**ON THE POSSIBILITY TO CHANGE THE**

**ISOTOPIC COMPOSITION OF PLUTONIUM FROM**

**THE SPENT MOX FUEL OF PWRs IN FAST**

**REACTORS**

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**Abstract**

The purpose of the paper is to investigate the use of fast reactors for changing the isotopic composition of Pu for a better reuse in thermal reactors. The possibility to change or adjust (the term «improve» can also be used) the isotopic composition of Pu from MOX SNF of thermal reactors is determined by the fundamental characteristic of fast reactors – their capability for nuclear breeding, and is directly related to the conversion ratio (CR).

It is obvious that in order to be effective for the whole two-component nuclear power, the change in Pu isotopic composition should not cause shortage of plutonium for fast reactors themselves. Therefore, fast reactors should have a high enough CR as well as fertile blankets. The only current technology to meet these requirements is that of fast sodium cooled reactors.

Single or multiple recycle of plutonium from thermal reactors in the fast reactor fuel, use of uranium-235 or special (target) assemblies with reduced plutonium content are the possible ways to achieve the purpose in the operating BN-800.

The paper examines the possibility of large-scale Pu improvement in commercial fast BN-type reactors with increased conversion ratio. The perspective of an experiment on the BN-800 reactor that would let one demonstrate this possibility in the experimental way is also discussed.

Measurable parameters of the change in the isotopic composition of plutonium in BN-800 are discussed in the paper.

# INTRODUCTION

Today, the world’s nuclear power rests on thermal pressurized water reactors that use uranium fuel. However, natural uranium resources are limited. Besides, after the uranium fuel lifetime is spent in a power reactor, such fuel contains fission products and high-radiation (reactor grade) plutonium.

Leading nuclear power countries (France, Japan) have been trying to solve the problems of Pu accumulation in the SNF (spent nuclear fuel) of light water reactors and saving of natural uranium resources by implementing partial nuclear fuel cycle closure due to transition to mixed uranium-plutonium MOX fuel (mixed oxide fuel) in thermal reactors [1—3]. MOX fuel is fabricated from plutonium extracted from uranium SNF after the required cooling decay time in spent fuel storage pools.

However, practical experience has shown that it is only possible to use such MOX fuel in thermal reactors once because it accumulates heavy Pu isotopes that do not fission in the thermal-neutron spectrum. The content of fissile isotopes in Pu to be used in PWRs (pressurized water reactors) should be at least 61% [4], whereas this content in plutonium from MOX SNF of PWRs is less than 50%.

Recycled Pu from PWRs can be utilized by its irradiation in fast reactors. The possibility to change (or adjust) the isotopic composition of Pu from spent uranium-plutonium fuel of thermal reactors is determined by the capability of fast reactors for nuclear breeding and is directly related to the conversion ratio CR [5]. If the reactor’s CR is more than 1, it is possible to change Pu isotopic composition, if it is less than or equals 1, the change is either ineffective or will require external fuel.

In general, the change in Pu isotopic composition will be effective for the two-component nuclear power if it does not cause shortage of plutonium for fast reactors themselves. To this end, fast reactors should have fertile blankets and a high enough CR. The only current technology to meet these requirements is that of fast sodium cooled reactors.

# WAYS OF CHANGING THE ISOTOPIC COMPOSITION OF PLUTONIUM

Fast reactors, unlike thermal reactors, take plutonium of any isotopic composition. Even isotopes (238Pu, 240Pu, 242Pu) fission in the fast-neutron spectrum, though not so effectively as odd (239Pu, 241Pu) isotopes do [6], their content tends to the so-called «equilibrium». If a fast reactor is loaded with fuel based on plutonium with a high content of even isotopes (unsuitable for a thermal reactor), the isotopic composition of such plutonium will change after irradiation in the fast-neutron spectrum and can become suitable for reuse in light water reactors. Conversely, if low-radiation plutonium is used, the content of even isotopes in it will increase after irradiation in the fast reactor. This idea to control the isotopic composition of plutonium by means of its irradiation in fast reactors (taking the ASTRID reactor as an example) was put forward by French specialists in 2017 [7]. However, there are only two operating sodium cooled fast power reactors in the world now – BN-600 and BN-800, and they are operating in Russia.

