

SPENT NUCLEAR FUEL REPROCESSING AND NUCLEAR MATERIALS RECYCLING IN TWO-COMPONENT NUCLEAR ENERGY

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The project Proryv [Breakthrough] now being implemented in our country is aimed at achieving a new quality of large-scale nuclear power, development, origination, and industrial implementation of a closed nuclear fuel cycle with thermal and fast reactors. Here the current status of the technologies for reprocessing spent nuclear fuel and recycling plutonium fuel is analyzed, and it is shown that the prerequisites for closing the nuclear fuel cycle in two-component nuclear energy in the mid-term from the mid-2030s are present.

A closed nuclear fuel cycle is being created in our country on the basis of advanced thermal and fast reactors. The key elements are the reprocessing of spent nuclear fuel and the management of radioactive waste, aimed at achieving the following goals:

- answers to deferred problems concerning the management of spent nuclear fuel and radioactive waste, consistent reduction of VVER and RBMK spent nuclear fuel, and prevention of further accumulation of spent nuclear fuel [1];
- bringing into the nuclear fuel cycle plutonium as a product of reprocessing spent VVER fuel for the fabrication of fast reactor fuel and full utilization of the energy potential of native uranium (^{238}U) by means of multiple recycling of fuel materials;
- fractionation of radioactive waste, extraction of long-lived minor actinides for secondary burning or transmutation in reactors, which, together with multiple recycling of plutonium, can significantly reduce the environmental hazard of radioactive waste based on the principles of radiation-equivalent disposal;
- technological support of the nonproliferation regime through the stage-by-stage elimination of the circulation of enriched uranium and the separation of pure plutonium.

The developed closed-NFC technologies together with the reactor technologies should secure competitiveness with other large-scale energy technologies [2, 3].

Modern closed-NFC infrastructure. The transition to a closed nuclear fuel cycle using fast reactors is accepted in our country as a strategic direction for the development of nuclear energy [4, 5]. An essential basis for this is the successful operation of the BN-600 industrial fast reactor, unparalleled abroad, at the Beloyarsk NPP. The No. 4 unit with BN-800 was commissioned in 2016, the initial purpose being the development of technologies for NFC closure. The RT-1 plant of the Mayak Production Association has been operating since 1977. Its modernization will allow the plant to operate until 2035 with the possibility of extending its operation period. RT-1 can reprocess up to 400 t of spent fuel per year from VVER-440, -1000, BN-600, and RBMK-1000 power reactors as well as transport and research reactors. Over its operating period the RT-1 plant has reprocessed more than 6000 t of spent fuel. The final products of the plant are uranyl nitrate with ^{235}U content 1%, triuranium octaoxide with ^{235}U content 14–17%, and recycled reactor-grade plutonium dioxide [6].

A plant for the production of mixed uranium-plutonium oxide fuel for BN-800 and the first phase (research hot chambers) of the Experimental Demonstration Center for the reprocessing of VVER-1000 spent nuclear fuel [7] were commissioned

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at the Mining and Chemical Combine. The second stage of the experimental demonstration center for the production of initial oxides suitable for the fabrication of mixed uranium-plutonium oxide fuel and/or REMIX fuel, fuel based on an unseparated unenriched mixture of reprocessed uranium and uranium-enriched plutonium, is to be commissioned in 2021 [8].

A pilot demonstration energy complex is being created at the Siberian Chemical Combine as a part of a power unit with a BREST-OD-300 reactor and closing the nuclear fuel cycle of on-site production, which includes modules for reprocessing irradiated mixed uranium-plutonium nitride fuel and fabrication/refabrication for manufacturing the starting fuel rods from imported materials and fuel rods from recycled materials. Currently, the construction of a fabrication/refabrication module of the Experimental Demonstration Power Complex for the production of mixed nitride fuel is being completed; commissioning is planned for 2022. Construction of a spent fuel reprocessing module is to commence in 2024 [9]. The technology for carbothermal synthesis will be implemented in the module for fabrication/refabrication of nuclear fuel [10]; the technology for combined (pyrochemical + hydrometallurgical) reprocessing of nitride fuel, which is also suitable for reprocessing mixed uranium-plutonium oxide spent fuel from fast reactors is to be implemented in the module for reprocessing spent fuel [10]. The development of a purely hydrometallurgical technology as a backup variant for reprocessing spent fuel from fast reactors is nearing completion.

