DEVELOPMENT OF NUCLEAR FUEL CYCLE TECHNOLOGIES – BASES OF LONG-TERM PROVISION OF FUEL AND ENVIRONMENTAL SAFETY OF NUCLEAR POWER.

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ABSTRACT

To-day nuclear power is one of the options, however, to-morrow it may become the main source of the energy, thus, providing for the stable economic development for the long time to come.

The availability of the large-scale nuclear power in the foreseeable future is governed by not only the safe operation of nuclear power plants (NPP) but also by the environmentally safe management of spent nuclear fuel, radioactive waste conditioning and long-term storage.

More emphasis is to be placed to the closing of the fuel cycle in view of substantial quantities of spent nuclear fuel arisings.

The once-through fuel cycle that is cost effective at the moment cannot be considered to be environmentally safe even for the middle term since the substantial build-up of spent nuclear fuel containing thousands of tons Pu will require the resolution of the safe management problem in the nearest future and is absolutely unjustified in terms of moral ethics as a transfer of the responsibility to future generations.

The minimization of radioactive waste arisings and its radioactivity is only feasible with the closed fuel cycle put into practice and some actinides and long-lived fission radionuclides burnt out.

The key issues in providing the environmentally safe fuel cycle are efficient processes of producing fuel for NPP, radionuclide afterburning included, a long-term spent nuclear fuel storage and reprocessing as well as radioactive waste management.

The paper deals with the problems inherent in producing fuel for NPP with a view for the closed fuel cycle.

Also discussed are options of the fuel cycle, its effectiveness and environmental safety with improvements in technologies of spent nuclear fuel reprocessing and long-lived radionuclide partitioning.

INTRODUCTION.

Energy generation has been and remains the basis of human existence. When evaluating various options of future evolution we have to be sure of the availability of reliable power sources and nuclear power is one of them. The21^{-st} century is starting in the name of the "growth limits". Primarily this is a global action on the human environment, its contamination and changes in the life quality. CO_2 releases and warming may not be beyond the scope of variations in the geologic history of the earth, however, this is not an argument in favour of repeating the fate of dinosaurs.

The state of the art of nuclear power is determined by the more rigid environmental requirements and competition with organic generators of energy9;. Political and social problems of nuclear power are results of putting into practice specific goals and their inherent technologies. The cost of nuclear kW/h is affected by the need to surmount environment related problems that are in many respects proceed from the heritage of the past, namely, the defence program implementation and the specific thinking in the area of peaceful atom. There is a significant risk of nuclear weapon proliferation and nuclear terrorism.

What are the requirements to be met by large-scale nuclear power as a basis of economic developments? These are safety of nuclear power plants (NPP); environmental safety in managing spent nuclear fuel (SNF) and radioactive waste (RAW) as well as non-proliferation of nuclear weapons. To achieve the modern goals and resolve global problems one needs to have novel technologies. Stable economic evolution may be only promoted by predicting the results of technologic innovations and developments of nuclear technologies.

The vector of the modern industrial development is the recycling of resources. Nuclear power involving materials that remain toxic during a long period of time has to provide, first, the recycle of those resources and, second, their minimal build-up (inventory) in the fuel cycle.

The currently cost-effective open fuel cycle cannot be considered to be environmentally safe even for the mid term since the build-up of substantial amounts of SNF containing thousand of tons Pu requires the near-term resolution of the issue pertaining to its safe management (1). The transfer of the responsibility to future generations is totally unjustifiable in terms of morals and ethics.

To minimize the build-up of SNF and RAW as well as their activity is feasible if the closed fuel cycle is put into practice with the attendant burning of actinides and some long-lived fission products.

The key issues of assuring the environmental safety of the fuel cycle comprise efficient technologies of

- production of fuel compositions for NPP including radionuclide afterburning;
- long-term storage and reprocessing of SNF;
- RAW management.

PROBLEMS INHERENT IN NPP FUEL PRODUCTION AS APPLIED TO CLOSED FUEL CYCLE.

The nuclear power in Russia is based on the VVER and RBMK reactors. The BN reactors are primarily experimental ones. In Russia the built up of SNF amounts to ~ 20000 t (against ~ 220 thousand tons in the world). By 2010 in Russia it will reach > 30000 t.

Currently RBMK spent fuel reprocessing has been acknowledged to be inexpedient and the concept of its long-term supervised storage is being put into practice.

