TRANSMUTATION OF LONG-LIVED NUCLIDES IN
THE FUEL CYCLE OF BREST-TYPE REACTORS

A.V. Lopatkin, V.V. Orlov, A.I. Filin
(RDIPE, Moscow, Russia)
1. Background

Radiation background is an integral part of nature around us. Radioactive nuclides, such as uranium isotopes, thorium, potassium-40, carbon-14, etc. occur widely in nature. Much attention has been paid to the consequences of high radiation exposure caused by human activities. However, lower natural radiation background may also turn negative for the mankind. Today, natural radioactive sources (uranium and thorium) are extracted from earth for the needs of nuclear power industry, which in turn is producing a large amount of new radioactive materials, namely, fission products, actinides, and irradiated structural materials. Some of these – actinides and certain fission products – are classified as high-level long-lived waste (HLW) for the purpose of assessing long-term environmental impact of nuclear generation. At present, there is a wide discussion in Russia and elsewhere on various aspects of HLW management, in particular, transmutation. Usually the term “transmutation” implies the following:

- If used in respect of actinides: incineration (fission) in neutron flux, i.e. conversion into fission products whose biological equivalent activity is 3 to 5 orders of magnitude lower than that of the source actinide mixture (Pu, Am, Cm, Np) if cooled for more than 5 000 years.
- If referred to long-lived nuclides from fission products: irradiation in neutron flux, to be converted in a nuclear reaction (mostly, (n,γ)) into a stable or short-lived stable nuclide.

The suggestions concerning transmutation of long-lived nuclides in a flux of charged particles (photons and other) have not been seriously looked into because of the low efficiency of these processes.

To be able to assess the required transmutation scope and choose proper strategy, it is necessary to have quantitative criteria. In most of the foreign and in many domestic studies, HLW transmutation has been treated for long as a task of doing away with the wastes, as completely as possible. If so, the task is hard and extremely costly to accomplish.

A group of authors’ [1] proposed to judge the demanded scope of HLW transmutation against such quantitative criterion as radioactive properties of natural uranium consumed for the purpose of nuclear generation. In this case, HLW should be transmuted until the biological equivalent activity of HLW sent to disposal diminishes to that of feed natural uranium. This means actually that it is suggested that the principle of radiation equivalence between the feed radioactive material and radioactive waste sent to disposal should be taken as a basis for working out the policy of radioactive materials management in the fuel cycle of nuclear power. Such equivalence can be established both at the time of waste disposal and in a historically short reliably predicted time interval (e.g. 200-1 000 years). Radiation hazard from natural uranium is an obvious criterion for comparing the risks from radioactive waste put in disposal. Uranium is a natural source of radioactivity, which remains with biosphere throughout the whole time of its existence. Taking uranium from nature, nuclear power upsets the existing natural balance by lowering it. To return activity to nature, in the amount biologically equivalent to that of natural uranium extracted from earth is to restore the natural radiation balance and hence ensure that the conditions of biosphere existence will not be impaired. In other words, with this concept, nuclear power will be waste-free in terms of radioactivity. Radiation equivalence approach allows reasonable minimisation of HLW weight and activity. Specific conditions of HLW disposal should correspond to local health and other standards and regulations. The most consistent implementation of the radiation equivalence principle will be if HLW are buried in uranium-mining areas worked out using new techniques that would meet environmental requirements.
In the existing open fuel cycle, in which the irradiated nuclear fuel is seen solely as waste, radiation equivalence can be struck only after 100,000 to 500,000 years of spent fuel cooling before disposal, i.e. is practically impossible.

To implement the radiation equivalence principle and transmute HLW, it is necessary to reprocess all spent fuel and separate it into appropriate fractions.

2. Transmutation demand for thermal reactors

Current nuclear power is based on thermal reactors. Let us discuss the long-term variation of potential biological hazard (PBH)\(^1\) of irradiated fuel using the example of VVER-1000 fuel. Similar discussion in respect of fast reactor BREST is given below. After a long storage, radiation characteristics of spent fuel from VVER-1000 show PBH variation in the bulk of the spent fuel accumulated worldwide (PWRs and BWRs). Following a 10-year cooling, the key PBH contributors are strontium-90 and caesium-137 with associated decay products, and plutonium (See Figure 1).