The world's first pilot demonstration energy complex must demonstrate stable operation of a full complex of facilities securing NFC closure. The on-site variant of fuel cycle organization will make it possible to work out technologies for a closed NFC with a short external fuel cycle time in the shortest possible time within a single site. The introduction of competitive fast reactors and a transition to a two-component structure of nuclear energy with a closed NFC are expected starting in the mid-2030s [1, 4, 5].

Plutonium recycling in thermal reactors. The optimal scheme for a closed nuclear fuel cycle for two-component nuclear energy presupposes the use of plutonium separated from the spent fuel of thermal reactors in the starting loads of competitive fast reactors, followed by a transition of the latter to a fuel self-supply regime. The use of plutonium in fast reactors makes it possible to maximize the use of the neutron potential of fissile materials and to switch to large-scale nuclear energy, 3–4 times higher than its current power level by 2100, with a minimum consumption of natural raw materials, without forcing the fuel breeding ratio and heat density of the fast reactor cores. However, considering a possible delay in large-scale introduction of fast reactors as well as provisioning of foreign nuclear power plants with VVER fuel, variants for recycling plutonium in thermal reactors are also being considered. Mixed uranium-plutonium oxide fuel is considered as conventional for this purpose [11]. In terms of plutonium content, spent mixed uranium-plutonium oxide fuel from VVER occupies an intermediate position between spent uranium and uranium-plutonium oxide fuel from fast reactors and it is much more easily reprocessed than mixed uranium-plutonium oxide spent fuel from fast reactors, and for this reason it is not considered in the present article.

A new REMIX fuel is proposed in order to use plutonium in VVER – a mixed uranium-plutonium fuel, obtained from an unseparated mixture of uranium and plutonium separated during the regeneration of spent VVER fuel with the addition of enriched uranium. In contrast to mixed uranium-plutonium fuel obtained from plutonium recovered during reprocessing of spent VVER fuel, such fuel makes it possible to recycle plutonium multiple times, at least up to seven recycles [12]. REMIX is a mixed uranium-plutonium oxide fuel based on an unseparated mixture of uranium and plutonium recycled from spent nuclear fuel with native or regenerated uranium enriched in ^{235}U to 19.75%. The use of such a mixture will reduce the consumption of enriched uranium by about 25% [13–17].

One of the variants of recycling such fuel in VVER is shown in Fig. 1. Such fuel contains about 1% ^{239}Pu and 4% ^{235}U and can be irradiated to burn-up 50 GW·day/t in 4 yr. Spent REMIX fuel contains 2% ^{239}Pu and 1% ^{235}U , and after reprocessing unseparated uranium and plutonium can be reused for the production of REMIX fuel with the addition of low enriched uranium [12–17].

It is impossible to ensure the recycling of long-lived actinides in VVER using REMIX fuel, so that a system with such fuel must be supplemented with reactors that ensure their transmutation, for example, fast or molten-salt incinerator-reactor [18].

In 2016, as part of a concept substantiation project, experimental fuel assemblies with REMIX fuel were manufactured and for the first time installed for irradiation in VVER at the Balakovo NPP. Fuel rods with the same fuel were supplied to the MIR research reactor. At present, the third micro-run of fuel irradiation in VVER is underway; the irradiation program in the MIR reactor is nearing completion [19]. In the event of successful completion of the cycle of operations, the fundamental feasibility of the fuel cycle with REMIX fuel can be considered as proven.

