PERSPECTIVE SOLUTIONS FOR THE BACK-END OF THE NUCLEAR FUEL CYCLE

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Abstract

Current status of the nuclear fuel cycle in Russia is characterized by significant increase in NPP units under construction based on reactor facilities with VVER-1000/1200/TOI. At the same time rapid expansion of the reactor fleet faces the back-end problems that have to be dealt with. These problems concern reprocessing of SNF and management of products generated as a result of the reprocessing.

The closed nuclear fuel cycle is a basis of package of services offered by Rosatom State Corporation at foreign markets.

1. INTRODUCTION

Due to the development strategy chosen, in recent years the RT-1 Plant (Mayak Production Association) has implemented several large-scale projects that brought the plant to a high level of technological development in terms of extension of the range of SNF types reprocessed, increase in reprocessing capacities of the plant, and systematic development of RW management infrastructure.

Full-scale reprocessing of SNF from the VVER-1000 was adopted. Capabilities of reprocessing SFA of foreign design (PWR and BWR) was demonstrated using reprocessing of a mock-up of TVS-Kvadrat (square fuel assembly) as an example. Test reprocessing of spent MOX-fuel from the BN-600 reactor was implemented. Today the RT-1 Plant is capable of reprocessing SNF of any type and composition.

However, in spite of intensive progress, the back-end technologies are far from being perfect and have a number of challenges concerning in the first place significant amount of waste generated (between 0.4 and 1 tonne of HLW glass per 1 tU). At the moment this aspect restrains large-scale reprocessing of SNF, which significantly decreases appeal of SNF reprocessing services both on domestic and foreign markets, and on the whole dictates necessity of optimization and improvement of HLW management technologies.

As of today, one of the main lines that offers advanced solution to the problem is partitioning of liquid HLW.

2. EXPERIENCE OF LARGE-SCALE HLW PARTITIONING

In the late 1970s USSR began to develop methods on producing separate fractions of radionuclides, so-called ‘partitioning method’. Development of partitioning methods is mainly represented by recovery of target elements or groups of elements from HLW and arrangement of their proper management.

The first in the world pilot plant for HLW partitioning (facility UE-35 (Rus. УЭ-35)) based on an extraction system with chlorinated cobalt dicarbollide (CCD) started its operation at Mayak in 1996. In 1999, at the same facility, additional unit was installed for recovery of actinides and REE from raffinates. The unit was based on precipitation technique. By 2001 complex extraction-and-precipitation technology was optimized, and the UE-35 facility was switched to the mode of regular commercial operation. The main task of the UE-35 facility was to extract caesium and strontium from HLLW of complex composition. These elements prevented waste from being directly vitrified into aluminophosphate glass. For the period from 1996 to 2002 the facility has reprocessed more than 1,400 m^3 of high-level solutions of high-salt composition (about 50 mln. Ci of caesium and strontium was extracted).

In the course of the UE-35 facility operation vast experience was gained, a lot of various process flowcharts and extracting agents were tested. The following basic versions of the main HLW-partitioning methods were tested at least on the test bench at Mayak:

- extraction technology of extracting strontium and caesium by means of a system based on crown-ethers;
- extraction technology of extracting strontium and caesium by means of a system based on chlorinated cobalt dicarbollide (CCD, carborane);
- technology of extracting and separating REE and TPE by means of an extraction system based on CCD;

- extraction technology of extracting REE and TPE by means of a system based on heteroradical phosphine oxide (HPO);
- extraction technique of extracting and separating REE and TPE by means of a system based on carbamoylmethyl phosphine oxide (CMPO);
- extraction technique of extracting and separating REE and TPE by means of a system based on N,N,N',N'-tetraoctyl diglycolamide (TODGA);
- separation of TPE and REE by displacement chelation chromatography with sulphocationites;
- sorption-extraction of caesium using inorganic electron-ion exchanger, i.e. potassium-copper ferrocyanide (FS-10);
- sorption-extraction of palladium by means of SCN carbon with subsequent sorption-finishing using VP-1Ap anion exchanger;
- sorption-extraction of technetium by means of selective anion exchanger (quaternary phosphonium compound) with further sorption-finishing using VP-1A;
- sorption-extraction of ruthenium using oxidized anion exchanger ANO-251 (rus. AHO-251) with pyridine nitrogen as a functional group.