![Figure 1. Potential biological hazard (ingestion) of irradiated fuel from VVER-1000 rated for 1 kg of irradiated actinides](image)

Enrichment 4.4%, burnup 40 MW*day/kg U.
Natural uranium: 9.76 kg of natural uranium.

After 100 years of storage, the key contributors are Am and Pu, and in the period from 1,000 to 100,000 years the main species responsible for PBH, is Pu. Therefore, from the viewpoint of reducing

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\(^{1}\) Potential biological hazard (PBH) is the dose of inner human exposure due to long-term consumption of radioactive materials. Individual biological risk (IBR) of a radioactive nucleus is defined as total exposure dose due to 1 Bq of activity of this nucleus, consumed with food or inhaled continuously in the course of 50 years, with due regard for biological specifics of removal and impact. PBH from a mixture of radioactive nuclei is a sum of individual PBH defined as IBH multiplied by decay rate of a particular nucleus.
the long-term hazard of spent fuel, it is necessary to extract Pu, Am, Cm, Cs and Sr from HLW subject to long-term burial. The extraction and transmutation of Pu and Am are of crucial importance. Other nuclides, considering their relatively short half-lives, may or may not be extracted, depending on the demand from the side of fuel recycling and RW management. Curium whose main isotopes (in terms of mass and activity) – $^{243}\text{Cm}$ and $^{244}\text{Cm}$ have $T_{\text{1/2}} = 29$ and 18 years, respectively, should preferably be cooled in monitored storage facilities until they decay into $^{239}\text{Pu}$ and $^{240}\text{Pu}$ which should be sent back to the fuel cycle for incineration. It is practically impossible to transmute $^{137}\text{Cs}$ and $^{90}\text{Sr}$ on account of low neutron cross-sections, so they can be put in monitored storage for 150 to 200 years till total decay ($T_{\text{1/2}} \sim 30$ years). The efficiency of these steps is illustrated in Figure 2. Two cases have been considered:

1) 0.1% of U, Pu, Am, Cm and Np go in waste subject to disposal (with the bulk of these actinides sent to transmutation or cooling), as well as 100% of other actinides and 2% of Sr, Cs, Tc and I present in spent fuel;

2) The same as in 1), but with 100% of all fission products sent to waste.

![Figure 2. Potential biological hazard (ingestion) of waste from 1 kg irradiated fuel of VVER-1000](image)

Calculations were performed assuming that SF is stored for 30 years prior to reprocessing. It was found that in Case 1 the radiation equivalence between RW and source uranium could be struck in 80 years after SF removal from reactor. In Case 2 the equivalence would be attained in 220 years, following the decay of $^{137}\text{Cs}$ and $^{90}\text{Sr}$. Radiation characteristics of 9.76 kg of natural uranium used for fabrication of 1 kg of uranium fuel with 4.4% enrichment ($^{235}\text{U}$), have been taken as a reference criterion in Figures 1 and 2.

Spent fuel taken annually from VVER-1000, contains approximately 230 kg of “civil”-grade Pu and 23 kg of minor actinides (~68% Np, 22% Am and 10% Cm). If SF is first cooled for a long time (e.g. 30 years) before reprocessing, short-lived FPs will decay completely, $^{137}\text{Cs}$ and $^{90}\text{Sr}$ concentrations will be twice lower, $^{241}\text{Pu}$ will decay into $^{241}\text{Am}$, and the concentration of $^{243}\text{Cm}$ and $^{244}\text{Cm}$ will diminish by a factor of two and more. Plutonium mass in SF will decrease by ~12%, and that of minor actinides (MA) will increase 2.2-fold with the changes in the composition which will
then include 31% of Np, 66% of Am and 3% of Cm. With the likely 30-year average cooling time of VVER-1000 spent fuel before the beginning of its mass reprocessing at the RT-2 plant (planned plant for reprocessing of all VVER-1000 spent fuel which start operating before the beginning start of fast reactor deployment), the values of 202 kg of Pu and 51 kg of MA (or 49.5 kg without Cm) should be taken as more correctly depicting the levels of their annual production in VVER-1000 in the analysis of the demanded capacity of transmutation facilities.