Changing the isotopic composition of Pu is important for nuclear power because of the operational activities of supplying fuel to the thermal-fast reactors system. Fast reactors aim at reducing Pu isotopic composition to equilibrium, when it remains unchanged despite Pu multiple recycle through the core. If the isotopic composition of plutonium loaded into a fast reactor contains more threshold isotopes than the equilibrium composition, then its irradiation in a fast reactor will lead to a decrease in the fraction of threshold isotopes, and, accordingly, vice versa.

Analysis has shown that it is production of fissile 239Pu from fertile 238U that makes a decisive contribution to the change in Pu isotopic composition, rather than fissioning of even Pu isotopes. However, within one fuel service lifetime, it is impossible to make Pu suitable for reuse in thermal reactors (achieve the required 61% fraction) as the rate of production of fissile plutonium isotopes in the cores of power fast reactors with MOX fuel is not high enough.

The possible ways to achieve the required content of fissile Pu isotopes in a fast BN800-type reactor [8, 9] are as follows:

* several consecutive recycles of plutonium through the reactor;
* increase in the relative contribution of 239Pu production due to decrease in the fraction of plutonium in the fuel. It can be done by replacing part of plutonium with 235U or by reducing Pu mass fraction in a few core fuel assemblies (special (target) assemblies) and increasing Pu mass fraction in the remaining fuel assemblies to achieve criticality. Let us assess the effectiveness of these ways.

### 2.1. Multiple recycle of Pu through BN-800

The calculations suggested that BN-800 core should be first loaded with fuel based on plutonium extracted from the MOX SNF of PWR [10]. Ignoring the presence of americium in the fuel and decay of 241Pu over lifetime, Pu effectiveness (in relation to the fission process) is assessed for the *j*-th recycle by using the formula [10]:

|  |  |
| --- | --- |
| , | (1) |

where  is the relative content of the *i*-th Pu isotope in the fuel for the *j*-th recycle (corresponds to the isotopic composition of MOX SNF of PWRs); are the mass coefficients of reactivity for corresponding Pu isotopes in BN-800 spectrum.

Mass coefficients of reactivity for Pu, 235U and Am isotopes are given in Table 1.

TABLE 1. MASS COEFFICIENTS OF REACTIVITY OF ISOTOPES IN BN-800 SPECTRUM

|  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- |
| Isotope | 238Pu | 239Pu | 240Pu | 241Pu | 242Pu | 235U | 241Am |
| Value of | 0,686 | 1,000 | 0,229 | 1,347 | 0,101 | 0,78 | -0,013 |

The coefficient  shows the effectiveness of the change in the isotopic composition of Pu over one lifetime. The coefficient calculated by the formula (1) is 1,0195 for the first lifetime.

If the CR over a lifetime is the ratio of the mass of Pu discharged from the core (including Pu from the lower axial blanket) to the mass of loaded Pu, the overall breeding effectiveness of Pu characterized by improved isotopic composition  can be calculated for the j-th recycle as the product of Pu effectiveness  and Pu conversion ratio *CR*. For the first lifetime, the conversion ratio is *CR*1=0,9588 for the core. Then, the overall breeding effectiveness of Pu for the first lifetime is:

|  |  |
| --- | --- |
| , | (2) |

This value indicates that changing the isotopic composition of Pu in BN-800 is not effective. By the start of the second lifetime there is a shortage of plutonium (over 2%), that is why addition of external plutonium is required. The same is true for all the subsequent recycles. The fuel from the core and the material from the lower axial blanket are supposed to be reprocessed together. The contained plutonium is mixed, whereas plutonium produced in the radial blanket is not taken into account. Table 2 shows change in the isotopic composition of plutonium extracted from the MOX SNF of PWR when it is irradiated in BN-800. It can be seen that despite a significant increase in the amount of fissile isotopes (by 3.1%) during the first recycle, the necessary fraction of fissile isotopes (not less than 61%) cannot be achieved after a series of 6 recycles of plutonium through the core of BN-800.

