OVERVIEW OF CURRENT RUSSIAN ACTIVITIES IN P&T AREA

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Introduction

The general policy of radioactive waste management is consistent with the long-term plans for nuclear power development adopted in each country. Russian activities aimed at setting up in the future a fuel cycle of nuclear power with reasonably minimised quantities of Radwaste subject disposal are being carried out by Minatom as part of the general Strategy for development of national nuclear power. [1] Several key missions of this Strategy deserve special mention:

- in the next 20-40 years, construction of advanced thermal reactors which will run on enriched uranium until the economically acceptable reserves of natural uranium are exhausted;
- reprocessing of all spent fuel of thermal reactors to separate plutonium and long-lived nuclides;
- development of a new generation of fast reactors which will meet the requirements placed on innovative reactors for large-scale electricity production (economic efficiency, safety, minimised Radwaste, proliferation resistance);
- after 2030, deployment of a system of innovative fast reactors, using plutonium separated from spent fuel of thermal reactors, and solution with their help of the totality of problems associated with transmutation of long-lived nuclides.

Task named “Transmutation” was set up in 2001, within the general Minatom’s programme, to develop a scenario for transition to the fuel cycle of future large-scale nuclear power (Figure 1) as part of the above Strategy and to resolve the technological problems of minimising the quantities of long-lived nuclides generated in the closed fuel cycle and subject to final disposal. Transmutation Task combines and arranges the work in the following directions:

- to study various scenarios for transition from the current state of the industry to large-scale nuclear power whose fuel cycle will provide radiation-equivalent management of long-lived radioactive waste, including its disposal;

- to develop radiochemical technologies meant for homogeneous (small additions to fuel) or heterogeneous (special fuel rods or assemblies) transmutation of minor actinides (MA) and long-lived fission products (LFP) in the closed fuel cycle of fast reactors;
- to optimise the procedures and conditions of MA and LFP transmutation;
- to verify nuclear data libraries for MA and LFP, and to update the cross-sections in various neutron energy regions;
- to investigate alternative approaches to MA and LFP transmutation.

Sections 1-4 describe main activities in the above areas, funded by Minatom.

Nuclear power development scenarios

Spent nuclear fuel (SNF) is the main source responsible for long-term (a thousand years or more) radioactivity hazard generated by nuclear power. Reliable prediction of SNF behaviour after final disposal during tens or hundreds of thousands of years is impracticable.

The main contributors to the long-term radiation hazard of SNF are actinides (plutonium, americium, curium) and some fission products (^{129}I, ^{99}Tn, etc.). Transmutation is regarded as a means of reducing the mass (activity) of long-lived nuclides subject to burial. This term is usually interpreted as follows:

- in case of actinides – burning (fission) in a neutron flux, i.e. conversion to fission products whose biologically equivalent activity – with more than 500 years of cooling – is lower by 3-5 orders of magnitude than that of the initial actinide mixture (Pu, Am, Cm, Np);
- in case of long-lived fission products (LFP) – exposure to a neutron flux for their conversion to stable nuclides or those easily decaying to stable forms through nuclear reactions, mainly (n,γ) .

The strategy for Radwaste management proceeds from the principle of radiation equivalence between the natural radioactive material (uranium, or thorium) utilised in the fuel cycle of nuclear power and its Radwaste to be disposed. This is a quantitative criterion, which can be calculated and optimised. Long-lived high-level waste (LHW) should be transmuted at least till to the biologically equivalent activity of waste to be disposed drops down to the level of natural uranium consumed. Such equivalence may be achieved either at the time of disposal or after a relatively short – in historical terms – period of time (e.g. 200-1 000 years), which can be reliably estimated. This approach affords reasonable reduction of the LHW mass and activity, which clearly does not obviate the need for ensuring compliance of the specific disposal conditions with the local health regulations, as well as with other rules and requirements. The principle of radiation equivalence will be met in the most consistent way if the waste is disposed at the uranium mining sites developed by new, environmentally friendly methods. A promising and recommended option is to mine uranium together with its long-lived decay products (^{230}Th, ^{226}Ra, ^{231}Pa), which would facilitate remediation of the mining areas.

