Proliferation-Resistant Fuel Cycle of Advanced Fast Reactors with Radiation-Equivalent Disposal of Radioactive Waste

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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.

2. Technological Support of Nonproliferation Regime

Large-scale deployment of nuclear power brings to the fore the issue of adherence to the Treaty on nonproliferation of nuclear materials usable for production of nuclear weapons. Such are the political realities of the second half of the 20th century and it may hardly be expected that this problem will lose its urgency in the next 30 to 50 years.

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.***

If continuing to use the nuclear technology of today, non-nuclear countries with long-term programmes of nuclear power development will require, sooner or later, their own capabilities for isotope separation and Pu/^{233}U extraction. Should these technologies and capabilities spread worldwide, international control over them would become unwieldy and less than reliable, and the prospects of nuclear disarmament would be questionable. With thousands of tonnes of uranium-235 and plutonium circulating in the global energy sector, it is impossible to keep track of all their uses.

An important task of new nuclear technology is to free civil nuclear power from the need in these dangerous elements. In this case, annual circulation of a large amount of nuclear fuel will not present the risk of proliferation. The safeguarding bodies could focus on the prevention of construction of illegal facilities for uranium enrichment and Pu/^{233}U separation.

This task can be fulfilled by future nuclear power based on breeder reactors which provide full fuel reproduction in the core (core breeding ratio >=1) and do not contain blanket. These can be fast reactors with high-density fuel (U-Pu or Th-U cycle), and thermal reactors with Th-U cycle (molten salts, heavy-water type with small burnup). The choice of the reactor type should be made with due regard for the scale of future nuclear power and available resources. We are assuming large-scale deployment of nuclear sources to 1000 GWe and more worldwide and to 100 - 300 GWe in Russia.

The large amount of Pu already accumulated in the spent fuel of thermal reactors, optimum neutron balance dictate the choice of a fast reactor and uranium-plutonium fuel cycle for large-scale deployment of nuclear power in the current and next centuries. The development of the Th-U fuel cycle will be on the agenda in most countries at the end of this century, when all cheap uranium is used up in thermal reactors (fig.1).

Nuclear power development around fast reactors, with a properly tailored U-Pu fuel cycle, will provide conditions for gradual reduction of the proliferation risk.

Fast reactors have no need of uranium enrichment and civilian nuclear power may well dispense with this technology in due course. Provision of the starting fuel inventories for fast reactors will allow clearing the plutonium stockpiles and utilizing the spent fuel kept in ponds at NPPs, especially as the radiation barrier to illicit uses of spent nuclear fuel declines with time. Initial separation of plutonium and fabrication of the first fast reactor cores should take place at enterprises of nuclear states or at international nuclear technology centers. Transfer of the plutonium kept in stockpiles and contained in spent nuclear fuel to fast reactors and their fuel cycle facilities, affording incomparably better protection, will eventually block this channel of weapons material proliferation.

In fuel cycle of fast reactors with CBR~1 do not need separating of Pu, this technology will not exist in civilian nuclear power with time.

The BREST reactors and BN reactors with nitride fuel are expected to implement the philosophy of the fuel cycle for the fast reactors of the new generation.

The fast lead-cooled reactor with UN-PuN fuel (BREST series) is under developing in the last decade, 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. BN reactor with UN-PuN fuel and CBR~1 is developing too.

Technological support of proliferation resistance in the fuel cycle being developed for BREST reactors and BN reactors with nitride fuel is provided along following lines.

- Transmuted actinides present in the fuel and rough fuel cleaning from FPs (so that 1% to 5% 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 and equilibrium fuel composition, 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.

- 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.
- The reactor design should exclude the loading of the target material for production of the weapons material in the core and reflectors.

In the fuel cycle under consideration, uranium-238 added to fuel during reprocessing "burns up" in the reactor while plutonium is an integral part of fuel and circulates in the closed cycle inside a highly active material.

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.

Owing to the physics specific to fast reactors, it is acceptable to have moderate removal of fission products from fuel during reprocessing (with 1-5 % left behind). Besides, americium, neptunium and some curium remain in fuel and will subsequently undergo transmutation. Together, these impurities set up a high level of radioactivity (~50-500 Ci/kg with 1% of fission products staying in fuel) and afford intrinsic physical (radiation) protection of the fuel against thefts. Reprocessing without plutonium separation amounts basically to fuel cleaning from fission products. Moderate purification -sufficient from the viewpoint of the effect on reactivity - allows a simpler process. Admittedly, this results in an increase of fuel radioactivity, which may lead to certain complications, e.g., at the fabrication stage, but not to any serious difficulties as the process is remotely controlled in any case. Besides, the fuel reprocessing technology should be open to national control over the configuration of the associated buildings and structures (for instance, from satellites).