TABLE 2. CHANGE IN THE ISOTOPIC COMPOSITION OF PLUTONIUM IN THE BN-800 CORE, %

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Isotope | Pu loaded into  BN-800, % | Pu discharged from BN-800, % | | |
| After 1 recycle | After 6 recycles | After 6 recycles with Pu added from the radial blanket |
| 238Pu | 4,02 | 3,14 | 0,68 | 0,66 |
| 239Pu | 37,88 | 43,95 | 53,90 | 56,74 |
| 240Pu | 33,48 | 32,80 | 32,13 | 29,43 |
| 241Pu | 11,42 | 8,42 | 5,12 | 4,99 |
| 242Pu | 12,04 | 11,69 | 8,17 | 7,97 |
| 241Am | 1,16 | 0,39\* | 0,24\* | 0,23\* |
| Fissile isotopes | 49,3 | 52,4 | 59,02 | 61,73 |
| Pu mass, kg/year  – core  – lower axial blanket  – radial blanket | 2279,2  –  – | 2080,8  104,5  – | 2136,34  104,9  – | 2072,1  104,9  49,2 |
| –––––––  \*Americium content 1 year after plutonium re-purification | | | | |

The goal can be reached if spent fuel from the core is reprocessed with fuel from the radial blanket. Weapon grade plutonium is produced in the radial blanket (it contains over 90% of 239Pu), and, despite a relatively modest annual production (only 49 kg), the contribution of such plutonium over 6 recycles proves to be enough to have the required fraction of fissile isotopes (over 61%). Besides, with the included plutonium from the radial blanket, the overall effectiveness of the change is  already for the first lifetime, so a small part of plutonium is unnecessary and has to be removed from the reactor fuel cycle.

Although ~2.2 ton of plutonium is recycled through the core per year, the annual output of Pu whose isotopic composition has been changed is not high because of the necessary multiple recycles.

Characteristic features of this way to change Pu isotopic composition are the constant mass of Pu circulating in the fuel cycle and absence of produced weapon grade Pu. On the other hand, its disadvantages are the necessity for multiple recycling of MOX fuel and, therefore, a long duration of such an adjustment cycle. Taking into account all the necessary interruptions (cooling decay time in the in-reactor storage and spent fuel pool, prolonged processes of fuel reprocessing and fabrication), a single recycle like that will take no less than 27 years.

The main conclusion about multiple recycle of Pu through BN-800 is that it is not commercially effective to improve the isotopic composition of plutonium from MOX SNF of PWRs in a reactor whose conversion ratio is low or close to 1 because it takes a long time and requires additional external plutonium. One recycle in a reactor with high CR is far preferable.

### 2.2. Replacing part of plutonium with uranium-235

When MOX fuel based on enriched uranium is used, part of Pu in the core should be replaced with uranium-235. In this case, relative production of 239Pu from fertile 238U increases as compared to its lower initial level, which makes it possible to get improved Pu with the required fraction of fissile isotopes over one lifetime. For this purpose, the mass fraction of Pu in the fuel has to be reduced from ~23% to 8-9%. For the reactor to remain critical, the content of 235U in the fuel is to be 11-17%. An illustration of the composition of such fuel (with BN-800 as an example) is provided in Table 3.

TABLE 3. COMPOSITION OF THE FRESH FUEL WITH ADDED URANIUM-235

|  |  |  |  |
| --- | --- | --- | --- |
| Isotope | Low enrichment zone, % | Moderate enrichment zone, % | High enrichment zone, % |
| 238U | 80 | 77 | 75 |
| 235U | 11 | 14,5 | 17 |
| PU | 9 | 8,5 | 8 |

When such fuel is irradiated in BN-800, production of 239Pu in it scarcely changes, as compared with regular uranium-plutonium fuel, though the amount of loaded plutonium in it decreases by over a half. Correspondingly, the relative fraction of produced 239Pu more than doubles, which makes it possible to get the required fraction of fissile isotopes over one lifetime (in this case the adjustment coefficient calculated by the formula (1) is 1,456). After irradiation, Pu composition is 238Pu / 239Pu / 240Pu / 241Pu / 242Pu = 2,4 / 56,4 / 25,7 / 6,7 / 8,7%. The fraction of fissile isotopes is 63% after irradiation. The annual output of plutonium improved in such a way is ~1250 kg per lifetime, or ~830 kg per year.

