Possible ways to solve the problems of using MOX fuel in thermal reactors

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Abstract. Modern nuclear power has a number of crucial problems, including the accumulation of spent nuclear fuel (SNF) and the projected shortage of nuclear resources. The conversion to a closed fuel cycle is the most promising solution to these problems. Mixed uranium-plutonium fuel (MOX fuel) has been successfully tested and commissioned under this concept by several European countries. However, its operation in reactors designed for uranium fuel has a number of problems, most of which have been resolved. Nevertheless, the spent MOX fuel has neutronic characteristics that do not allow its reuse in thermal reactors and solve the problem of SNF accumulation. This is due to the accumulation of heavy (even) plutonium isotopes, which are not fissionable in the thermal spectrum. In fast reactors, it is possible to create conditions under which the isotopic composition of plutonium from the spent MOX fuel from thermal reactors changes in such a way that it becomes suitable for reuse in thermal reactors. This process is called improving. The concept for the development of two-component nuclear energy based on the joint operation of thermal and fast neutron reactors and the creation of a closed fuel cycle was adopted in Russia to implement this process and solve crucial problems of nuclear power.

1. Introduction
In researches carried out at the IAEA within the framework of the INPRO project on the initiative and with the participation of Russian experts, estimates were made of the structure of the world nuclear power under possible scenarios of its development, taking into account the irregularity of regional development, the availability of nuclear resources, technological and infrastructural readiness of some regions for the large-scale development of nuclear power [1]. Analysis of the scenarios for the development of the world nuclear energy suggests that for an open fuel cycle, even with a reasonable scale of development, by the end of the century there will be a shortage of nuclear resources.

The solution to this problem lies in the creation of a nuclear energy system of a closed fuel cycle. Comprehensive researches of the use of energy plutonium are a prerequisite for the transition to such a system. Special attention should be paid to the problems of using plutonium in the fuel of thermal reactors, such as pressurized-water reactor (PWR), which from now until at least the middle of the century will dominate among other types of reactors.

The conversion to a closed fuel cycle and the creation of two-component nuclear power based on the joint operation of thermal and fast reactors is a strategic direction for the development of nuclear power in Russia. The implementation of this concept will make it possible to develop nuclear power of any required scale without restrictions on nuclear resources and emissions into the environment.
2. Return of plutonium to the nuclear fuel cycle, MOX fuel

The world practice of operating power reactors has a successful experience of returning (recycling) plutonium to the nuclear fuel cycle in the form of MOX fuel.

MOX (mixed oxide) fuel is nuclear fuel composed of a mix of uranium and plutonium dioxides. The main reason that the nuclear energy industry today is pursuing the path of using MOX fuel is the goal of disposing of the stockpiles of excess plutonium by burning it in reactors. MOX fuel fabrication involves using depleted or natural uranium, in the form of uranium dioxide, and reactor-grade or weapons-grade plutonium.

MOX fuel proponents thus forward these two arguments: Firstly, we get rid of the excess plutonium; and secondly, we burn uranium that is unusable in thermal reactors, while producing energy as well [2]. However, the use of MOX fuel is associated with many problems due to the significant difference between plutonium isotopes and uranium isotopes in their nuclear and physicochemical properties.

3. Differences between MOX fuel and uranium fuel and problems of using MOX fuel

Differences in physicochemical properties in terms of melting point, heat conductivity, residual energy release and the yield of gaseous fission products (GFP) are not critical in themselves, but can have a dangerous effect on the thermohydraulic parameters of the reactor core under certain operating conditions and in case of accidents.

From a neutron-physical point of view, the main difference between plutonium and uranium as fuel for thermal reactors is that the absolute value of the microscopic absorption and fission cross sections for plutonium in the thermal spectrum is more than 2 times higher than for uranium, which is why the average neutron energy turns out to be higher. This entails a number of features, the main one of which is a decrease in the efficiency of the control and protection system (CPS), boric acid, and burnable absorbers. In addition, the use of MOX fuel leads to an increase in the power peak and a change in some reactivity coefficients. In total, this makes it difficult to control the reactor and worsens its stability.