In the current open nuclear fuel cycle, which manage SNF as nothing but waste, radiation equivalence may be achieved only after 100 000-500 000 years of fuel cooling. It means that radiation equivalence is nonsensical from the pragmatic viewpoint. Some prior studies have shown that such radiation equivalence is feasible, given the following:

- The whole SNF amount from thermal reactors is reprocessed with partitioning, for plutonium, MA and LFP to be transferred to the fuel cycle of fast reactors.
- Fast reactors generating electricity and operating in a closed fuel cycle burn the bulk of actinides and transmute LFP.
- Plutonium, americium and some other long-lived nuclides are removed from waste to a fairly high degree.
- High-level waste should go through interim storage prior to final disposal.

In 2001, RDIPE and RRC-KI using system models of nuclear power, carried out the set of scenarios studies on nuclear power transition to the fuel cycle, depicted in Figure 1, according the Strategy implementation. The dynamics of changes in the installed capacities of plants with thermal and fast reactors was studied with regard to various estimates of Russian natural uranium reserves. The researchers also explored the possibility of resolving the problem of Pu and MA transmutation in a growing system of fast reactors without resorting to additional dedicated transmutation units. Thermal reactors in that study were exemplified by VVERs and RBMKs whose commissioning dynamics to the year 2020 was taken as determined in the Strategy. Fast reactors were represented by blanket-free BREST-1200 (lead-cooled) or BN-800 (sodium-cooled) with U-Pu-MA nitride fuel and core breeding ratio of 1-1.05 designs. Beginning with the year 2030, their commissioning rate varies depending on the reprocessing plant capacity to remove plutonium from SNF of thermal reactors required for the first fast cores loadings. The closed fuel cycle of fast reactors affords transmutation of actinides both generated in the fast reactors themselves and contained in the irradiated fuel of thermal reactors. The latter actinides are incorporated in the fuel of the first charge. Transmutation of minor actinides in fast reactors accompanies energy generation, leaving the efficiency of the nuclear power system practically unaffected. Transmutation of LFP was omitted in this study as a less urgent problem at the current stage of the work.

It has been demonstrated that transmutation of minor actinides is fully feasible in the system of thermal and fast reactors under consideration. Computation results for one of the examined cases are given in Figures 2-4. It was assumed that construction of new thermal generating capacities would not proceed beyond the year 2020 (meaning VVER-1000s with the operating life of 50 years). Beginning in 2030, BREST-1200 power reactors would be put in operation at a rate of 1 unit per year. The SNF of thermal reactors will have accumulated~570 t of plutonium (including 320 t of ^{239}Pu and ^{241}Pu) and 120 t of MA (Figure 2). By the year 2100, using this plutonium together with their own small breeding, fast reactors can raise their capacity to 82.8 GW (Figure 3). Minor actinides from SNF of thermal reactors will be included in the first charges of fast reactors till the year 2080 (Figure 4) and will be completely "burned" in the closed cycle of fast reactors towards the end of the 21st century. It has been shown that, given some 3% of MA in the starting fuel charge as loaded, about 40% of MA will burn up over the first fuel irradiation time.

In 2002, RDIPE and RRC-KI have continued scenarios studies on future nuclear power development. Some cases are under investigation, which, in addition to the above, envisage introduction of thermal reactors with the Th-U fuel cycle after the year 2050.

Radiochemical technologies

The initial round of R&D to support development of U-Pu-MA fuel reprocessing methods for the new generation of fast reactors, including transmutation of minor actinides, was carried out in 1999-2001. The development was based on the following assumptions:

- Uranium and plutonium are inseparable at all stages of the process (technological support of the non-proliferation regime).
- Neptunium and americium are not removed from fuel (uranium + plutonium) during reprocessing, or – if removed – are taken back for recycling; these elements may also be added to the regenerated fuel.
- In reprocessing, it is advisable to extract separate fractions of Cm, Cs and Sr for subsequent cooling and of I and Tc, for subsequent transmutation.
- The disposed waste must have no more than 0.1% of U, Pu, Am and Cm; 100% of the remaining actinides (Th, Pa, Bk, Cf); 1% of Cs, Sr, Tc and I.

