FUEL CYCLE OF FAST REACTOR BREST WITH NON-PROLIRATION, TRANSMUTATION OF LONG-LIVED NUCLIDES AND EQUIVALENT DISPOSAL OF RADIOACTIVE WASTE

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ABSTRACT

The declared objectives in the fuel cycle of fast reactor BREST achieved by the following measures. Proliferation resistance of the fuel cycle being developed for BREST reactors is provided along two lines: reactors physics and design features; spent fuel reprocessing technology excluding plutonium separation at all process stages. Surplus neutrons produced in a chain reaction in a fast reactor without 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 lead blanket by leakage neutrons without detriment to the inherent safety of this reactor.

Keywords: nonproliferation, reprocessing, transmutation, actinides, fission products, monitored storage.

1. STARTING PREMISES

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

The most realistic solution to the energy problems is offered by large-scale nuclear power (NP) capable of taking in a significant portion of the growing fuel demand. Serious expansion of nuclear sources - by an order of magnitude against the current level - can be achieved only around 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 serious 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 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 (nonproliferation);

- fast reactors should be cheaper than existing LWRs, to make them competitive with fossils and gas in most countries and regions.

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.

2. TECHNOLOGICAL STEPS TO CONSOLIDATE PLUTONIUM NONPROLIFERATION

Modern civilian nuclear power technologies are the result of conversion of the military nuclear technologies developed in the mid-twentieth century. They contain several elements that may contribute to the risk of nuclear proliferation. These include

- *Separation of uranium isotopes,*
- *Separation of plutonium and/or uranium-233 from spent fuel,*
- *Long-term storage of spent fuel,*
- *Storage of separated plutonium.*

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 onto proliferation will be closed also because Pu currently residing in storages and in spent fuel stored in cooling ponds, will be gradually moved to fast reactors and their fuel facilities, which offer higher proliferation resistance.

To ascertain that BREST fuel satisfies the nonproliferation 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 authorized 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 100-1000 Ci/kg), hence providing its inherent protection against thievery.

Fresh (not irradiated yet) fuel has the following actinide content (% at): ^{235}U - 0.26; ^{238}U - 85.94; ^{237}Np - 0.018; ^{238}Pu - 0.027; ^{239}Pu - 9.12; ^{240} Pu - 3.72; ^{241}Pu - 0.564; ^{242}Pu - 0.236; ^{241}Am - 0.09; ^{243}Am - 0.072. Spent fuel removed from the reactor contains actinides in about the same proportion and also 8.8% (mass) of fission products.

Proliferation resistance of the fuel cycle being developed for BREST reactors is provided along two lines:

- reactors physics and design features;
- radiochemical technique of spent fuel reprocessing.

Moreover, both lines can be implemented in appropriately upgraded (i.e. using nitride fuel and using no blankets, with CBR~1) sodium-cooled fast reactors whose development has been started recently in IPPE.

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 thievery 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 FAs, its insignificant variation with burnup (CBR~1) and moderate power density in the core, afford quasicontinuous on-load refuelling during low-load operation. With closed fuel cycle facilities arranged on NPP sites, it becomes possible to do without out-of-pile storage for spent and fresh fuel, which are most vulnerable to thefts.

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 nonproliferation requirements. Therefore, it is necessary to improve the existing 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.

The main feature required of a reprocessing technology is that it leaves no room for Pu separation from uranium wherever in the process, which means that the two should always go together in a certain ratio. Inseparability of U and Pu should take its root in 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.

The fuel cycle will be placed on NPP sites, which excludes the need for long-distance fuel shipments and removes the associated risk of accidents and thefts. Therefore, the radiochemical plant and related facilities should be compact and cost-effective, with annual nitride fuel production at the level of 20-50 t.

Studies are under way to investigate the possibilities of keeping uranium and plutonium inseparable and of satisfying the requirements concerning fuel purification and waste fractioning (see below) afforded by various radiochemical techniques, such as:

- aqueous, with and without organic extractants;
- molten chloride electrolysis, with actinide reduction into metals or nitrides;
- metallurgical refining, with no nitride breakup in any reprocessing stage;
- in molten fluorides;
- gas fluorination;
- high-temperatrure annealing (as an initial stage of fuel reprocessing);
- electrolysis of molten fluorides;
- recrystallization in molten molybdates and phosphates, etc.

The basic process routes, equipment mix, etc. have already been worked out. The developers are now reviewing the requirements for radiochemical techniques, estimating the costs and investigating the technical feasibility of the project.

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 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.

3. RADWASTE MINIMIZATION

In addition to U-Pu inseparability, the reprocessing technique should satisfy some other requirements meant to improve the radiation balance between the fuel cycle waste and natural uranium used in it.

Surplus neutrons produced in a chain reaction in a fast reactor without 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 lead blanket by leakage neutrons without detriment to the inherent safety of this reactor.

The radiation 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 their activity thousand-fold, approximately. It is assumed in the fuel cycle concept suggested that going to waste are

- 0.1% of uranium, plutonium, americium and curium,
- 100% of the other actinides,
- (1-5)% of cesium, technetium and iodine,
- 100% of all other fission products.