Table 1. Safety related characteristics of MOX as compared to UO₂ [3].

| Characteristic Item                      | Change from UO₂                      | Effect                                      |
|-----------------------------------------|--------------------------------------|---------------------------------------------|
| **Physical-chemical:**                  |                                      |                                             |
| Melting point                           | Lowers by 20-40 °C                   | Adverse effect                              |
| Heat conductivity                       | Decreases                            | Adverse effect                              |
| Fission gas release                     | Increased release                    | Adverse effect                              |
| Non-gaseous element release             | Possible increase                    | Cesium and some others                      |
| **Nuclear:**                            |                                      |                                             |
| Fission/ absorption cross-section       | Larger; strong resonance above thermal energy | Reduced control rod/ boron worth           |
| Power peaking                           | Increased peak ratio                 | Complicated MOX rod configuration needed    |
| Reactivity coefficient                  | Change of absolute value             |                                             |
| **At low Pu enrichment:**               |                                      |                                             |
| Doppler coefficient                     | More negative                        | More rapid reactivity change in case of transient; reduced reactor shutdown margin |
| Void coefficient                        | More negative (BWR)                  |                                             |
| Moderator temperature coefficient       | More negative (PWR)                  |                                             |
| Fission yield and actinide production   | Increased iodine, tritium, and actinide production | Increased hazard in accident                |
| Decay heat                              | Increased (moderately)               | Negative effect on residual heat control and long-term waste management |
| Delayed neutron fraction                | Reduced fraction                     | Difficulty in reactor control               |
| Prompt neutron                          | Shorter life time                    | Difficulty in reactor control               |
These features lead to a significant difference in the characteristics of the reactor core with MOX fuel as compared to uranium cores. [4]:

- decrease in the efficiency of the CPS and the boron regulation system (without modernization of the reactor, only a part of the uranium fuel loaded into the reactor can be replaced with MOX fuel);
- decrease in the effective fraction of delayed neutrons;
- increase in the unevenness of the distribution of energy release over the fuel element;
- smoother decrease in the multiplying characteristics of fuel assemblies as they burn out;
- increase in residual heat generation and a slower rate of its decrease over time;
- increasing the fluence on the reactor vessel;
- increased gamma and neutron radiation from fresh and irradiated fuel;
- increasing the yield of gaseous fission products.

In addition to the listed issues directly affecting the control, safety and life of the reactor; MOX fuel has a number of common problems that make it much more difficult to use:

- stricter safety requirements at all stages of fuel handling (production, transportation, storage, use);
- the complexity of the manufacture of fuel elements to compensate for the enhanced power peak;
- limiting the duration of plutonium storage during the production of MOX fuel;
- less power generation;
- spent MOX fuel is impractical to reprocess, which does not allow its reuse.

4. MOX fuel experience

The experience of using MOX fuel has more than half a century history. Currently, MOX fuel is operated in more than 40 commercial reactors in Japan, France, Germany, Switzerland, Belgium et al.

The following design solutions are successfully applied at foreign commercial reactors to compensate for the above-mentioned features of MOX fuel, which have a negative effect on the core characteristics [4]:

- use of enriched boron in the CPS and boron regulation system;
- profiling of fuel assemblies with several types of fuel elements with different Pu content;
- introduction of restrictions on the location of MOX-fuel assemblies in the core;
- increase in the free volume under the cladding of a fuel element and/or a decrease in the helium pressure under the cladding.

It is planned to apply similar solutions when introducing MOX fuel at VVER-1000 reactors as part of the program for the disposal of weapons-grade plutonium. It should be noted that due to the peculiarities of the isotopic composition, MOX fuel from weapons-grade plutonium in terms of its neutron-physical characteristics, and, consequently, consumer qualities, occupies an intermediate position between uranium fuel and MOX fuel from energy plutonium.

MOX fuel operates similarly to uranium in terms of operating conditions, the number of fuel cycles and burnup. This option makes it possible to significantly reduce the accumulation of uranium spent nuclear fuel in these countries, but does not completely solve the problem: the problem of storing uranium spent nuclear fuel from light-water reactors in these countries has been converted into the problem of storing MOX-SNF.

5. Russian programs including the introduction of MOX fuel

As part of the program for the disposal of weapons-grade plutonium, Rosatom is conducting research to substantiate the safety and licensing of loading experimental and standard MOX fuel assemblies into VVER-1000 reactors.

