FUEL CYCLE OF BREST-1200 WITH NON-PROLIFERATION OF PLUTONIUM AND EQUIVALENT DISPOSAL OF RADIOACTIVE WASTE

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Abstract

Depletion of hydrocarbon resources and concerns regarding greenhouse gas emissions are expected to lead to increased reliance on nuclear power in the Russian Federation in the 21st Century. Increased reliance on nuclear power for electricity production is best achieved using innovative reactor designs and fuel cycles that are more efficient, inherently safe, proliferation resistant and produce less long lived radioactive waste. The BREST series of reactors is being developed within the Russian Federation to meet these criteria. The current states of development work on the BREST-1200 reactor is described.

1. INTRODUCTION

Significant growth of global fuel and energy demand expected in the 21st century will most likely be accompanied by depletion of cheap hydrocarbons and an increase in the emissions resulting from fossil fuel combustion.

The most realistic solution to the energy problem is offered by large-scale nuclear power (NP) capable of supplying a significant portion of the growing fuel demand. Serious expansion of nuclear sources — by an order of magnitude compared to the current level — can be achieved only using fast reactors in a closed fuel cycle. Large plutonium stockpiles accumulated in the first stage of nuclear power development dictate the use of fast reactors with uranium-plutonium fuel, which have significant advantages over other reactor types and the thorium-uranium cycle.

The geography and scale of energy supply anticipated in the next century, impose new requirements on nuclear reactors and closed fuel cycle technology, in particular:

- Full Pu reproduction in the core with breeding ratio (BR) ~1. The slowdown in the expected rate of capacity growth and large amounts of plutonium accumulated in the first stage of nuclear power development, eliminate the need for quick doubling of plutonium, which allows the use of reactors with BR~1 and moderate power density in the core;
- Natural safety of reactors with deterministic exclusion of the most dangerous accidents such as prompt runaway, loss of coolant, fire, steam and hydrogen explosions, which lead to fuel failure and catastrophic release of radioactivity;
- Lower radiation risk from radwaste (RW) owing to the transmutation of the most hazardous long lived actinides and fission products (FP) in reactors and thorough treatment of RW to remove these elements, with provision of a balance between the activity of RW put to final disposal and that of uranium extracted from earth;
- Facilities of a closed fuel cycle should not be suitable for Pu extraction from spent fuel for the purpose of its further use for weapons production; fuel should be physically protected against thefts (non-proliferation);
- Fast reactors should be cheaper than existing light water reactors (LWRs), to make them competitive with fossil fuels and gas in most countries and regions.

Needless to say, technical measures alone cannot prevent proliferation of nuclear weapons because there is always a loophole for illegal use of the now fully matured technologies of uranium enrichment and Pu separation from the spent fuel of existing nuclear power plants (NPPs), which is

stored for a long time in cooling ponds. The danger can be eliminated only through consolidation of the international nonproliferation policy and associated safeguards.

In this context, the new nuclear technology should not open new possibilities for production of weapons-grade materials. Furthermore, it should be totally unsuitable for such applications and in this respect, nuclear power development around fast reactors, with a properly tailored fuel cycle, will provide conditions for gradual reduction of the proliferation risk.

Fast reactors do not need enriched uranium, which will allow abandoning enrichment with time. The first cores of fast reactors will be fabricated using existing stockpiles of Pu and spent fuel that will be removed from their current storages and sent for reprocessing with recovery of plutonium. This initial recovery of Pu and fabrication of the first cores for fast reactors should be carried out at existing facilities in nuclear countries or in international nuclear centers.

With time, this door into proliferation will be closed also because Pu currently residing in storage and in spent fuel stored in cooling ponds, will be gradually moved to fast reactors and their fuel facilities, which offer higher proliferation resistance.

The Research and Development Institute of Power Engineering (RDIPE) has been working in the last decade on a concept of a fast lead-cooled reactor with UN-PuN fuel (BREST series), which relies on considerable domestic expertise in fast reactors and marine nuclear systems with PbBi coolant. The studies carried out so far show that these reactors can satisfy all of the above requirements. The reactor survives any credible accident without fuel failure, has full internal Pu reproduction in the core (CBR~1), does not use uranium blankets and transmutes minor actinides (MA) as a part of the main fuel. These features make it possible to simplify reprocessing technology to a not too deep fuel purification from fission products, with Pu extraction from spent fuel neither required nor possible. Fuel reprocessing should preferably be set up on NPP sites in order to avoid large shipments of highly radioactive and fissionable materials.