Processes involving crown-ethers to extract minor actinides, as well as extracting agents of other series (HPO, CMPO) were tested out. Within the project Breakthrough (Proryv) a process was developed, where N,N,N,N – tetraoctyl diglycolamide (TODGA) was used as an extracting agent in polar diluent, i.e. meta-nitrobenzofluoride, with further chromatographic separation of americium and curium. Dynamic hot tests of the method of Am and Cm extraction from real HLW were performed by means of a system based on TODGA. Am recovered made 99.9%. The method of Am and Cm separation was successfully demonstrated using the pilot facility at Mayak.

Results of the activities performed at Mayak confirm a fundamental possibility of partitioning of high-level raffinate with recovery of caesium and strontium (in combination or separately), REE and TPE (in combination or separately). At the same time there is a possibility to obtain not only a contaminant-free fractions, but pure fractions.

3. PARTITIONING TODAY

The current status of development of partitioning methods in Russia includes basically tests of methods of extracting and separating actinides within Breakthrough Project and a range of works on choosing highly-selective ligand with a view to separating actinides and REE, as well as Am/Cm.

In consideration of renewed interest to partitioning methods, the main target of Rosatom enterprises at the moment is to create a competitive and customer-centric product (service) in the sphere of the back-end (SNF and RW management), i.e. commercialization of partitioning technologies.

Within the task, the focal point is the following partitioning processes (methods):

- extraction and production of a final form of Cs-St fraction that makes more than 95 % of activity of RW from SNF reprocessing. This fraction is characterized by medium-lived (half-life period is ~ 30 years) radionuclides, which predetermines a possibility to repeatedly decrease activity of the fraction in the course of the storage period, to switch RW to ILW and, accordingly, to bury the waste at shallow depth within the limits of the current legislation;
- extraction and recovery of minor actinides (Np, Am, Cm). These radionuclides define radiotoxicity of RW in the long term (more than 1000 years). This fraction should be stored for a long term in a maximum compact and reliable form, i.e. in the form of ceramic-like or glass matrix. A possible option for a foreign customer is burning (transmutation) of Am and Np in fast neutron reactors (BN-600, 800, 1200) or, over a long term, in a molten-salt / liquid-metal reactor. At the same time, intermediate storage of curium is envisaged with a view to transforming it into Pu in the course of time;
- extraction of REE and their immobilization into the final matrix with further long-term storage;
- recovery and decontamination of platinum-group metals (Ru, Pd) for the purpose of recycling with further industrial application;
- optional extraction of elements with a view to using or analyzing (Tc, Am, I, Kr, Np, Cs, Sr).

In the context of the RT-1 Plant operation and depending on tasks assigned, partitioning methods can be used for:

— reprocessing of complex liquid waste accumulated in the course of previous activities or current HLLW generated in the course of SNF reprocessing for the purpose of their subsequent solidification, including:
  - decontamination of HLLW with changing its status to ILW for further cement-solidification and burial;
  - removal of fractions of some radionuclides (or groups of radionuclides) from HLW subject to vitrification with a view to eliminating impact of the components on glass properties.

— extraction from HLW and purification of fractions of some radionuclides (or groups of radionuclides) with a view to:
  - obtaining Cs and St fractions for subsequent return to a country of fuel origin;
  - producing radionuclides of practical interest for science and industry.

4. PROMISING TRENDS

To improve the back-end, it is essential not only to reduce amounts of RW generated but provide conditions for its maximum safe and economically-acceptable ultimate disposal. Class 1 RW is long-lived HLW with high heat-generation, safe burial of which is the most challenging issue, because it remains potentially hazardous for environment and population during several billion years. At the same time, amount of long-lived heat-generating HLW to be buried is of less importance than its specific heat-generation.

The most promising trends in this field are partitioning methods that will allow removing the main heat-generating fraction in the form of Cs or St at the first stage, and that can significantly decrease RW radiotoxicity at the next stage by recovering minor actinides (Am, Cm).

The estimations performed indicated that after extracting fractions of Cs+Sr and Am+Cm, RW vitrified in borosilicate glass will be classified as Class 2 at the end of the period of about 50 years from the moment of vitrification. According to the criteria of RW acceptance for burial effective in the territory of the Russian Federation (Federal Rules and Regulations NP-093-014 / Rus. НП-093-014), specific heat-generation of solidified RW of Class 2 must not exceed the value of 100 W/m^3.

It is worth noting that according to outcomes of the UE-35 facility operation at the RT-1 Plant till 2002, Cs-Sr recovery factor made 97% (extraction from HLW). But at that point degree of this fraction recovery was not a first-priority task among the tasks set back then. At the moment technological capabilities available provide recovery of the complete Cs-Sr fraction from HLW (up to 99.0%). Further increase in recovery degree requires additional extraction cascades.