Figure 2 shows the possibility of striking the radiation balance in a thermal reactor system including a transmutation facility. The decision on the type of the transmutation facility is dictated by long-term nuclear policy. At least two approaches are possible:

1) Nuclear power sector is based on thermal reactors and operates until economically acceptable natural reserves of uranium come to an end. After that, nuclear power is phased out. Fast reactors (FR) are not used at a practically meaningful level.

2) Nuclear power is developed based on fast reactors using Pu present in spent fuel of thermal reactors which also remain in operation for a long time, together with FRs.

With the first approach, nuclear energy sources become but a short episode in the history of mankind, with the need to take special measures to get rid of accumulated HLW. Plutonium can be transmuted in thermal reactors operating in a closed fuel cycle, despite the poor present economics of this step. This option will require cardinal changes in “fresh” fuel management strategy and will provide ~20% increase in effective fuel resources of the nuclear power sector based on thermal reactors. Minor actinides can be transmuted at the same time, which will somewhat lower fuel burn-up. Plutonium and MA can be transmuted in dedicated transmutation facilities widely discussed in literature: accelerator-driven and fusion sub-critical installations, thermal and fast reactors with molten fuel, etc. If implemented, this approach will in fact shut the door to rapid development of large-scale nuclear power sector based on fast reactors.

The second approach virtually resolves the issue of resource limitations in nuclear power development because of an ~100-fold increase in the efficiency of U incineration, which affords the use of much more expensive uranium. All Pu accumulated in spent fuel of thermal reactors, will be involved in the fuel cycle of fast reactors, becoming its integral part. Minor actinides can also be efficiently transmuted in fast reactor cores. With lower (or nil) demand for Pu breeding, surplus neutrons can be used to transmute long-lived fission products. Fast reactors with small reproduction of surplus Pu (i.e. more then they need for their own purposes) allow implementing transmutation fuel cycle in large-scale and continuing to grow nuclear power.

3. Transmutation fuel cycle

In reality, radiation equivalence can be attained if a large-scale nuclear power sector is developed based on safe cost-effective fast reactors of a new generation and a closed fuel cycle, along with thermal reactors (advanced VVERs, etc.) fueled with uranium. Fast reactors of a new generation with full Pu reproduction in the core and without breeding blanket will serve not only for energy production but also to incinerate transuranic species and long-lived FPs produced by thermal and fast reactors.
In a transmutation fuel cycle including SF reprocessing with the possibility of waste fractioning, radiation equivalence can be attained provided certain requirements are met. As follows from the earlier studies performed by the authors [2], radiation equivalence is attained if:

- All SF is put in reprocessing.
- U, Pu and Am extracted during reprocessing, are totally incinerated (transmuted) as a result of repeated in-pile irradiation, first of all, in fast reactors.
- Extracted Cm is stored for about 100 years to decay into Pu which is then incinerated in reactors.
- No more than 0.1% of reprocessed U, Am and Cm, 0.01-0.1% of Pu, 1-10% of Np and 1-5% of $^{137}$Cs, $^{90}$Sr, $^{99}$Tc, and $^{129}$I go in waste put in disposal.
- Np is extracted and stored for some time to be used later as source material for production of radioisotope sources on the basis of $^{238}$Pu, or is immediately transmuted as part of FR fuel.
- Cs and Sr are extracted during reprocessing and can be either used as heat/radiation sources or stored in a dedicated storage facility for about 200 years until total decay into stable elements.
- $^{99}$Tc and $^{129}$I extracted during reprocessing, are converted in a stable state (i.e. transmuted) in neutron flux as a result of irradiation in FR blanket.

The key prerequisites for attaining radiation equivalence, are reprocessing of all irradiated fuel with appropriate fractioning, fast reactors used to incinerate the bulk of actinides, and intermediate storage of high-level waste prior to final disposal.