A characteristic feature of this improvement method is a considerable gain (by ~30%) in Pu mass.

However, if the effect of consumption of 235U on the adjustment process is included in the formula (2), the integrated coefficient of the change in Pu quality over one lifetime can be calculated as

|  |  |
| --- | --- |
| , | (3) |

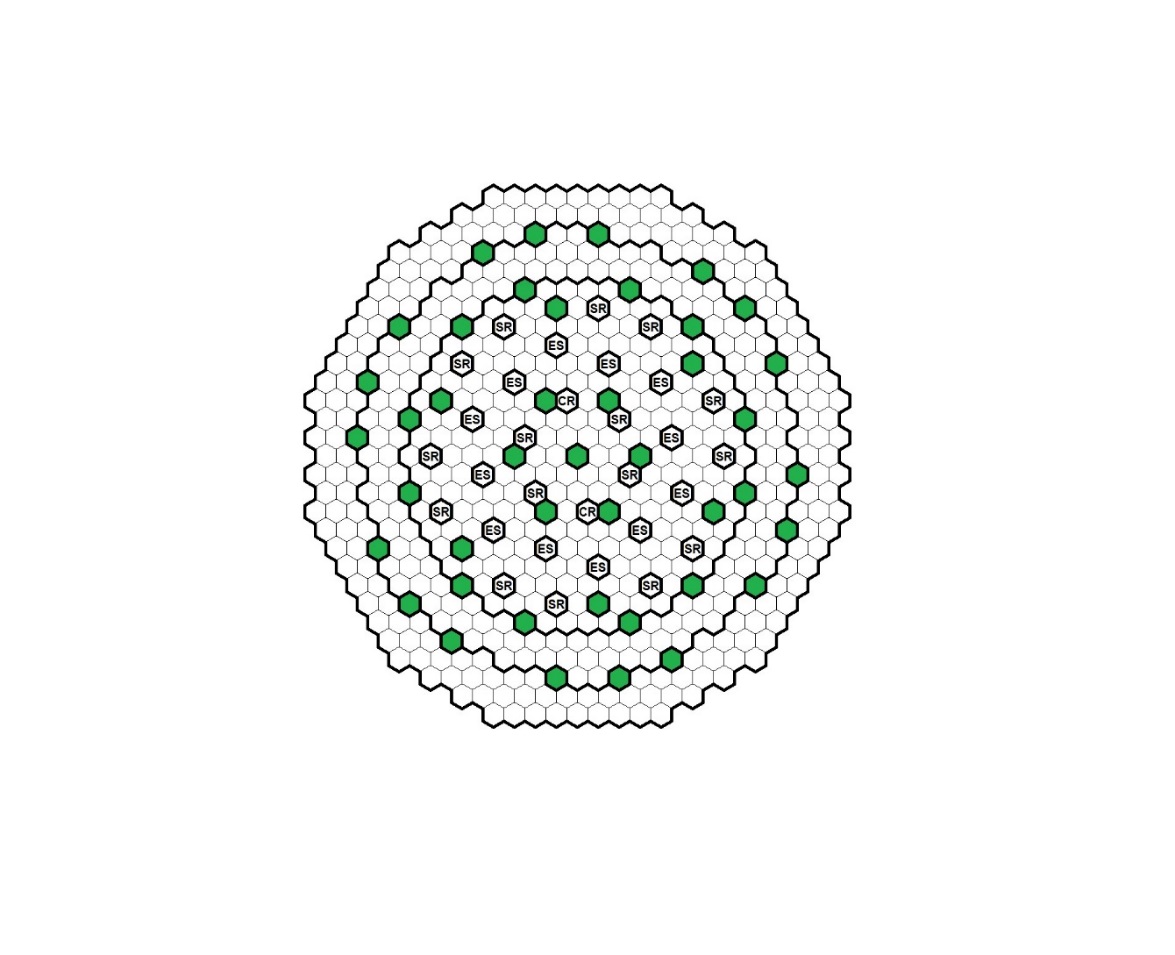
where *М*U is the mass of 235U in the fuel of the fast reactor.