BREST has several physical traits, which make it proliferation-resistant:

- Transmuted actinides present in the fuel and rough fuel cleaning from FPs (so that 1% to 10% of them remain in the fuel) facilitate fuel protection against theft at all stages of the fuel cycle.
- With full Pu reproduction in the core (CBR~1) there is no need to use uranium blankets, which precludes production of weapon-grade plutonium in these reactors and eliminates the need for Pu extraction.
- With CBR~1, the fact that spent fuel composition is very close to that of fresh fuel, implies that Pu is neither extracted nor added to the fuel. To adjust fuel composition, another portion of ^{238}U is added into the main fuel to compensate for the burnup of this component.
- With small reactivity margin in the core, it is not possible to load into reactor fuel assemblies containing source material for Pu production. Small reactivity worth of fissile actinides (FAs), its insignificant variation with burnup (CBR~1) and moderate power density in the core, afford quasi continuous on-load refuelling during low-load operation. With closed fuel cycle facilities arranged on NPP sites, it becomes possible to eliminate out-of-pile storage for spent and fresh fuel, which are most vulnerable to theft.

On-site fuel facilities eliminate the need for long-distance shipments of fuel and hence remove the danger of accidents and theft associated with them.

Surplus neutrons produced in a chain reaction in a fast reactor without a uranium blanket and the high flux of fast neutrons, allow efficient transmutation of not only all actinides in the core but also long-lived fission products (I, Te) in the lead blanket by leakage neutrons without detriment to the inherent safety of the reactor.

The radioactivity balance between natural uranium used for energy production in a closed system and resultant long-lived high-level waste (LLHLW) can be attained based on the transmutation of actinides and long-lived fission products in BREST reactors, extraction and utilization of Sr and Cs, with HLW put in monitored storage for about 200 years before final disposal in order to lower its activity approximately a thousand-fold. It is expected that in this fuel cycle concept the waste will contain a small portion of uranium, plutonium, americium and curium, strontium, cesium, technetium and iodine, and 100% of all other actinides and fission products.

2. TECHNOLOGICAL ASPECTS OF THE NONPROLIFIRATION OF FISSIONABLE MATERIALS AND EQUIVALENT DISPOSAL OF RADIOACTIVE WASTE

To ascertain that BREST fuel satisfies the non-proliferation requirement, calculations were performed on the critical mass of a "bare ball" containing fuel composition without reflector. The BREST fuel thus calculated was compared with the critical mass of metallic uranium enriched to 20% with ^{235}U (828 kg), which is authorised by IAEA for circulation and is classified as Class 4 Hazard, i.e. not dangerous as regards the possibility of its use for nuclear weapons production. "Bare" critical mass of fuel composition in a BREST reactor with an equilibrium core (i.e. containing uranium, plutonium, neptunium, americium and curium isotopes) amounts to 850 kg in case of metallic fuel and 1530 kg with nitride fuel. This means that BREST fuel is unsuitable for nuclear weapons production, provided actinides are not separated from it during reprocessing.

The physical traits of fast reactors allow reprocessing in which 1% to 10% of fission products remain in the fuel. Also left in the fuel for transmutation are Am, Np and some Cu. Altogether, these impurities account for the high radiation level of the fuel (approximately 50 Ci/kg), hence providing its inherent protection against theft.

The existing commercial technology of spent fuel reprocessing based on aqueous extraction and other radiochemical techniques studied now (fluorides, electrochemical refining in molten salts, etc.) are tailored to Pu extraction and hence cannot satisfy the non-proliferation requirements. This was one of the reasons why the R&D in this area were halted in the USA, along with the fast reactor programme. Furthermore, this will also be the major obstacle to large-scale deployment of nuclear power in future. Therefore, it is necessary to improve the existing technology and look for new reprocessing techniques. The new techniques should take advantage of the possibilities opened by reactors of the new generation, and should be proliferation-resistant.

In this context, the main feature required of a reprocessing technology is that it leaves no possibility for Pu separation from uranium anywhere in the process. This means that the two should always go together in a certain ratio. Inseparability of U and Pu should be ensured by the chemical processes and equipment used in reprocessing. Any potential variations in process parameters–temperature, pressure, agents used, etc., should not enable Pu extraction or result in significant increase of Pu content in fuel composition, i.e. the reprocessing technology should be inherently resistant to proliferation.