Thermal reactors running on enriched uranium today are expected to operate for a long time alongside the system of fast reactors in the nuclear energy mix. The irradiated fuel of modern thermal reactors will be reprocessed to provide plutonium and uranium for deployment and makeup of fast neutron facilities. In the long term, as cheap uranium reserves dwindle, thermal reactors may be converted to the uranium-thorium fuel cycle. Uranium-233 for the first cores of these reactors may be produced in fast reactor blankets. To rule out the proliferation hazard, it is possible to exclude the stage of uranium-233 separation during the blanket fuel reprocessing by accumulating uranium-233 to the levels required by thermal reactors.

The main deterrent to large-scale development of nuclear power in the future is the modern industrial method of irradiated fuel reprocessing relying on extraction from aqueous solutions, together with other processes under investigation (gas phase fluorination, electrochemical reprocessing in molten salts) as they were originally meant for plutonium separation and are at variance with the nonproliferation requirements. It is for this reason that such developments were halted in the USA, along with the fast reactor efforts. Therefore it is necessary to improve the existing methods of reactor fuel reprocessing and to look for new techniques that would be tied in with the possibilities offered by reactors of the new generation, would meet the requirements of large-scale energy production and the principle of nonproliferation.

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. 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-temperature 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.

It should be noted that the problem of nonproliferation will not be settled by technical means alone, as there is no eliminating all possibilities for illicit use of the mature technologies for uranium enrichment or plutonium separation from spent fuel kept in storage ponds at NPPs for long periods of time. The only way to avert this danger is to improve the international political regime of nonproliferation and the associated measures of control, protection and enforcement. Such activities may be largely facilitated in the case of a nuclear technology which is free from separation of plutonium (and uranium-233) and does not require uranium enrichment.

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

As may be inferred from Fig. 2, 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.3. 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.

4. Summary

This paper describes the starting premises and the current lines of development work dealing with the fuel cycle of the new generation of fast reactor 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 regeneration technology excluding plutonium separation at all process stages.

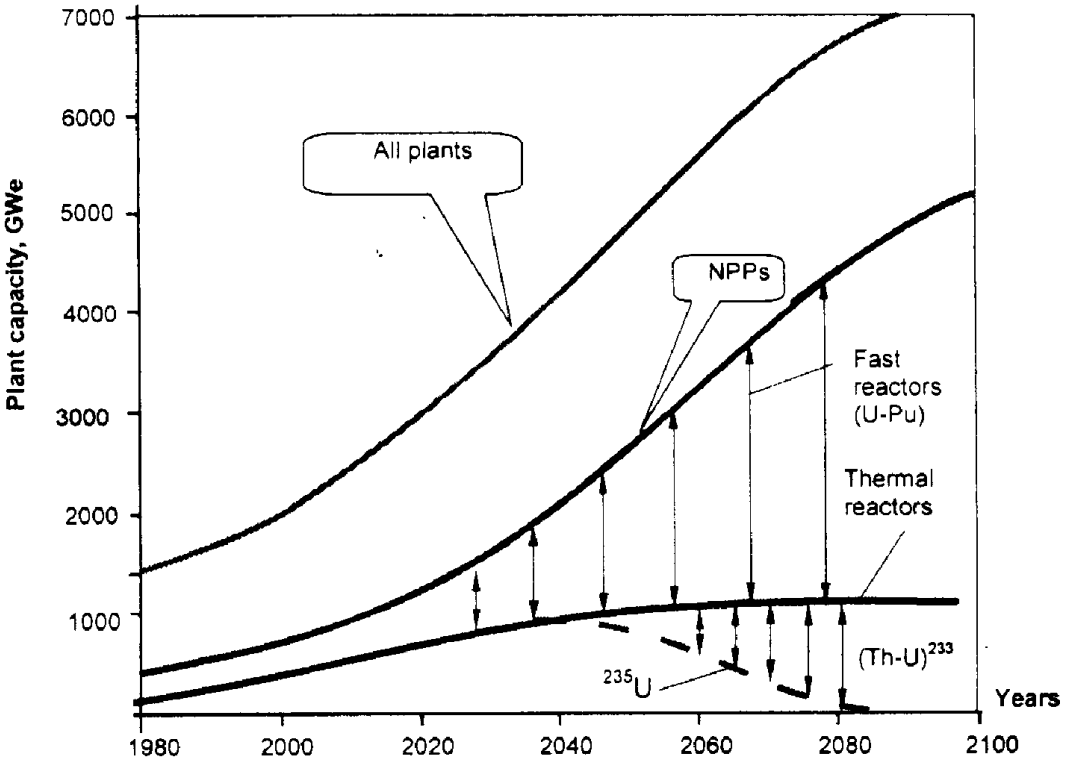

*FIG.1: Tentative scenario of nuclear capacity growth, including fast reactors (given potential cheap U reserves of ~10 mln t). (***) Strategy of nuclear power development in Russia in the first half of the 21st century. Summary. Ministry of the Russian Federation for Atomic Energy. Moscow, 2000.