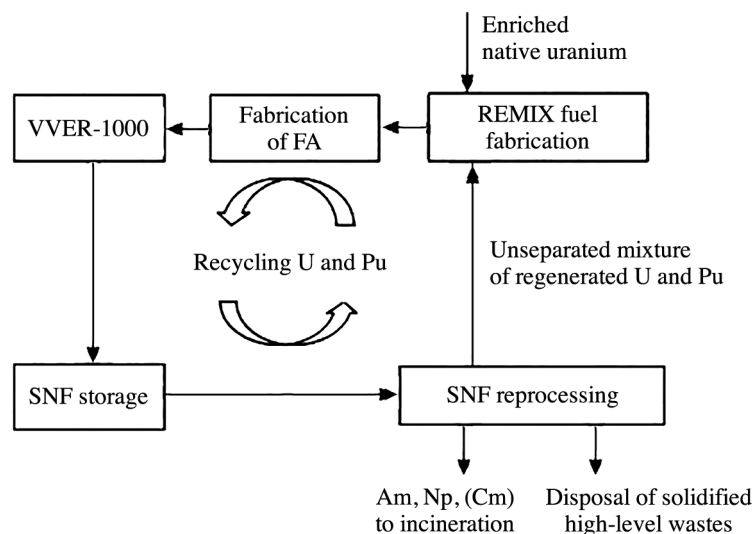


Fig. 1. VVER fuel cycle with REMIX fuel.

It should be noted that since the close chemical composition is close, there are almost no differences in the reprocessing of spent uranium and REMIX fuel, so that the possibility of reprocessing spent REMIX fuel at the RT-1 plant and at the Experimental Demonstration Center for the reprocessing of VVER-1000 spent nuclear fuel is beyond doubt.

Plutonium recycling in fast reactors. Plutonium recycling as applied to fast reactors is being optimized in BN-600 and BN-800 reactors and is planned in BN-1200, BREST-OD-300, and BR-1200. It should be noted that until now our country had no industrial experience in operating fast reactors using mixed uranium-plutonium fuel. Such experience is being gained in BN-800, which already partially uses mixed uranium-plutonium oxide fuel in the existing hybrid zone and which is to be switched to full load with such fuel in the next 2–3 years. The use of conventional mixed uranium-plutonium oxide fuel is also not excluded in next-generation fast reactors, but most promising is the introduction of a new, dense, mixed uranium-plutonium nitride fuel that has not been previously used in power reactors.

The reprocessing of BN-800 spent fuel has not yet been performed. In 2012 and 2014, eight fuel assemblies with BN-600 spent mixed uranium-plutonium oxide fuel with a burnup 73–89 GW·day/t and hold-up time of about 20 years were reprocessed in the RT-1 plant. The reprocessing included dissolution, fining, extractive separation, and separation of uranium and plutonium with purification from fission products without mixing with the spent fuel of thermal reactors, and purification of uranium and plutonium in the corresponding refining cycles with mixing with the spent fuel of thermal reactors. No heightened losses of plutonium to waste during reprocessing were recorded, the plutonium content in the raffinate was less than 0.1 mg/L, and no plutonium was found in the insoluble residues [20]. All this allows us to assert that reprocessing of both BN-800 spent fuel and, if necessary, VVER at RT-1 will not cause difficulties even taking into account the design limitations of RT-1.

A combined reprocessing technology – the Pyro-Hydro (PH) process – has been developed for reprocessing spent uranium-plutonium nitride and oxide fuel from fast reactors with a short hold-up time; it presupposes the joint separation of uranium and plutonium, taking into account the requirements of technological support for the nonproliferation regime, as well as its purely hydrometallurgical variant [20, 21]. Both variants also call for the extraction and separation of americium and curium, obtaining as the desired product a mixture of uranium, plutonium, and neptunium oxides as well as uranium-americium oxide and uranium-curium oxide mixtures. Alternatively, mixed oxides of a mixture of uranium, plutonium, neptunium, and americium can be obtained [21–23]. A pyrochemical process stage of the combined technology is under development [24]; research on the hydrometallurgical process stage of the combined technology is nearing completion (Fig. 2) [23].

Extraction of actinides for incineration and reduction of radioecological hazard of buried high-level waste. In accordance with the methodology of radiation-equivalent management of radioactive waste to reduce the radioecological hazard in two-component nuclear power, the following measures are assumed:

- exclusion of the disposal of spent fuel from VVER and fast reactors;