At the initial stage, the researchers explored the feasibility of meeting the above requirements with the use of the following procedures:

- Upgraded Purex process (Bochvar Institute).
- Electrolysis of molten chlorides with reduction of actinides to metals or to nitrides – LINEX process (Bochvar Institute, RIAR).
- Metallurgical process, with nitrides kept intact at all stages of reprocessing (Bochvar Institute and collaborators).
- Pyrochemistry (ion exchange reactions) in molten fluorides and chlorides (Bochvar Institute, RRC-KI, IHTE).
- Dry fluoride volatility process (VNIICT, RRC-KI).
- Re-crystallisation in molten molybdates and phosphates (Bochvar Institute), etc.

Basic flow sheets have been prepared and equipment components identified. Laboratory experiments have been conducted to validate the key features of process design. The capital and operational costs have been estimated and the feasibility of technological options has been assessed. No procedure examined has shown fundamental difficulties for commercialisation with a production rate of 25-50 t of irradiated fuel per year; and the main requirements (see above) can be met under nominal conditions of the process. It is also possible to fulfil the Radwaste partitioning requirements by combining various methods. The difference in the costs of fuel processing by the examined methods does not exceed ±10%. According to economic estimations, the cost of a plant for fuel reprocessing and refabrication will make no more than 15% of the cost of the nuclear power plant with two BREST-1200 units (2x1 200 MWe) to be catered for by this plant. Studies are in progress on non-aqueous methods, including ion-exchange reactions and electrolysis processes. Work on dry volatility process and advanced Purex processes have been suspended due to its relatively easy transformation for the plutonium extraction.

In the recent years, RIAR has been engaged in studies (DOVITA Programme) on fuel manufacture technology, in manufacture of mock-up fuel rods with mixed $(U,Np)O_2$, $(U,Pu,Np)O_2$ and $(U,Pu,Am)O_2$ fuel and their irradiation in BOR-60 to a burn up of 13-20% as well as in post-irradiation examination of these fuel rods. These efforts are in fact the first practical attempts at MA transmutation; it provide experience of handling fuel with considerable MA amount (up to 5%) and demonstrate the effect of minor actinides on the irradiated fuel qualities.

Along with its industrial applications, this effort affords verification of the computation procedures involved in MA transmutation analyses.

Optimisation of MA and LFP transmutation procedures and modes

Computational studies on modes of MA and LFP transmutation in fast reactors have been performed at RDIPE and IPPE for the last 3 years, with focus placed on nuclear safety issues. Currently, for various reasons, transmutation of minor actinides is considered as small additions (no more than 3% by mass) to the core fuel (homogeneous approach). In this case, there is no need for setting up special production of targets for the separated MA fractions. Heterogeneous transmutation of minor actinides (as separate fuel assemblies) is also under investigation. Transmutation of LFP will take place in the fast reactor blanket, while the neutron spectrum will have to be made considerably "softer".

Nuclear data for MA and LFP

Prior to analysis of actinide transmutation in fast reactors and before validating the technology of the closed nuclear fuel cycle, the evaluated neutron cross-sections were revised for the following isotopes: ^{237}Np, ^{241}Am, ^{242}Am, ^{243}Am, ^{243}Cm and ^{244}Cm (IPPE). Complete files of recommended neutron cross-sections were compiled for the BROND-3 library, with refined fission, inelastic-scatter and neutron-capture cross-sections, fission neutron yields, gamma quantum generation cross-sections, as well as covariance matrices of errors in fission and radiation capture cross-sections. Neutron cross-section files are being prepared for curium isotopes and some long-lived fission products and this work will be continued in 2003.