Radwaste minimization

In addition to U-Pu inseparability, the reprocessing technique should satisfy some other requirements meant to improve the radioactivity balance between the fuel cycle waste and natural uranium used in it. As may be inferred from Fig. 1, plutonium, americium and curium are the most dangerous elements in radioactive waste from the viewpoint of biological impacts. Therefore, they have to be transmuted in a closed fuel cycle, with only a very small proportion of them sent to waste. ^{90}Sr and ^{137}Cs are also separated during fuel reprocessing to be cooled in a monitored storage facility
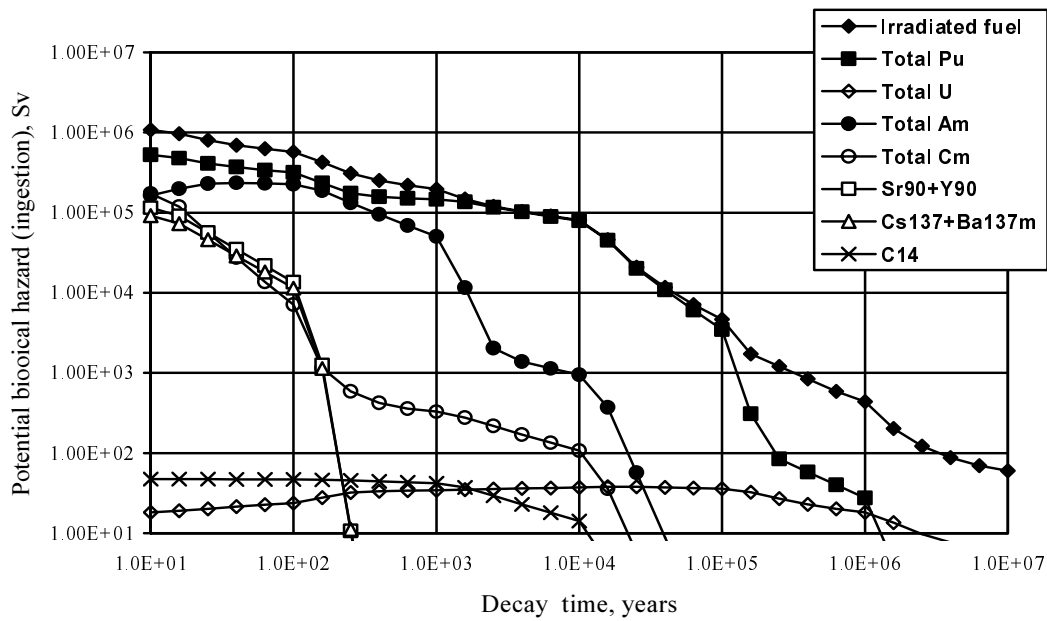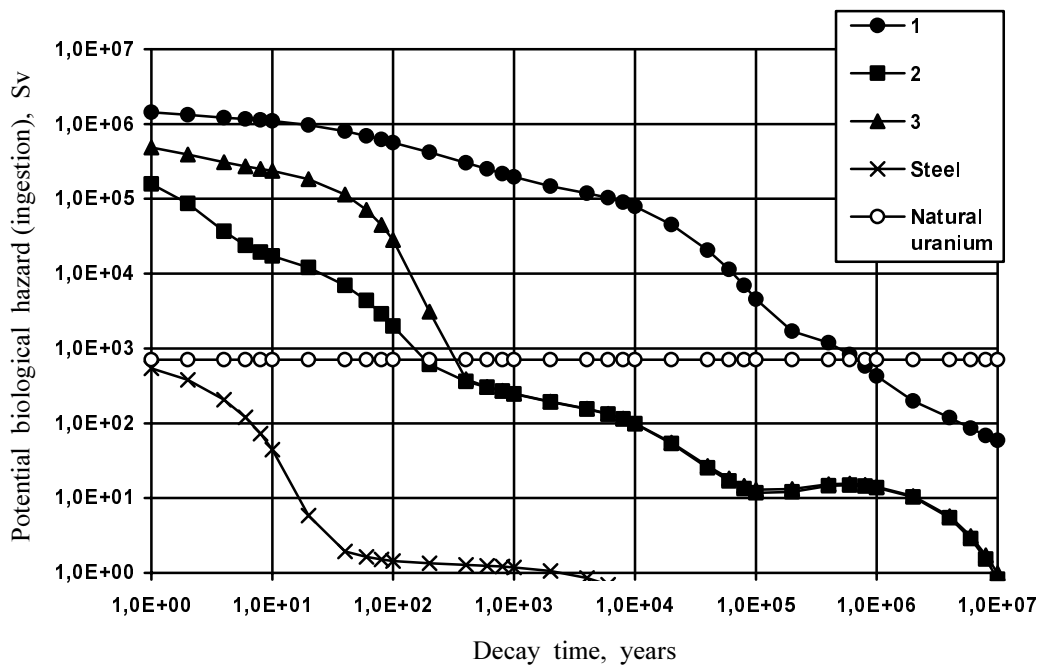
FIG. 1. Potential biological hazard (ingestion) of irradiated fuel from BREST-1200 rated for 1 kg of irradiated actinides (1.06 kg of nitride fuel).