In respect to possible minimization of RW amount to be buried in deep geological formations, additional recovery of stable fission elements (Mo, Zr, etc.) from liquid RW is considered a promising direction. Table 1 presents a list of limit weight-contents of HLW components incorporated into borosilicate matrix.

TABLE 1. LIMITATIONS ON VITRIFICATION OF HLW COMPONENTS IN BOROSILICATE GLASS.

HLW component	Content of a component in glass, wt. %	Limit mass fraction of a critical component, %
UO_3	0.18	5
PuO_2	0.03[b]	
NpO_2 (50%)[a]	0.16	0.034 (PuO_2)
AmO_2	0.36	0.6 (TUE)
CmO_2	0.05	
SrO (3%)	0.02	-
Y_2O_3	0.33	-
ZrO_2	2.91	20
Cs_2O (3%)	0.06	-
BaO	0.83	-
TcO_2 (100%)	0.60	-
RuO_2	1.51	
Rh_2O_3	0.21	4 (precious metals)
PdO	0.50	
La_2O_3	0.79	
Ce_2O_3	1.79	
Pr_2O_3	0.69	
Nd_2O_3	2.68	7 (REE)
Pm_2O_3	0.15	
Sm_2O_3	0.51	
Eu_2O_3	0,13	
NiO	0.33	1, 0.5
Cr_2O_3	0.49	0.5, 0.6
Fe_2O_3	1.69	4.5, 9
Total TUE	0.6	0.6
Total precious metals	2.23	4
Total REE	6.74	7
Degree of HLW incorporation in terms of oxides, %	17	-
Weight of glass with 1 tU after recovery of Mo (degree of HLW incorporation makes 17%), kg	240	-

[a] amount of a component extracted from SNF and to be solidified as an HLW fraction is specified in brackets

[b] values indicating component contents that limit HLW incorporation into glass is highlighted with a thickened line

Results of the performed estimations demonstrated that the limit on incorporation of four components (groups of components), i.e. PuO_2, TPE, REE and Cr_2O_3, will be reached when about 17 wt. % of HLW components is incorporated into borosilicate matrix without Mo and Cs+Sr fractions. This indicates absence of any prospect to increase degree of component incorporations with a view to reducing amount of HLW vitrified. Therefore, with significant degree of certainty one can say that amount of vitrified HLW, without Cs+Sr fraction (even after Mo extraction from the stream) will be not less than 240 kg per 1 t of SNF reprocessed.

5. INNOVATIVE SOLUTIONS FOR THE BACK-END

When using partitioning methods, alternative and innovative option is to arrange practical use of heat-generation parameters of Cs–Sr and to find physical-and-chemical matrix for Cs–Sr with a view to recycling after specified lifetime of Cs–Sr product runs out.

One of the possible solutions to practical application of useful properties of the heat-generating fraction extracted from HLW is manufacturing of a practical heat source with a view to using it at the production site. Mayak specialists estimated technical parameters of such Sr source and possible economic effect of using radioisotope heat sources (RHS) as additional heat sources for the RT-1 (heating of the air of air-supply centers and special-network heating systems). It has been found out that it is technically possible to use additional heat sources (RHS) instead of 50 % of hot water from ventilation centers. Design parameters of a practical Sr-90 RHS are presented in Table 2.

TABLE 2. DESIGN PARAMETERS OF RADIOISOTOPE HEAT SOURCE (RHS).

Parameter	Result
Heat power of capsule	1300 W
Volume of active part	1.364 L
Number of capsules	5
Material of shielding	Steel 3 (cast iron)
Thickness of shielding	360 mm
Total power of RHS assembly	6500 W
Weight of RHS assembly	~ 5000 kg
Dimensions of RHS assembly	height: 60 cm diameter: 12 cm

Design of kilowatt-class Sr-90 RITEG (radioisotope heat and electric generator) was also developed to produce energy. The possible applications are power supply of low-current energy consumers (e.g. physical protection elements, observation systems), accumulation of generated power by accumulation systems (charging). Design parameters of RITEG are shown in Table 3.

TABLE 3. DESIGN PARAMETERS OF A RADIOISOTOPE HEAT AND ELECTRIC GENERATOR (RITEG).

Parameter	Result
Volume of active part	1.3 L
Heat power	1025 W
Weight of RITEG assembly	3500 kg
Temperature of functional surface	570ºC
Maximum temperature of active part	900ºC

6. CONCLUSION

Innovative solutions in the sphere of SNF management, current level of development of HLW partitioning methods in combination with promising solutions concerning adopting effective ways of practical (useful) application of extracted fractions on an industrial scale allow offering the Customer balanced solutions concerning the back-end and providing sustainable development of the atomic sector around the world.