The closing of the NFC comprises recycling major and minor actinides as well as higher toxicity fission products. The closing of the NFC in terms of uranium is ensured by VVER-440 SNF reprocessing at the RT-1 plant and producing molten uranyl nitrate hexahydrate containing 2.4 – 2.6 % ^{235}U to be further used for the fabrication of secondary fuel for RBMK. The RT-1 reconstruction will enable the reprocessing of VVER-1000 SNF as well as alien PWR and BWR SNF (3).

With this aim in view at A.A. Bochvar VNIINM under development are processes of producing oxide, nitride, cermet and other types of fuel to recycle U, Pu, MA into available and now under development reactors (VVER, BN, BREST and others).

The urgency of the fissionable material recycle proceeds from a reduction in the available resources of uranium in Russia and the higher cost of uranium mined in Russia compared to the world prices.

One of the important problems inherent in the fuel cycle closing is recycling U into the VVER reactors. The ^{235}U concentration of VVER SNF is 1.2-1.3%. Uranium recycling involves problems of providing recovered fuel of the needed condition (uranium-236 compensation and uranium-232 neutralization). The accumulation of ^{232}U with an increase of NF burn-up, the fabrication of MOX fuel using civil Pu issue a challenge of arranging a remote production of fuel from recycled products. The fabrication of MOX fuel from U and Pu product managed at a reprocessing plant enables the minimization of process stocks of fissionable materials and production of oxide powders of high quality homogeneous U-Pu solutions. The processes of weapon's grade Pu conversion to MOX fuel are a priority direction of developments providing for the utilization of the valuable material and the non-proliferation conditions.

PROVISION OF FUEL CYCLE EFFICIENCY AND ENVIRONMENTAL SAFETY VIA IMPROVED TECHNOLOGY OF SNF REPROCESSING AND LONG-LIVED RADIONUCLIDE PARTITIONING.

The higher efficiency and environmental safety of SNF reprocessing is achieved through the minimization of all types of RAW arisings and emissions of radioactive substances to the environment as well as through creation of inherently safe processes.

Isolation of fragmented VVER SNF from a structural material will optimize the subsequent reprocessing of SNF. This operation is also important for SNF reprocessing by "dry" methods. In future structural material processing and recycling will minimize the build-up of RAW of this class.

Development of equipment for chopping spent fuel assemblies of VVER-1000 will enable the reprocessing of alien spent fuel assemblies. Defective SNF can be reprocessed via chopping defective spent fuel assemblies in cans.

Application of continuously operated dissolver will increase the throughput by more than a factor of 3 and reduce capital and operating expenditures for gas clean-up during dissolution.

Optimization of the existent technology at the expense of RT-1 updating will substantially improve the NSF reprocessing parameters to acquire the following results:

- 1/3 reduction of the number of extraction stages and a respective decrease of Pu inventory in technological cycle;
- reduced application of salt forming agents (bivalent iron, sodium carbonate etc.) which will result in ~ 250 kg reduction of vitrified waste per 1 t of reprocessed SNF;
- 30% decrease of extractant consumption per each ton of reprocessed SNF at the respective reduction of organic RAW.

With the burn-up extended to ≥ 50 GW day/t the accumulation of ^{232}U makes the existent parameters of separation of U and Pu from each other and fission products (10^7-10^8) excessive.

In this case a single-cycle Purex process will make it feasible to co-strip U and Pu at the ratio specified for the MOX fuel refabrication and to reduce substantially intermediate and low activity waste arisings. As a reprocessing option for high burn-up SNF of VVER a reprocessing cycle may be proposed that comprises

- U and Pu co-extraction from HNO3 solutions;
- Pu and in part U stripping at specified U:Pu mole ratio (MOX fuel production for thermal or fast reactors).
- Stripping the main portion of U, its conditioning for further utilization.

Should the need arise the process may also contemplate the extraction of neptunium. The improved quality of SNF reprocessing products and safety of extraction process are achieved by the application of triisoamyl phosphate (TIAP) – an extractant that compared to TBP is an order of magnitude less soluble in HNO_3 solutions and has a much higher capacity for Pu which eliminates the formation of the second organic phase at essentially all concentrations of Pu (2).

Salt-free reagents and electrochemical methods substantially reduce the arisings of HALW, IALW and LALW. The reduced amount of IALW and concentration of radionuclides will lower down the expenses for vitrification that ensures the reliable immobilization of radionuclides.