Calculation of the integrated coefficient of the change in Pu quality over one lifetime by the formula (3) shows the main disadvantage of the given method of Pu adjustment - considerable consumption of 235U, which exceeds its saving due to reuse of Pu.

### 2.3. Reduction in Pu mass fraction in a few core fuel assemblies

If this method is employed, Pu isotopic composition is adjusted in a few core FAs (target FAs) rather than in all core FAs. In this case, the reactor criticality is achieved by increasing Pu mass fraction in the remaining FAs that do not take part in the adjustment process. Then, the reactor is only a source of neutrons for the target FAs where Pu isotopic composition is changed.

When target FAs with reduced (to ~10%) Pu mass fraction are placed in the core, the heat generation is 2-2.5 times less in such FAs and correspondingly higher in regular FAs. That is why the number of such FAs is limited. No more than 43 target FAs with reduced (to ~10%) mass fraction of plutonium from MOX SNF of PWRs can be placed in the core of BN-800 so that the design constraints on the maximum heat generation are not broken. An example of arranging the target assemblies in BN-800 is shown in Figure 1.



 – shim rod,  – control rod,  – emergency shutdown rod

FIG.1. *An example of arranging 43 target assemblies to change Pu isotopic composition in BN-800*

The change in the isotopic composition of Pu in the target FAs is shown in Table 4. The required fraction of fissile Pu isotopes is achieved in these assemblies during irradiation over one lifetime. However, because of the limited number of such FAs, the annual output of plutonium is not large and amounts to around 100 kg/year.

TABLE 4. ADJUSTMENT OF PU ISOTOPIC COMPOSITION IN 43 FAs AND BN-800 CORE

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Pu isotopes | Pu composition in the 43 FAs | | Pu composition in the basic loading | |
| Initial composition, % | Composition after irradiation (core+LAB), % | Initial composition, % | Composition after irradiation (core+LAB)\*, % |
| 238Pu | 4,0 | 2,2 | 1,2 | 0,9 |
| 239Pu | 37,9 | 57,0 | 67,4 | 63,1 |
| 240Pu | 33,5 | 25,2 | 23,4 | 27,1 |
| 241Pu | 11,4 | 6,4 | 3,2 | 3,9 |
| 242Pu | 12,0 | 8,4 | 4,6 | 4,6 |
| Fissile fraction, % | 49,3 | 63,4 | 70,6 | 67,1 |
| Mass, kg/year | 73,6 | 98,3 | 1831 | 1773 |
| \*LAB – lower axial blanket | | | | |

This way of changing the isotopic composition of Pu is characterized by a considerable increase in its mass (by ~30%). Its advantage is the excellent opportunity it offers for changing the fraction of fissile isotopes by varying the fraction of initial plutonium.

The adjustment coefficient calculated by the formula (1) for the target FAs only is *K*к=1,518. However, if decline in quality and burnup of Pu in the basic loading are taken into account, the integrated coefficient for the whole core is less than 1:

|  |  |
| --- | --- |
| , | (4) |

It points to the conclusion that improvement in the isotopic composition of Pu in the 43 target FAs is due to the worsening of Pu isotopic composition and increased burnup in the basic reactor loading. Besides, using this adjustment method provides a relatively low annual output of Pu with the changed isotopic composition – less than 100 kg/year. This result is sufficient to demonstrate the potential of plutonium reuse in PWRs, but not for business development.