4. Transmutation in fast reactors of BREST type

Transmutation fuel cycle can be built around lead-cooled fast reactors of the BREST type in a closed uranium-plutonium cycle (See Table 1), which have been developed by RDIPE together with other institutes for the last 10 years. BREST reactor development has been carried out proceeding from the following considerations:

- Total Pu reproduction in the core without uranium blankets, with breeding ratio (BR) ~1 and moderate power density.
- Natural (inherent) safety of the reactor, with the most dangerous accidents, such as fast runaway, loss of coolant, fires, steam and hydrogen explosions resulting in fuel failure and catastrophic radioactive releases, excluded deterministically.
- Minimisation of the radiation hazard of radwaste due to in-pile transmutation of the most hazardous long-lived actinides and fission products and thorough radwaste cleaning from the above in order to attain radiation balance between the radwaste put in disposal and uranium extracted from earth (waste-free energy production in terms of radioactivity).
- Impossibility of using closed fuel cycle facilities to extract Pu from irradiated fuel (nonproliferation of nuclear materials).
- Provision of the economic competitiveness of fast reactor plants with modern thermal reactor NPPs (VVER, RBMK, BWR, PWR, CANDU).
Let us discuss the radiation balance of a BREST-1200 reactor. The reactor is assumed to be running in an equilibrium state with U, Pu, Am and Np recycling in the fuel cycle and reactor made-up with natural uranium and Np and Am from three VVER-1000. Plutonium produced in VVERs, goes into the first cores of newly commissioned BREST reactors. Curium from BREST and VVER is stored for about 100 years until $^{243}\text{Cm}$ and $^{244}\text{Cm}$ decay into Pu isotopes. The resultant Pu is returned to the fuel cycle of BREST reactors. The contributions of individual species into PBH of irradiated fuel of BREST reactors are shown in Figure 3. In the time period from 40 to $10^5$ years, PBH of BREST spent fuel is defined by Pu and Am. With the above recycling, when the bulk of U, Pu, Am, Np and Cm are recycled and transmuted and only 0.1% of them goes to waste, as well as 100% of other actinides and 5% of Sr, Cs, Tc and I of those present in spent fuel, radiation equivalence between RW and source natural uranium can be reached after 180 years of storage (See Figure 4). If 100% of Cs and Sr are sent to RW, radiation equivalence can be struck in 350 years. The time needed to reach the balance depends on Cs and Sr decay. PBH of the waste is compared with PBH from 13.7 kg of natural uranium, which is actually the mass of $\text{U}_{\text{nat}}$ needed to produce in a thermal reactor 1 kg of fuel for the first core of BREST reactor. This value was defined taking into account that the first load would be recycled 12 times in the course of reactor lifetime, i.e. 60 years.

Figure 3. Potential biological hazard (ingestion) of irradiated fuel from BREST-1200 rated for 1 kg of irradiated actinides (1.06 kg of nitride fuel)
Figure 4. 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 EP823.
Steel: 123 g of stainless steel.
Natural uranium: 13.7 kg of natural uranium.

The data given in Sections 2 and 4 only illustrate the transmutation fuel cycle and the possibility of reaching radiation equivalence. The full radioactivity balance in a well-developed nuclear power system can be found in [2].

4.1 Some remarks concerning natural uranium consumption

- For the purpose of comparison, Sections 2 and 4 discuss natural uranium consumption typical for the current nuclear power and that of the nearest future (within the 21st century). Thermal reactors operate in an open fuel cycle. Critical loads of BREST reactors of the first generation will be fabricated from reprocessed spent fuel of thermal reactors. However, with the growth and long-term functioning of the fast reactor system, natural uranium consumption will be going down tending to 1 t of U/fission products, i.e. will decrease ~100-fold as compared to the level assumed in Figures 2 and 4. Obviously, this will complicate radiation balance attainment. It will be necessary to provide deeper cleaning of the waste put in disposal from actinides, to resolve carbon-14 issue, by all means, to transmute Np, Tc and I. (There is no need to do this within the next century. It will suffice just to extract and store these materials, which can hardly cause serious difficulties, considering their low specific activity [3]).

- Potential biological hazard from natural uranium has been calculated considering all products of U decay. If long-lived decay products – 231Pa, 230Th and 236Ra are separated from U in the course of mining and returned to the earth or sent to waste, then the value of natural uranium PBH, used in the comparison, should be reduced approximately 10-fold. This also imposes higher demands on RW cleaning from long-lived nuclides.
Higher requirements for the cleaning of RW sent to waste do not undermine the effort to develop an on-site reprocessing cycle for BREST fuel. This technique is suitable for the bulk of the fuel of future reactors. The RW generated by the on-site cycle – and their mass will be less than 10% of that of the recycling fuel - will be in need of additional reprocessing, probably, at some central facility.

