no major changes are required to existing SNF reprocessing technology. The disadvantage is high concentrations of salting out agents in solutions.

2.2. PRORYV project

PRORYV project provides for creation of the new generation nuclear power technologies on the basis of closed nuclear fuel cycle using fast reactors. The engineering pilot demonstration energy complex (PDEC) is being designed. It is being plane to construct at The Siberian Chemical Combine (Seversk, Tomsk Region). According to the project, PDEC includes a fast reactor BREST-OD-300 with lead coolant and SNF reprocessing and fuel production plant [9]. The basic technology of PDEC is combined (pyrochemical SNF reprocessing and hydrometallurgical partitioning of TPE). Completely hydrometallurgical technology is considered as an alternative. A modification of PUREX process is used for SNF reprocessing in the completely hydrometallurgical technology.

The partitioning of TPE technology has been developed within the framework of the project since 2013. Two alternative liquid- liquid extraction processes have been proposed to date. The first process based on UNEX-T solvent and the second one based on well-known ligand N,N,N’,N’-tetraoctyldiglycolamide (TODGA).

UNEX-T solvent is synergistic extraction mixture of ,6-bis(1-aryl-1H-tetrazol-5-yl)pyridines (ATP) with cobalt chlorinated dicarbollide (CCD) in a polar diluent [10, 11]. UNEX-T solvent selectively separates Sr, Cs and actinides (III) from nitric acid media. UNEX-T solvent demonstrates high stability to hydrolysis and radiolysis. The extractability of radionuclides in 1 M nitric acid decreases in the range: Am(III) ≈ Cs > Pu(III) > Sr(II) > Pu(IV) > Eu(III). Separation factors of Am/Eu reaches 100. Pb, Pd, Ni and Co are extracted remarkably. UNEX-T process includes extraction of actinides (III) from HLLW contained no more than 1 M HNO_3; stripping of actinides (III); stripping of Cs and Sr; solvent regeneration with nitric acid solution.

TODGA in F-3 diluent is used in the second TPE partitioning process. F-3 significantly increases the capacity of solvent and prevents the third phase formation [12]. The process includes actinides (III) and REE co-extraction from HLLW contained 3-4 M HNO_3, Zr and Pd scrubbing using solution of complexones, HNO_3 scrubbing with salting out agent solution, selective actinides stripping with buffered DTPA solution and REE stripping. The dynamic test using mixer-settlers set-up was carried out. The feed solution contained about 4.5 g/L REE and trace amounts of ^{241}Am. Not less 99,97 % of americium were recovered. Decontamination factor for the removal of REE from Am product was about 100. Most of the zirconium, molybdenum and palladium were in the raffinate [13].

TODGA/F-3 solvent was tested using a similar solution of PUREX raffinate contained more than 1 g/L Am and about 4.5 g/L REE [14]. About 1.5 g/L of Am were removed at extraction step and separation factors of REE/Am were no less 5 at TPE stripping step. The test showed efficiency of TODGA/F-3 solvent for gram amounts of TPE.

The feasibility of replacing the fluorinated diluent F-3 with a hydrocarbon diluent is being considered.

Am and Cm are not separated in UNEX-T or TODGA process. Displacement complexing chromatography was chosen for TPE separation. The technology of separation of americium and curium was demonstrated in 2016 at PA “Mayak” [15]. Tokem-308 cation-exchange resin with grain size 0.2 mm was used. About 14 g of Cm was separated including 9 g of pure Cm fraction containing 6 activity % of Am. The Cm-Am fraction contained about 4,6 g ^{244}Cm and about 40 g ^{241,243}Am. The Am fraction contained less than 0.8 mass. % of Cm and less than 0.1 activity % of ^{154,155}Eu.

3. COMPREHENSIVE PARTITIONING PROGRAM

The partitioning of TPE from HLLW is actively being investigated in Russia today within framework of two projects discussed above. It is also be noted that commercial partitioning of Cs-Sr from HLLW was carried out in Russia. About 1600 m^3 of acidic HLLW with total β-activity 54 million Ci was reprocessed at "Mayak" Production Association using liquid-liquid extraction with mixture of cobalt chlorinated dicarbollide and polyethylene glycol in F-3 diluent [16]. But despite the developments in the field of partitioning, the creation of technology of partitioning of all critical radionuclides from HLLW is a very difficult task. A program of the comprehensive partitioning was recently proposed in Russia. The key provisions of the program:

- Review approaches and technologies of partitioning estimate of technology readiness, choice of technologies for further researches;
- Assessment of the legislation on radioactive waste management and developing of a regulatory framework taking into account the specificities of comprehensive partitioning;
- Compilation of the initial requirements for comprehensive partitioning;
- Development and test of partitioning and immobilization of each group of radionuclides (cs, sr, am, cm, np, tc, pd);
- Developing and testing a comprehensive technology including partitioning and immobilization;
- Testing of recycling of the fuel containing minor actinides and tc (irradiation, dissolution, processing).

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