# CHANGING THE ISOTOPIC COMPOSITION OF PU FROM MOX SNF OF PWRs IN COMMERCIAL FAST BN-TYPE REACTORS

Let us study the previously mentioned ways of changing Pu isotopic composition in the designed commercial high-power fast reactor [8, 9, 11]. This reactor differs from BN-800 in a bigger diameter of the fuel element (9.3 mm versus 6.9 in BN-800), in a much longer fuel lifetime (5 years versus 1.5 years) and in the full use of radiation-resistant materials (cladding steel ЭК164) [12]. Pu fraction in the fuel of such a reactor is smaller by almost a third than in BN-800, so the effectiveness of the change in Pu isotopic composition should be higher in it. Yet, by calculation, neither single nor double irradiation of plutonium from MOX SNF of PWRs in the high-power fast reactor makes it possible to get the desired isotopic composition: the fraction of fissile isotopes is 59,4% even after two recycles. The change in Pu isotopic compositions is shown in Table 5.

TABLE 5. CHANGE IN PU ISOTOPIC COMPOSITION IN A HIGH-POWER FAST REACTOR

(2 RECYCLES). CRITICAL MASS FRACTION OF PU IS 21,06%

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Isotope | Loading the core, kg  1st recycle | Discharge from the core+LAB, kg  1st recycle | Loading the core, kg  2nd recycle | Discharge from the core+LAB, kg  2nd recycle |
| 238Pu | 352 | 217 | 200 | 147 |
| 239Pu | 3321 | 4412 | 4105 | 4723 |
| 240Pu | 2935 | 2698 | 2516 | 2491 |
| 241Pu | 1001 | 562 | 527 | 415 |
| 242Pu | 1056 | 911 | 861 | 735 |
| 241Am | 102 | 161 | 151 | 138 |
| Fuel total, kg | 8767 | 8960 | 8360 | 8650 |
| Fissile isotopes, % | 49,3 | 55,4 | 55,4 | 59,4 |

However, if spent FAs are reprocessed with the FAs of the radial blanket, two recycles will be enough. 109 kg of plutonium is produced in the radial blanket per year, of which 104 kg is 239Pu. In this case, the fraction of fissile isotopes is 56% after the first lifetime and 61.4% after the second.

The adjustment coefficient calculated by the formula (2) is  for the first lifetime, which is much higher than for BN-800, even without including plutonium from the radial blanket.

The overall duration of the adjustment process (with allowance for the cooling decay time in the in-core storage, prolonged processes of fuel reprocessing and fabrication, etc.) is not less than 25 years and the annual output of Pu whose isotopic composition has been changed amounts to ~900 kg per year.

FAs with reduced mass fraction of Pu can be placed in the core of the high-power fast reactor, as it was done in the case of BN-800. Up to 48 target FAs can be placed in this reactor without violating the design constraints. To get the desired fraction of fissile isotopes, Pu mass fraction in the MOX fuel of such FAs should constitute 13-14%, but it must be increased in the remaining regular FAs for the reactor to remain critical.

The annual loading of Pu intended for improvement in the 48 target FAs is about 125 kg. Isotopic compositions of Pu loaded and discharged after adjustment are shown in Table 6.

TABLE 6. ISOTOPIC COMPOSITIONS OF LOADED AND DISCHARGED PU IN A COMMERCIAL HIGH-POWER FAST REACTOR, %

|  |  |  |
| --- | --- | --- |
| Isotope | Loading the core | Discharge from the core+LAB |
| 238Pu | 4,02 | 1,89 |
| 239Pu | 37,88 | 57,52 |
| 240Pu | 33,48 | 26,11 |
| 241Pu | 11,42 | 5,19 |
| 242Pu | 12,04 | 8,02 |
| 241Am | 1,16 | 1,27 |
| Fraction of fissile isotopes | 49,30 | 62,71 |
| Total Pu, kg/year | 125 | 161 |

As in the previous cases, this adjustment method is characterized by a considerable (~30%) gain in the mass of improved plutonium. Its disadvantages are a relatively modest annual improvement and a certain worsening in the isotopic composition of Pu from the basic reactor loading.