Waste composition:
1 - 1 kg of irradiated fuel + 132 g of steel
2 - 5% (Sr, Cs, Tc, I) + 100% other FP + 0.1% (U, Pu, Am, Cm) +100% (Th, Pa, Np, Bk, Cf) + 132 g of steel EP823
3 - 100% FP + 0.1% (U, Pu, Am, Cm) +100% (Th, Pa, Np, Bk, Cf) + 132 g of steel ЭП823
   ***Steel** - 123 g of stainless steel*
   ***Natural uranium** - 13.7 kg of natural uranium*

FIG. 2. Potential biological hazard (ingestion) of high-level waste from BREST-1200 rated for 1 kg of irradiated actinides (1.06 kg of nitride fuel and 0,132 kg of steel).

which are then passed on for transmutation, storage, or utilisation. The remaining waste contains fission products (with small percentages of cesium, strontium, technetium and iodine) and minor quantities of actinides. This waste will be also kept in monitored storage for 200 years to achieve full decay of short-lived nuclides. After 200 years, the specific activity of the waste will not be over 50 Ci/kg ($\beta\pm$ decays mainly), specific heat release will be about 0.02 W/kg, and the potential biological hazard (PBH) will approach that of natural uranium consumed. Such waste may be diluted to a required level, enclosed in a durable matrix and buried, e.g. in spent uranium mines. Thus the natural radiation balance of the earth can be preserved during the projected long term operation of nuclear power.

For example, the balance between potential biological hazard (PBH) by ingestion of radioactive waste and used natural uranium is shown in Fig.2. In this example waste consists of actinides and fission products from irradiated fuel and irradiated SS cladding of fuel elements. The results are calculated for 1 kg of irradiated actinides (1.06 kg of nitride fuel and 0,132 kg of steel). The PBH of waste is compared with PBH of 13.7 kg of natural uranium. This mass of natural uranium was needed to produce, in a thermal reactor, the Pu in 1 kg of fuel for BREST-1200 first loading. This mass takes into account that the first loading will be recycled 12 times during the reactor lifetime of 60 years. The PBH of natural uranium includes activity of all decay chains of uranium isotopes. Radioactivity balance of the fuel waste and used natural uranium will be achieved after 200 years of waste cooling, if waste contains not more than 0.1% (wt.) of recycled actinides and 5% of Cs, Sr, Tc, I.

Additional (desirable) requirements. Extraction of Np (which may be sent to waste) and Cm (to be stored out-of-pile for 50–70 years, with Pu resulting from Cm decay sent back to the reactor). Np, Cm should be extracted so that 1−10% of them remain in fuel (see Table I).

TABLE I. TENTATIVE REQUIREMENTS FOR REPROCESSING

RW cleaning from actinides	no more than 0.1% left
Fuel cleaning from fission products	1−10% left
Sr and Cs extraction from RW	1−5% left
I and Tc extraction from RW	1−10% left
FA cooling before reprocessing	1 year

Several refining technologies have been appraised against the above requirements: aqueous, molten fluorides, gas fluorides, and electrochemical refining in molten salts. Investigations were also made into the use of unconventional refining techniques such as metallurgical processes or direct annealing of fuel. The research on these and some additional technologies are continuing now. All these techniques were found to be basically capable of assuring inseparability of U and Pu during reprocessing and providing the requisite level of fuel separation from actinides and fission products.

(1) *Aqueous extraction* is practically the sole technique used now in countries with developed nuclear power (Purex process). Basically, it can be modified to suit the non-proliferation requirements. The modified procedure will be simpler than the existing technique because it will not include Pu extraction from organic material. At the same time, the aqueous technique has certain drawbacks in the context of the above requirements:

- Large amount of equipment and rooms, on account of low permissible fuel content in process solutions;
- Large amount of low-level liquid waste;
- The need for long-term preliminary cooling of irradiated fuel to reduce radiation exposure of organic agents and the extent of their decomposition, and also large consumption of these agents;
- Danger of a fire.

These shortcomings justify an extensive effort to develop an improved process, find the optimum mix and size of equipment and perform a cost-benefit analysis for a case when reprocessing is performed on an NPP site.