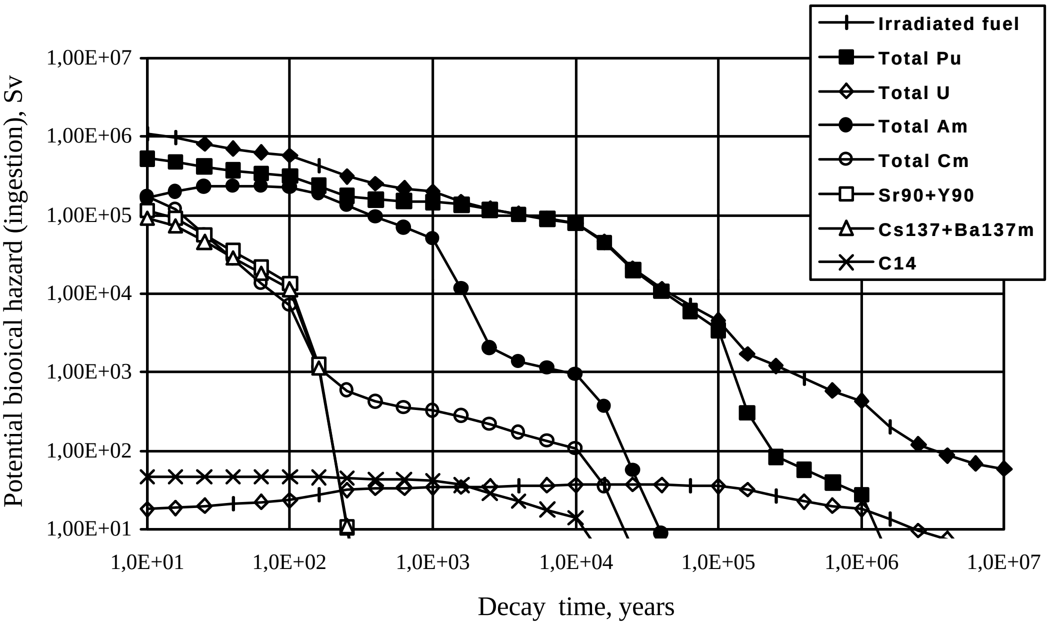

FIG. 2. Potential biological hazard (ingestion) of irradiated fuel from BREST-1200 rated for 1 kg of irradiated actinides (1.06 kg of nitride fuel).

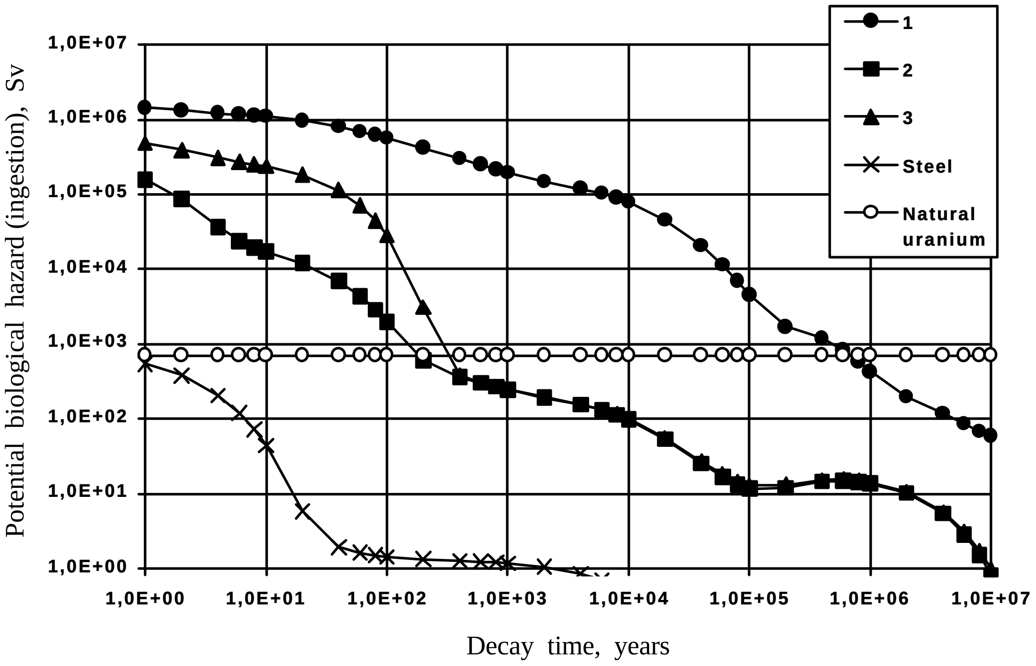

FIG. 3. *Potential biological hazard (ingestion) of high-level waste from BREST-1200 rated for 1 kg of irradiated actinides (1.06 kg of nitride fuel and 0,132 kg of steel)*

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 Ý Ï823*

Steel* - *123 g of stainless steel

Natural uranium - *13.7 kg of natural uranium*