# CONCLUSION

Fast reactors with nuclear breeding are the only effective tool to change the isotopic composition of Pu from MOX SNF of PWRs for its reuse in thermal reactors.

The operating BN-800 reactor can be used to demonstrate the possibility of changing the isotopic composition of imported plutonium from MOX SNF of PWRs by multiple recycling of the fuel in the fast reactor, by using uranium-235 or by mounting special (target) assemblies with reduced plutonium content. Each of the methods has its own advantages and disadvantages. However, BN-800 alone will not lead to large-scale export of services on the improvement of imported plutonium. It will only let one demonstrate this possibility in the experimental way, with the use of representative batches of plutonium (up to 100 kg a year), and elaborate all the features of international and national legislation in the field of nuclear material accounting and control. A large-scale project on Pu improvement (with Pu recycle of 1 ton per year or higher) can be planned when a series of commercial high-power fast reactors with an increased conversion ratio are commissioned.

# **REFERENCES**

1. Mixed Oxide (MOX) Fuel. World Nuclear Association, 2017.
2. [Management of Recyclable Fissile and Fertile Materials](http://www.oecdbookshop.org/oecd/display.asp?K=5L4S0FM8MSHD&lang=EN&sf1=seriesidentifier&st1=ser-00671p1&sf2=availabilitycode&st2=50,55,60,80,100,120,140,160,180,200&sp2=and%20not&sf3=versioncode&st3=3&sort=sort_date/d&ds=nuclear%20development&m=4&dc=18&plang=en). OECD/NEA, 2007.
3. Lemasson D., Le Mer J., Garzenne C. Scenario of a symbiotic nuclear fleet composed of PWRs and SFRs. — In: ICONE20, USA, July 30—August 3 2012, p. 269—278. <https://icone20-power2012-54223>.
4. Camarcat N, Settimo D, Dhedin J et al. (2020) Irradiation of Second Generation EDF Plutonium in Russian Fast Reactors using MOX Pellets Fabricated in Marcoule (France). Atomnaya energiya, vol. 128, No. 2, 2020, pp. 73–75.
5. Kagramanyan V.S., Lytkin V.B., Troyanov M.F. Breeding Characteristics of Fast Breeder Reactors and their Determination. Atomnaya energiya, vol. 46, No. 4, 1979, pp.232-236.
6. Shoshina А.N., Burievsky I.V, Poplavskaya E.V. Influence of the Fuel Isotopic Composition on the Main Characteristics of a Fast Reactor. MEPhI International Student Scientific Conference «Polar Lights – 2007». Collected abstracts. Saint Petersburg, 29.01–3.02.2007. [https://polar.mephi.ru/ru/conf/ 2007/pdf/sbornik2007.pdf](https://polar.mephi.ru/ru/conf/%202007/pdf/sbornik2007.pdf).
7. Grouiller P., Coquelet C., Venard C. Plutonium recycling capabilities of ASTRID reactor. IAEA-CN-245-348. 2017. <https://www.researchgate.net/publication/317380638_Plutonium_recycling_capabilities_of_ASTRID_reactor>.
8. NPPs with Sodium-Cooled Fast Reactors. Textbook edited by Shcheklein S.E and Tashlykov O.L. Part 1. Yekaterinburg, Ural Federal University, 2012.
9. Matveev V.I., Khomyakov Y.S. Technical Physics of Sodium Fast Reactors. Textbook for higher educational institutions. Edited by Rachkov V.I. MPEI Publishing House, Moscow, 2012.
10. Grouiller P., Coquelet C., Venard C. Plutonium Recycling Capabilities of ASTRID Reactor. Vienna, IAEA-CN-245-348, 2017.
11. Kuznetsov I.A., Poplavsky V.M. Safety of NPPs with Fast Neutron Reactors. Edited by Rachkov V.I. Izdat Publ., Moscow, 2012.
12. Nikitina А.А., Ageev V.S., Leontyeva-Smirnova M.V. et al. Advances on Structural Materials for Fast Reactor Cores. Atomnaya energiya, vol. 119, No. 5, 2015, pp. 292-300.