For the purpose of testing the MA-related neutron data libraries, the first draft input decks have been prepared, which represent the results of earlier domestic experiments on MA irradiation (IPPE, ITEP, VNIIEF, RDIPE). Computational modelling of those experiments (RDIPE) with the use of neutron cross-sections from the libraries ENDF/B-VI, JENDL-3.2 and BROND-3, suggests the following conclusions:

- For the neutron spectrum in the core of a large fast reactor, the difference between the experimental and calculated fission rates of ^{237}Np, ^{241}Am and ^{243}Am – critical isotopes for analysis of a long-term radio toxicity balance – does not exceed 11% irrespective of the nuclear data library used; no measurements for curium isotopes were made in those experiments.
- For systems with harder or softer neutron spectra, the difference between the experimental and calculated fission rates for curium isotopes may reach 30%; better agreement is observed for americium isotopes and ^{237}Np, with greater discrepancies found in a system with a hard spectrum.

A round of experiments is planned to conduct irradiation of small fission chambers and thin MA foils in facilities with different neutron spectra.

MA neutron data libraries are being verified at IPPE using the results of previous experiments in BN-350 where separated actinide isotopes were subjected to irradiation. Similar irradiation work has been done in BOR-60 and is still going on. The experimental data analysis is expected to yield results in 2003.

Alternative approaches to MA and LFP transmutation

As mentioned above, fast reactors are capable of effecting transmutation of the minor actinides accumulated in SNF of thermal reactors and produced in the course of their own operation. On the other hand, computational studies show that under certain circumstances (increase in the pre-reprocessing average spent fuel cooling times to 50 years and more, higher burn up of thermal reactor fuels, etc.) there may appear problems with transmutation of minor actinides inside starting charges of fast reactors. In such a case, various approaches are possible including provision of dedicated reactors for transmutation of long-lived nuclides, e.g.:

- Sub critical blankets with a target driven by accelerator;
- dedicated blankets of thermonuclear reactors;
- critical reactors with liquid fuel (molten-salts and metals);
- critical reactors of traditional design with solid fuel.

Russian Institutes working through ISTC projects or under direct contracts with foreign collaborators are performing extensive experimental and calculation studies on alternative approaches to transmutation, its fuel cycles and associated technological issues. Recent years have seen completion of about 30 ISTC Projects dealing with the transmutation concepts, nuclear data and technologies. Among the efforts in progress or just starting at present, it may be worthwhile to mention ISTC Projects #1486 (RRC-KI, VNIIEF, RIAR; US and Japanese funding), #1786 (RRC-KI, US funding), #1606 (VNIITF, RRC-KI, IHTE, VNIICT; European funding), and #2267 (JINR, European funding).

Project#1486 titled "Experimental and theoretical justification of the cascade scheme of the sub critical MSR for transmutation of long-lived radwaste of the nuclear fuel cycle" is under way now. Project duration: 2 years, started March 2001. The goal of this project is to develop the optimal reactor flow sheet for the cascade sub critical MSR. The main experimental effort is placed on simulation of cascade neutronic effects on cold molten-salt assemblies fuelled by uranium and metallic neptunium.

Project#1786 (with the implementation period between March, 2002 and February, 2004) is aimed on development of a fusion reactor blanket and shielding concept and optimising it from the viewpoint of actinide transmutation, on consideration of the basic design features and determining the integral reactor parameters. It is required to determine the transmutation rate, the total thermal power of supported light-water fission reactors (LWR) and the efficiency of reducing the total mass and long-term biological hazard of high-level waste by bringing in a thermonuclear facility specially developed for transmutation.

Project#2267 (early 2002 through November of the same year) is focused on developing and licensing a prototype sub critical electronuclear facility – SAD. The facility will consist of a lead or lead-bismuth target, a blanket with U-Pu fuel, a lead reflector, shielding, and experimental channels. The target is to be exposed to a proton flux with the energy of 660 MeV and power of 0.5 kW, the thermal power of the blanket is set at ~20 kW, and $K_{eff} \approx 0.95$. SAD is planned to construct at JINR (Dubna) for performing research in physics and technology to support the electronuclear method for transmutation of Pu, MA and LFP.