The work performed has shown the possibility of ensuring the safety requirements for neutron-physical characteristics and the required rates of plutonium disposal. At the same time, the neutron-physical characteristics of the discharged MOX fuel also does not allow its reusing [5].
The problem of plutonium reuse in thermal reactors is caused by a decrease in the proportion of fissile nuclides. In fast reactors, an increase in the proportion of fissile isotopes is possible, which makes it possible to obtain plutonium with neutron-physical characteristics suitable for further use in thermal reactors. This process is called improving [6].

6. Plutonium improvement in fast reactors

The analysis of the improving process is carried out on analytical models. We will simulate the behavior of isotopes in fuel, which is processed entirely and is not divided into separate, distinct parts.

This work is focused on the problem of improving the composition of plutonium by irradiating plutonium during one fuel assembly run. For this case, the traditional form of the system of equations by isotopic kinetics is suitable (1). For simplicity, we restrict ourselves to the trimmed composition of plutonium from the four isotopes Pu-239, Pu-240, Pu-241, Pu-242 and take into account the presence of U-238.

We will find the effective cross sections for the area of jointly processed materials, taking into account their contribution to the corresponding reaction rates. Irradiation conditions in a fast industrial reactor are sufficiently stable, therefore, the neutron cross sections and flux can be taken constant. We will neglect reactions other than fission and capture. The equations for the rates of change in the number of nuclei in the reactor are:

\[
N_{i}^{t+1} = \phi \left( \sigma_{c}^{i} N_{i}^{t} + \sigma_{f}^{i} N_{i}^{t} \right) \cdot N_{i}^{t} + \sigma_{c}^{i} N_{i}^{t} - \sigma_{f}^{i} N_{i}^{t} - \sigma_{c}^{i} N_{i}^{t} - \sigma_{f}^{i} N_{i}^{t} \]

\[
\text{where } i \text{ is a given isotope number; } N_{i}^{t} \text{ is the number of nuclei of the } i\text{-th isotope; } \phi \text{ is neutron flux.}
\]

The components of the rate of change in the number of nuclei have quite traditional components: \(-\sigma_{c}^{i} N_{i}^{t}\) – the rate of removal of the isotope due to the reaction of capture, \(-\sigma_{f}^{i} N_{i}^{t}\) – rate of removal of the isotope due to the reaction of fission.

Initial conditions are required to obtain a solution. Let us define them at the initial moment of time \(t = 0\). Let's denote them as follows:

\[
N_{238}^{0} = N_{238}^{0} \quad N_{239}^{0} = N_{239}^{0} \quad N_{240}^{0} = N_{240}^{0} \quad N_{241}^{0} = N_{241}^{0} \quad N_{242}^{0} = N_{242}^{0}
\]

It is quite natural that all values of the number of nuclei, cross sections and flux in formulas (1) and (2) are positive in the mathematical sense.

The solution to system (1) with initial conditions (2) can be written in the form of formula (3), the initial version of which was proposed by Bateman [7]:

\[
N_{i} = \sum_{m=1}^{i} \left\{ N_{i-m+1} \prod_{j=1}^{m-1} \sigma_{c}^{j} \left[ \sum_{k=1}^{m} \exp \left( -\sigma_{f}^{k} t \right) \right] \prod_{j=1}^{m} \left( \sigma_{c}^{i} + \sigma_{f}^{i} \right) \right\}
\]

where isotopes are numbered according to the lines in (1):

\(i = 1\) this is U-238,
\[ i=2 \text{ this is Pu-239}, \]
\[ i=3 \text{ this is Pu-240}, \]
\[ i=4 \text{ this is Pu-241}, \]
\[ i=5 \text{ this is Pu-242}, \]
\[ \sigma^k_f \quad \text{cross-section of "removal" of the } k\text{-th isotope } \sigma^k_f + \sigma^k; \]
\[ \sigma^j_c \quad \text{the capture cross section of the } j\text{-th isotope with the formation of the } (j+1)\text{-th isotope; } \]

the product for indices that do not meet the imposed conditions is equal to 1, i.e. \( \prod_{i=3}^{8} f(i) = 1 \)

The change in the fraction of odd isotopes for a specific option is shown in Figure 1.