Table 1. Main characteristics of BREST-1200 [1]

<table>
<thead>
<tr>
<th>Characteristic</th>
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<tbody>
<tr>
<td>Power, MWe</td>
<td>Fuel charge (U+Pu)N, t</td>
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<tr>
<td>- thermal</td>
<td>Pu/(239Pu+241Pu) charge, t</td>
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<tr>
<td>- electric</td>
<td>Fuel life, years</td>
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<tr>
<td>Core</td>
<td>Time between refuellings, years</td>
</tr>
<tr>
<td>- diameter, mm</td>
<td>Inlet/outlet lead temperature, °C</td>
</tr>
<tr>
<td>- height, mm</td>
<td>Maximum temperature of fuel cladding, °C</td>
</tr>
<tr>
<td>Fuel rod diameter, mm</td>
<td>Maximum lead flow rate, m/s</td>
</tr>
<tr>
<td>Fuel pitch in a square lattice, mm</td>
<td>Steam temperature at SG outlet, MPa</td>
</tr>
<tr>
<td>Fuel</td>
<td>UN+PuN(+MAN)</td>
</tr>
</tbody>
</table>

4.2 Physics of BREST transmutation

- Minor actinides should be transmuted in the core. Transmutation may be homogeneous (i.e. as part of fuel) or heterogeneous (in dedicated fuel rods or fuel assemblies) (See Section 5 for further details). All MA isotopes have their own critical mass without reflector, i.e. they surpass 238U as regards their reproduction properties. Their addition to the core will not increase the critical charge of BREST reactor. All isotopes are efficiently incinerated in the core spectrum. MA transmutation requires neither extra neutron expenditure nor significant extra costs, while contributing to the main reactor objective, i.e. electricity generation.

- Tc and I (probably, other long-lived fission products as well) should preferably be transmuted some distance from the core, in a blanket, with appropriate softening of neutron spectrum. It is quite realistic to spend ~0.5 neutron per one fission in such transmutation. In this, some 250 kg of Tc and I will be transmuted during effective year of irradiation. The realistic rate of transmutation is ~7-10 % annually, i.e. I and Tc loading in the blanket will be 2 500-3 500 kg.

5. Homogeneous and heterogeneous transmutation of Am, Cm, Np in BREST core

This issue is discussed in detail in [4]. Below are summarized the main points of the discussion.

MA addition alters core performance. The level of 5% h.a. has been chosen as acceptable safe MA inventory in the core. Given the critical core charge of 56 t h.m., this will actually mean 2 800 kg, which is equivalent to 126 annual MA yields (Np and Am) in VVER-1000, with 3-year
cooling prior to SF reprocessing and with extraction of MA and Pu, or to 57 annual yields (without Cm) if SF is first cooled for 30 years (See Section 2). In effective operation year, ~100 kg of MA from VVER-1000 will be burned while the remaining amount will be safely “kept” in the closed fuel cycle. It would suffice to take Pu for the critical charge from 37 annual outputs of VVER-1000 spent fuel with 30-year cooling. With such proportion between Pu and MA, there will be no problem with MA disposition at the RT-2 plant. MA-Pu balance shows that the transmutation of MA from thermal reactors will be carried out with their lower concentration in fuel, or only in some of BREST-1200 reactors.

There might be homogeneous (as part of fuel) or heterogeneous (in dedicated fuel rods or fuel assemblies) MA transmutation in the core. In both cases, the transmutation rate in the core centre may run to 8.5% for Np and 10% for Am in effective irradiation year.

Equilibrium MA content in BREST fuel is ~0.7% h.a. With Cm extraction during reprocessing (the presence of Cm raises decay heat release 4-fold and neutron activity in fuel 300-fold) radiation performance of fuel does not differ much from that of equilibrium uranium-plutonium fuel (in the absence of MA recycling), except for gamma dose rate which increases an order of magnitude. MA effect in case of in-pile irradiation is also insignificant. If Am and Cm extracted from VVER fuel, are added to the fuel (keeping to the authorized 5% h.a.), decay power in fuel increases ~5-fold, neutron activity – 1.5 times, gamma dose rate ~100-fold. It appears acceptable in case of remote production, but requires greater attention to fuel cooling at all production steps.