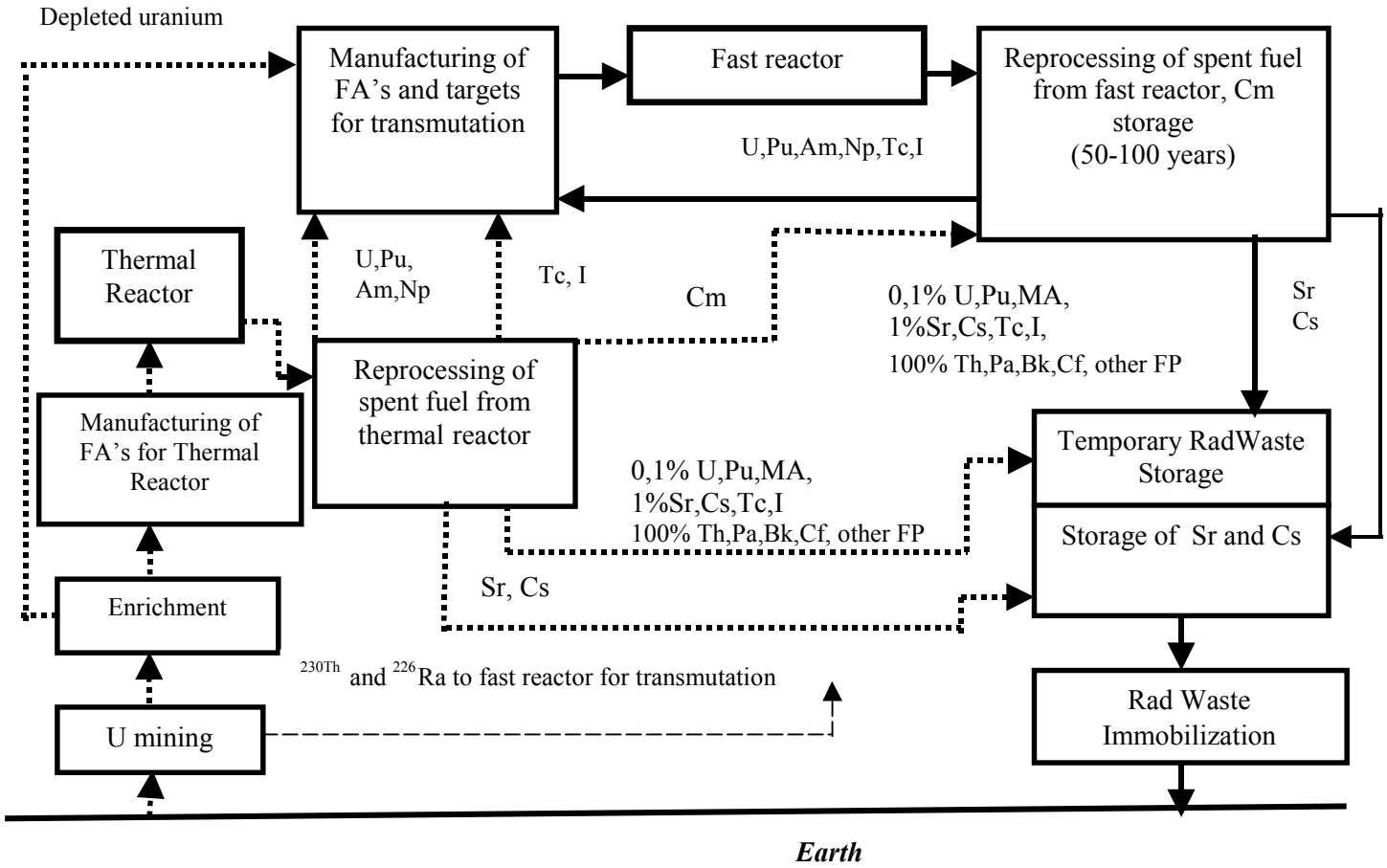
Finally, studies on integral evaluation of Molten-salt Reactor technology potential as applied to commercial long lived actinides transmutation are underway within Project #1606 titled "Experimental study of molten-salt technology for safe, low waste and proliferation resistant treatment

of plutonium and minor actinides in accelerator-driven and critical systems". Project#1606 is being carried out in close co-operation with EU MOST project. The major developments expected as result of project #1606 are the following: reactor physics & fuel cycle considerations; experimental study of fuel salt key physical and chemical properties; corrosion studies in natural convection loop. The first experimental data, are already obtained in terms of the project for the selected Na,Li,Be/F salt system, which include system phase behaviour, solubility of plutonium trifluorides/oxides and transport properties. The construction of corrosion loop made from Ni-based alloy for studies with PuF_3 and Redox control in Na,Li,Be/F system is under way now.