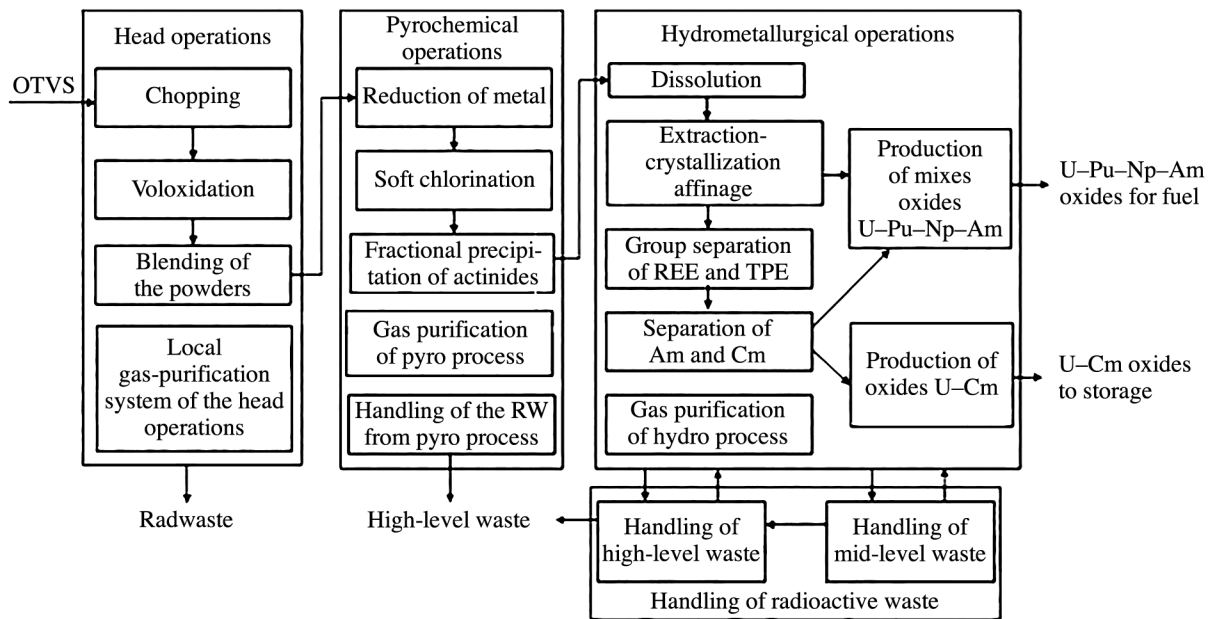


Fig. 2. Schematic flow diagram of the combined technology for reprocessing mixed uranium-plutonium nitride and oxide spent fuel (PH-process).

- reprocessing of VVER spent fuel with specified fractionation to transfer at least 99.9% of plutonium and actinides (neptunium and americium) as well as long-lived fission products into the fuel cycle of fast reactors;
- multiple recycle of actinides in fast reactors with transmutation of actinides and long-lived fission products;
- extraction and hold-up of curium for 70–100 yr, followed by the use of fission products, mainly plutonium, in fast reactors;
- deep cleaning of radioactive waste to be buried from actinides (the content of actinides in radioactive waste does not exceed 0.1%).

At the first stage of the formation of a closed NFC in the absence of a sufficient number of fast reactors, the option of recycling plutonium in the composition of REMIX fuel or mixed uranium-plutonium oxide fuel in VVER with the release and transmutation of actinides in fast or molten-salt reactors is considered [25].

Technologies for the group separation of transuranic and trans-plutonium elements have been developed and tested within the framework of project Breakthrough. The extraction system N, N, N', N'-tetraoctyldiglycol-amidemethanobenzotrifluoride is currently under consideration [8, 22]. In 2017–2018, hot dynamic tests of the technology for the separation of americium and curium from real high-level waste were performed using a system based on N, N, N', N'-tetraoctyldiglycolamide-methanobenzotrifluoride; americium extraction >99.9% was attained. A test based on the use of sulfonic cation exchangers for the extraction of curium and americium from the concentrate of rare earth and transplutonium elements obtained from the raffinate of the extraction processing of spent VVER-440 fuel was performed at the Mayak PA pilot plant. At the refining stage, using the cation exchanger Tokem-308 with grain size 220 μm in the separating columns, about 14 g of curium were extracted, of which 9 g was a curium–americium fraction with activity <6%. The mixed curium–americium fraction contained about 4.6 g of curium and about 40 g of americium. In the americium fraction, the content of curium is <0.8 wt% and $^{154,155}\text{Eu}$ <0.1% in terms of activity [21, 23]. Thus, in terms of reprocessing spent fuel from both VVER and fast reactors, a technology for the extraction and separation of americium and curium has been developed and tested, which makes it possible to proceed to the substantiation of fuel with americium for bringing fast reactors into the fuel cycle.