An analysis of the equations obtained for the number of nuclei shows that it is possible to express the rate of improving \( V_{Dd} \) (change in the fraction of fissile isotopes) is described by the formula:

\[
V_{Dd} = \varphi \left\{ \sum_{i \neq f} u^{238} (1-x) + \frac{\sigma_c^{Pu240} d_{Pu240} - \sum \sigma_c^f d_i}{\sum \sigma_c^f d_i} \left[ 1 - Dd \right] - \left[ \sum \sigma_c^f d_i - \sum_{i=non-f} \sigma_c^f d_i \right] \right\} \]  

\[ x \quad \text{the proportion of plutonium in heavy nuclei; } \]
\[ \sum_{i \neq f} \sigma_c^f d_i \quad \text{capture index on fissile isotopes (conversion to non-fissile isotopes); } \]
\[ \sum_{i \neq f} \sigma_c^f d_i \quad \text{fissile isotope removal index; } \]
\[ \sum_{i=non-f} \sigma_c^f d_i \quad \text{non-fissile isotope removal index. } \]

Stationary fraction of odd isotopes:

\[ Dd_{\infty} = \frac{(\sigma_c^{Pu239} + \sigma_c^{Pu240} a_b^{Pu241} - \sigma_c^{Pu240} a_b^{Pu241} + \sigma_c^{Pu240} (\sigma_c^{Pu241} - a_b^{Pu242}))}{\sigma_c^{Pu242}} \]  

\[ a_b^{Pu242} - a_{b-cf} \]

Equation (5) was obtained under the following condition:

\[ \sigma_c^{Pu242} > \sigma_c^{Pu239} > \sigma_c^{Pu240} > \sigma_c^{Pu240} > a_b^{Pu242} \]  

The maximum initial plutonium content in the fuel \( x_{max} \), below which the improvement process for the current composition of plutonium takes

\[ x_{max} = \left[ 1 + \frac{d_{Pu239}(\sigma_c^{Pu239} - \sigma_c^{Pu239}) - d_{Pu240}(\sigma_c^{Pu240} - \sigma_c^{Pu240}) + d_{Pu241}(\sigma_c^{Pu241} - \sigma_c^{Pu241}) - d_{Pu242}a_b^{Pu242}}{\sigma_c^{Pu239}} \right]^{-1} \]  

\[ d_i \quad \text{nuclear fractions of the } i\text{-th isotope of plutonium in plutonium. } \]

As an example, consider plutonium from MOX-fueled thermal reactors with a power generation of 45 GW / tU. Its isotopic composition: Pu-239/Pu-240/Pu-241/Pu-242/Pu-238=37%/32%/16%/12%/4%. For simplicity, exclude Pu-238. In this composition, the fraction of odd isotopes is 54.6%. We will irradiate the resulting composition in the low enrichment zone BN-800 with an end shield, initial plutonium from a thermal reactor on its own MOX with an energy production of 45 GW / tU [8]

The change in the fraction of fissile plutonium for different initial fractions of plutonium over time is shown in Figure 1.

This and other researches show the possibility of creating a technological platform for a closed fuel cycle based on fast reactors. Such a platform within the framework of two-component nuclear power will solve the problem of reusing of MOX fuel and will significantly improve the situation with the accumulation of spent nuclear fuel. In addition, it will provide an opportunity for expanded reproduction of fuel and implementation of the concept of radiation-equivalent waste management.
Figure 1. Graph of changes in the fraction of fissile isotopes versus time with varying the initial fraction of plutonium in the composition [8]

7. Conclusion
According to experts, a shortage of natural uranium will come in the near future, while the existing systems for the regeneration of fissile materials, based on thermal reactors, cannot solve the problem of either waste or the fuel balance. However, the shortage of commercially available fissile material is not a limiting factor for the sustainability of nuclear power. The solution is to create a nuclear power system that will increase the use of the energy potential of nuclear materials through expanded fuel breeding in fast reactors and improved fuel use characteristics of thermal reactors, as well as closing the fuel cycle for all significant actinides. This approach will provide humanity with the ability to develop nuclear power of any required scale without restrictions on fuel resources and emissions into the environment.

The creation of such a system is a strategic direction for the innovative development of nuclear power in Russia.

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