The practical implementation of minor actinide and other radionuclide partitioning depends on the development of a respective transmutation process, the usability of some isotopes in national economy as well as improvement of environmental safety of HALW disposal. The operations of ^{129}I, ^{3}H, ^{14}C and ^{99}Tc partitioning can be incorporated into the main technology of SNF reprocessing. With a view of immobilizing ^{129}I – one of fission products having the longest life and ^{3}H it is advisable to subject SNF to heat treatment. The oxidative treatment of sheared fuel within 300-500^{0}C allows an essentially quantitative removal of those radionuclides from fuel and their immobilization for supervised storage (^{3}H) or transmutation (^{129}I).

For the removal of minor actinides and residual quantities of other transuranium elements the classic process using organophosphorous acid has been upgraded. The benefits of extractant of this class are high factors of transplutonium and rare earth element separation.
Lower heat and radiation loads on a vitrified materials, fractional HALW solidification in accordance with physicochemical and geochemical properties make the disposal of HALW more reliable. Aside from cesium and strontium it is advisable to remove noble fission products, primarily, palladium and rhodium. Noble metal fission products in sum are removed by sedimentation with reagents that combine the properties of strong reductants and "soft" ligands.
The commercial directivity of SNF reprocessing calls for the minimization of operations not involved in the production of fuel. The build-up of non-utilizable civil Pu and ^{237}Np at the RT-1 plant raises the reprocessing cost and requires extra funds to be allocated for safe storage. However, in the long-term the environmentally safe nuclear power is feasible via burning most toxic radionuclides and achieving the radiation-migration balance between HALW and nuclides recovered from uranium ores (4).
The scope of RAW is to a substantial extent determined by the total capacity of reprocessing plants, namely, the area of industrial premises, communication length, number of equipment items etc. The application of dry processes enables a drastic reduction in RAW scopes, however, the practical use of those processes for the large-scale commercial reprocessing of RAW arising at NPP involves serious problems. Eventually only the gas-fluoride technology allows an effective recycle of U, however, the competitiveness of the particular method with the available aqueous reprocessing technology is problematic.
Of promise is the combination of various options of pyrochemical and aqueous methods of SNF reprocessing. As it has already been mentioned a number of long-lived fission products can be removed via the heat treatment of SNF prior to its dissolution. The selective leaching of neutron poisons will enable the recycle of oxide SNF and higher effectiveness of using fissionable materials. Of challenge are methods of SNF reprocessing that are based on fluids having an abnormally high penetration and dissolution abilities. The advantage of supercritical extraction is high processing rates and a minimal scope of waste, namely, only fission products weight. One of the unresolved issues of this method is to ensure the radiochemical process safety under high pressure, however, the feasibility of implementing a small-scale process allows designing an explosion safe equipment.
"Dry" processes may form the basis of at a site plant for SNF reprocessing and U recycling as well as of complexes for reprocessing fuel and targets of transmutation reactors and facilities. In this connection at SSC RF VNIINM under consideration are processes of SNF reprocessing in molten salts (fluorides, molybdates etc.) as well as thermal methods of shearing and reprocessing, specifically, as applied to hard to shear cermets. The basic criterion of applicability of any technology must be its cost effectiveness and this needs implementation of R&D including a complex trial of a particular technology and its equipment in pilot facilities using actual products and the subsequent technical and economic comparison.

CONCLUSIONS.

The reprocessing of SNF resolves two main interrelated goals, namely, fissionable material recycle and environmental safety of nuclear power – radiation-migration equivalence of accumulated RAW which is provided by burning fissionable materials and highly toxic radionuclides as well as by the usability of some radionuclides in national economy and by producing safe forms of radionuclides suitable for disposal.
SNF reprocessing combined with secondary fuel refabrication will minimize the fissionable material accumulation in the fuel cycle and increase its cost effectiveness. The reconstruction of the RT-1 plant will increase the involvement of recovered U and Pu into the fuel cycle of VVER and BN reactors, will put the closed nuclear fuel cycle into practice and will offer a challenge to participation in the world market of SNF reprocessing.
Analysing the main trends in the world developments of reprocessing technologies one can draw a conclusion that there are a variety of approaches to the resolution of this issue. The methods under development have technological niches that are determined by specific conditions in countries-designers. Designing alternative methods of SNF reprocessing serves to advance the infrastructure and flexibility of nuclear technologies for resolving global problems facing the humanity.

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