In case of heterogeneous transmutation of the mixture of Am and Cm extracted from VVER-1000 fuel, decay power is about 30 times higher, gamma dose rate is ~50 times greater than those of equilibrium fuel of BREST using its own Np and Am. The initial multiplication properties of (Np+Am)N fuel and of equilibrium BREST fuel are approximately the same, however, burn-up performance differs dramatically. On in-pile irradiation, decay heat in (Np+Am)N fuel rods may be some 1.5-2.5 times greater (depending on fuel composition and density) than that in BREST fuel placed nearby, which is intolerable and calls for addition of inert material. This, however, impairs transmutation efficiency and changes core performance. Loading intervals for Np- and Am-containing FAs are at variance with the standard loading of BREST fuel. “Pure” critical mass of Np+Am mixture is less than 100 kg. Moreover, this circumstance makes heterogeneous transmutation undesirable from nonproliferation point of view as well. Though it is very difficult to make a charge from this material, the very possibility of doing so cannot be excluded.

6. Local radiation equivalence

The developed groundwork for a transmutation fuel cycle allows making fundamental strategic decisions concerning the lines of nuclear power development in Russia. The cornerstones of future nuclear power are reprocessing of all fuel from thermal and fast reactors, transmutation of long-lived nuclides, predominant deployment of fast reactors of the new generation, high-active fuel and relatively “clean” wastes. However, this is but a first step in the development of the scientific basics for creation of environmentally acceptable nuclear power. The next step is substantiation of the areas of final disposal of radioactive waste based on the preservation of local radiation balance and nature. Most consistently, this condition can be met in uranium mining areas. Pioneered in Russia by B.V. Nikipelov, work has been started on developing reference principles for such disposal. Needless to say, this does not mean rejection of existing developments in the area of RW disposal; they just should be reappraise. The key modern principle in the substantiation of the environmental strategy of
RW disposal is multibarrier configuration in case of high-level waste, which is hard to prove for the time of RW storage of $10^4$-$10^6$ years.

The problem of preserving local radiation equivalence is closely connected with uranium mining technique and management of associated long-lived Pa, Th and Ra actinides. Once separated from uranium, they are sent to surface tails, which seriously disturbs local radiation balance, so that the problem of rehabilitation becomes insoluble. It is hardly possible to change what has already been done, but in future, in a sensibly organized nuclear power sector, the approach to U mining should be different. From the viewpoint of contamination and subsequent rehabilitation of the area, uranium mining is no less important than the RW minimisation and disposal technique. Definition and substantiation of requirements for environmentally safe uranium mining needs further investigations as well.

Apart from radiation-equivalent activity, it is necessary to take into consideration heat balance, radionuclide migration as a result of matrix failure, etc. Thus, if migration is taken into account, it is necessary to pay attention not only to Pu, MA, $^{99}$Tc, $^{129}$I, but also to $^{116}$Sn, $^{137}$Cs, $^{79}$Se, $^{231}$Pa, which play insignificant role in the analysis of the total radiation balance of nuclear power.

The line mentioned in the title of the Section, requires comprehensive systematic studies.

7. Conclusions

Transmutation of long-lived nuclides produced as a result of nuclear generation, should be set up proceeding from the principle of reasonable sufficiency, expressed as radiation equivalence between the radwaste sent to disposal and source natural uranium. In this case, introduction of fast reactors of new generation (such as BREST or other reactors based on similar philosophy) will resolve transmutation problems even with the thermal-to-fast reactor capacity ratio of 2:1. The authors of the “Strategy of nuclear power development in Russia” [5] foresee, and substantiate their prediction, that fast reactors of the new generation will account for no less than 2/3 of nuclear capacity in future large-scale nuclear power sector. Fast reactors will be the basis of a transmutation fuel cycle, which will remove the need of creating additional transmutation facilities.
REFERENCES


[4] I. Ganev, A. Lopatkin, V. Orlov, Homogeneous and Heterogeneous Transmutation of Am, Cm, Np in BREST Cores, (To be published soon in Atomnaya Energiya).