REFERENCES

[1] Ministry of Russian Federation for Atomic Energy (2000), *Strategy of Nuclear Power Development in Russia in the First Half of the 21st Century,* Summary, Moscow.

Figure 1. **Preliminary flow sheet for transmutation nuclear fuel cycle**



Radio toxicity of used uranium = Radio toxicity of disposed RAW

Figure 2. **Mass content of fissile Pu and minor actinides (Np+Am+Cm) in the spent fuel of thermal reactors in the adopted case of Nuclear Power Development (VVER-1000, RBMK-1000 and VVER-440) without SNF reprocessing**

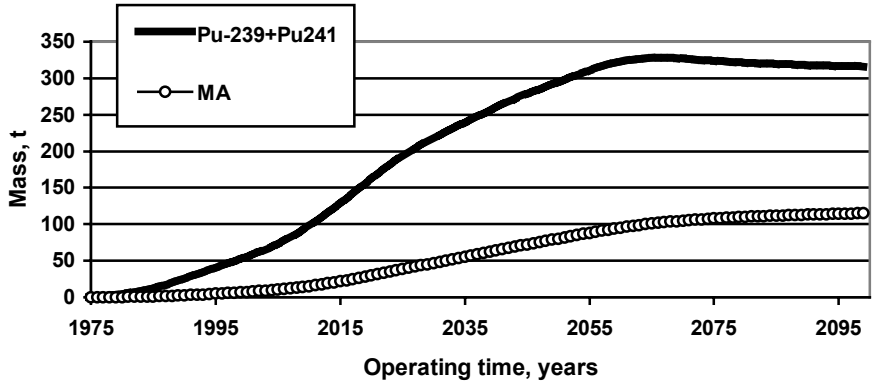


Figure 3. **Changes in the installed capacity of NPPs with thermal and fast reactors**

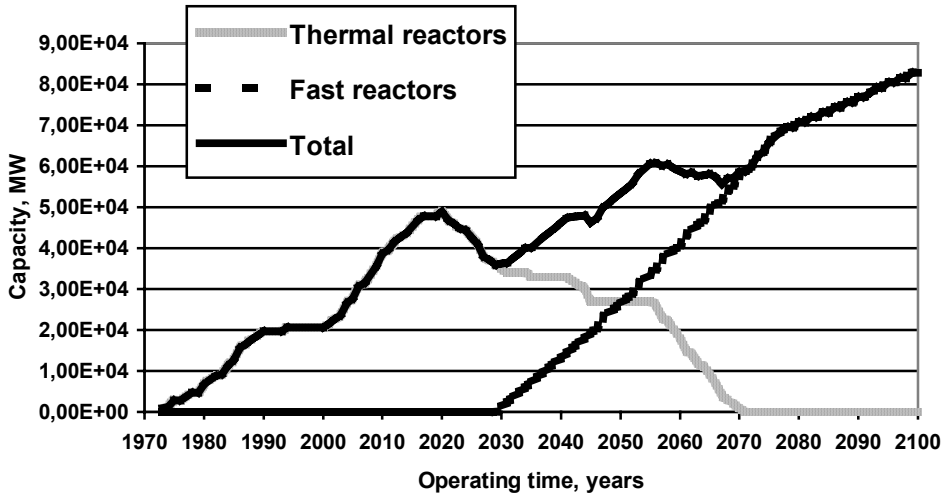


Figure 4. **Balance of Pu and minor actinides (MA=Np+Am+Cm) in the spent fuel of thermal reactors**