Radioactive waste management. Systems for managing radwaste, whose environmental aspects are regulated by classification and formulation of requirements for the physicochemical characteristics of packages and the conditions for their disposal (acceptance criteria), have been or will be implemented at the operating RT-1 plant, at the pilot-demonstration center, now under construction, for reprocessing of VVER spent nuclear fuel, and in a to be built module for reprocessing

BREST-OD-300 spent nuclear fuel at the pilot demonstration energy complex [26, 27]. The requirements are fulfilled at existing and planned facilities. The economic component of the problem of radioactive waste disposal is regulated by the tariffs introduced for radioactive waste disposal [28], which are determined for each class of radioactive waste. As a result, the disposal costs of a radwaste producer are determined by the class and final volume of the generated wastes, including the volume of the package (container). The work in the field of two-component nuclear energy must be directed precisely at the systems cost reduction of the entire radwaste management cycle.

On reprocessing of spent fuel from fast reactors with extraction of actinides with post-reactor exposure 7 yr and burnup 70 GW·day/t, the volume of vitrified radioactive waste (borosilicate glass) in accordance with regulatory requirements will be about 0.7 m³/t of spent nuclear fuel. The data in [29] show that rejection of actinide extraction into a separate fraction and inclusion of actinides in the radwaste (vitrification together with the content of fission products in the spent fuel) lead to significantly larger volume of disposed waste – the volume of the matrix increases by about 0.6 m³/t of spent fuel [30].

A similar volume reduction of the final radwaste with the exclusion of actinides from the radioactive waste is observed on reprocessing of spent fuel from fast reactors [29]. Vitrified waste is subject to hold-up in order to reduce the specific heat release to 1 W/L and subsequent deep underground disposal. The elimination of actinides significantly reduces the long-term biohazards of disposal, i.e., the requirements of the isolation barriers of the storage facilities.

A radwaste management system is examined in detail in [23, 29, 31]. The volumes of radwaste requiring disposal in deep formations are determined by two parameters – the absorbed dose over the storage period and the heat release at the commencement of disposal. To reduce the volume of radioactive waste requiring burial in deep geological formations it is necessary to:

- separate the actinide fraction from the radioactive waste and send the separated isotopes to incineration;
- validate the possibility of increasing the amount of fission products brought into the matrix or after separation of the actinide fraction from the waste and after storage for about 60 yr (two half-lives of the main dose-forming cesium and strontium) in order to digest the vitrified waste, thereby renewing the matrix and reducing the volume 3– 4-fold.

Conclusion. The problem of managing spent fuel in two-component nuclear power is to reprocess spent nuclear fuel from thermal and fast reactors and return not only uranium and plutonium but also actinides into the fuel cycle. Analysis shows that the demonstration of NFC closure for plutonium at the industrial level has already commenced. Moreover, the prerequisites have been created for the industrial demonstration of technologies for NFC closure not only for plutonium but also for actinides, including from the standpoint of the reprocessing of spent fuel, namely:

- mixed uranium-plutonium oxide fuel for BN-800 is being fabricated using the energy plutonium separated at RT-1, mixed uranium-plutonium nitride fuel is fabricated to validate its use in fast reactors and REMIX fuel to justify its use in VVER;
- reprocessing of spent mixed uranium-plutonium oxide fuel was demonstrated at RT-1;
- technologies for extraction of actinides, including the extraction and separation of americium and curium, have been developed and tested on real spent fuel and high-level waste.

In Russia, the conditions are now ripe for demonstrating technologies for fuel cycle closure and gradual transition to two-component nuclear energy with a closed fuel cycle, namely:

- a second start-up complex of the Experimental Demonstration Center for the reprocessing of spent nuclear fuel from thermal reactors is close to completion and commissioning;
- capacity for the fabrication of mixed uranium-plutonium oxide and nitride fuel has been created or is about to be completed;
- RT-1 is capable of reprocessing spent fuel from fast reactors (subject to design constraints);
- new capacities for reprocessing spent fuel from the BREST-OD-300 reactor using new technologies for reprocessing, extracting, and separating americium and curium are being planned.

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