(2) *Reprocessing based on molten fluorides*. Spent fuel is transformed into a molten salt such as LiF-NaF-UF$_4$ (PuF$_3$), at temperatures of more than 650^0C, which also contains fluorides of FPs and other actinides. Gaseous, volatile and the most noble (electrochemically) elements are isolated in the initial stage of the process. Next, actinides are extracted from the melt in oxidation/reduction and fluorine/oxygen exchange reactions. With actinides having similar electrochemical potentials, it proves possible to operate the process in such a way that it is not feasible to isolate a fraction with high content of Pu.

(3) *A process in which spent fuel is brought in a gaseous state such as UF$_6$*. About half of FP elements produce non-volatile fluorides and hence are separated during fluoridation. The remaining elements can be separated into requisite components by using traps with different temperatures. Uranium and plutonium fluorides are kept together. In principle, superheating of the mixture and thermal decomposition of PuF6 could isolate Pu. There are technical and administrative measures, however, that exclude such superheating in the process being developed.

(4) In the process under consideration, *chlorides of fuel components* (such as UCl$_3$, PuCl$_3$) are *subjected to electrolysis in a potassium and lithium chloride environment*. During anodic dissolution of nitride fuel, noble metals, molybdenum, technetium and zirconium pass into slime. Refining is carried out in electrolyzers with a solid or pool cathode, at temperature of 550^0C–650^0C. As a result of the electrochemical process, uranium, plutonium, the majority of minor actinides and 2% to 10% of rare-earth elements are precipitated on the cathode.

With molten fluoride, gas fluoride and electrochemical molten salt techniques of spent fuel reprocessing it will be possible to reduce the amount of equipment involved in the process and, very likely, also the amount of low-level waste, by an order of magnitude as compared to the aqueous technology. The above techniques have been tested in laboratories but have not been commercialized yet and hence require further analytical and experimental work to support their commercial application. These technologies may involve some increase in the relative content of Pu in the fuel subjected to reprocessing, which is unwelcome from the viewpoint of non-proliferation and nuclear safety. This matter needs further investigation.

(5) A concept has been suggested for *metallurgical refining* of spent fuel. The technology is based on high-temperature extraction of nitride fuel components in liquid gallium with subsequent separation by metallurgical methods. The concept requires many analytical and experimental studies to support it. Obviously, the main problem with this technology will be the choice of construction materials capable of surviving the harsh environment (liquid gallium) and high temperatures (1000^0C–2000^0C).

(6) For the *annealing technique* of fuel refining, only the basic analysis has been performed so far, with some model calculations on nuclide release from oxide and nitride matrices. The calculations relied mostly on analytically evaluated, but not experimentally validated, coefficients of nuclide diffusion in a matrix, which means that this technique also needs considerable experimental and analytical effort to substantiate it. The main attraction of this option is its simplicity (the fuel matrix is refined, heated and kept in this state for some time to allow for nuclide release from grains into the gas environment). Even if this technology fails to live up to the current optimistic predictions about its future, it might become an initial stage of fuel refining in some other baseline technologies. The main difficulty in implementing this technique will be selection of structural materials that can perform at very high temperatures (2000^0C–2400^0C).

(7) The following technologies are also being studied:

- Combined technology with high-temperature annealing and electrolysis in molten fluorides;
- Regeneration based on high-temperature fluorination.

3. SUMMARY

This paper describes the starting premises and the lines of development work dealing with the fuel cycle of the fast reactor BREST-1200 with the desired characteristics. This work is being carried on as part of the Minatom Programme "Fuel cycle of the large scale nuclear energy based on fast reactors with non-proliferation of plutonium and equivalent disposal of radioactive waste" which comprises as its main objectives:

- Investigation of various irradiated fuel regeneration technologies which exclude plutonium separation at all the process stages while ensuring appropriate waste fractionation;
- Study of radiation conditions for different technologies, substantiation of requirements for radwaste fractionation;
- Investigation of nuclear safety in process setups;
- Technical and economic comparison of technologies and choice of one option for further development.

The declared objectives in the fuel cycle under investigation are achieved by the following measures:

- Radiation equivalency in radwaste disposal
  - Waste fractionation,
  - Transmutation of Pu, Am, Cm, ^{99}Tc, ^{129}I,
  - Waste cooling for about 200 years in a monitored storage prior to geological disposal;

- Non-proliferation of plutonium
  - Adoption of a nuclear reactor with CBR~1 and of a fuel regeneration technology excluding plutonium separation at all process stages.

This paper deals only with the initial work stage, with the main results expected in 2000–2001.