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 for 200 years till they reach full decay. Besides, subject to separation are ^{99}Tc, ^{129}I and neptunium which are then passed on for transmutation, storage, or utilization. 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 (ingestion) of radioactive waste and used natural uranium is shown on fig.2. In this example waste consists of actinides and fission products from irradiated fuel and irradiated SS cladding of fuel elements. The results rated for 1 kg of irradiated actinides (1.06 kg of nitride fuel and 0,132 kg of steel). PBH of waste is compared with PBH of 13.7 kg of natural uranium. This mass of natural uranium was needed to produce in thermal reactor Pu included in 1 kg fuel of BREST-1200 first loading. And this mass takes in to account that the first loading will be recycled 12 times during reactor life time 60 years. The PBH of natural uranium (includes) activity of all decay chains of uranium isotopes. Radiation balance of the fuel waste and used natural uranium will be achieved after 200 years of waste cooling if waste contains not more 0.1% (wt.) of recycling actinides and 5% of Cs, Sr, Tc, I.

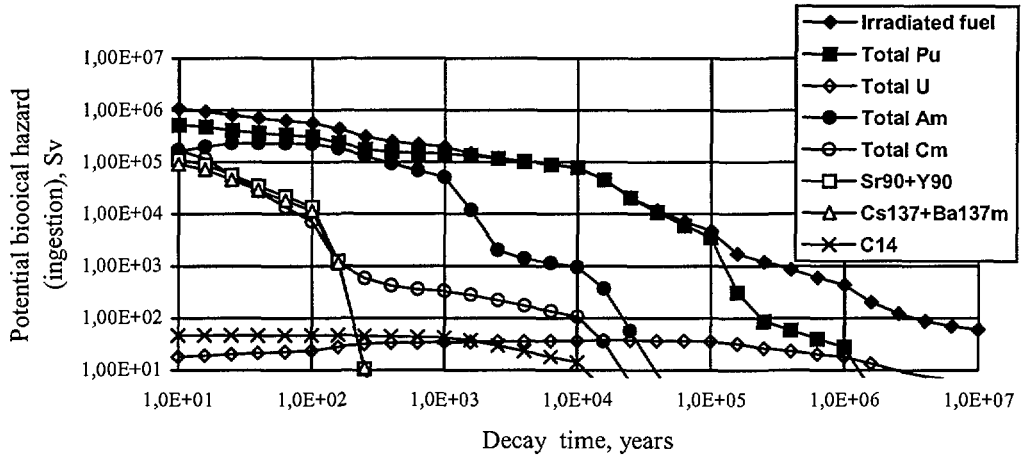

Figure 1: potential biological hazard (ingestion) of irradiated fuel from BREST-1200 rated for 1 kg of irradiated actinides (1.06 kg of nitride fuel).

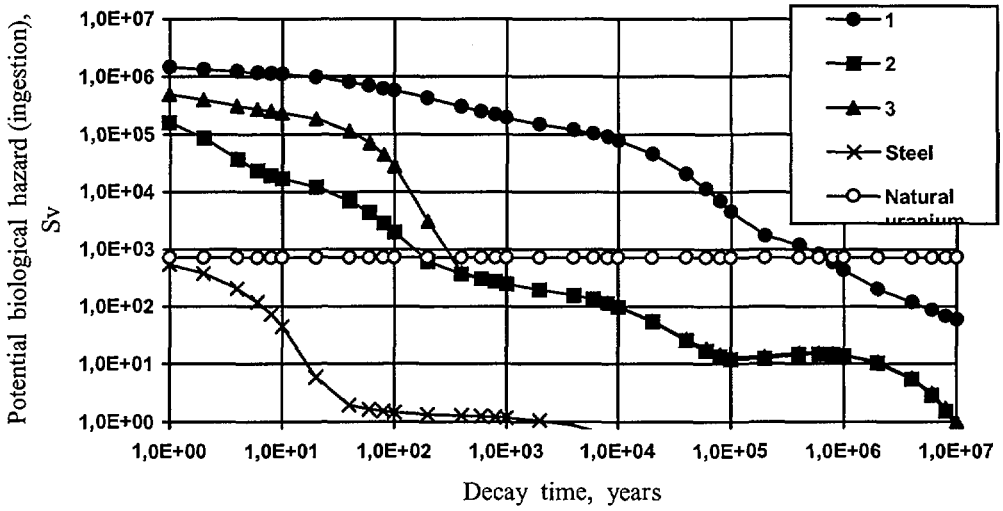

Figure 2 : *potential biological hazard (ingestion) of high-level waste from BREST-1200 rated for 1 kg of irradiated actinides (1.06 kg of nitride fuel and 0,132 kg of steel)*

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

4. CONCLUSION

This paper describes the starting premises and the current lines of development work dealing with the fuel cycle of the fast reactor BREST with the sought-for 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.

Nonproliferation of plutonium:

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