

Prospects for the Use of MOX-Fuel for New Type Nuclear Reactors.

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Operating experience and scientific and technical studies determine the limitations of production and implementation of MOX fuel for WWER/PWR/BWR reactors for the following main reasons:

- *the high activity of the non-irradiated MOX fuel and the related need for additional safety research when using existing equipment for handling fresh fuel;*
- *due to the high specific activity of plutonium-239, which is several orders of magnitude higher than the activity of uranium-235, it will be necessary to reduce the number of leaky fuel rods by gas leakage and direct contact in order to maintain acceptable reactor water activity at the NPP operation. fuel with water;*
- *loading even a portion of the MOX fuel core reduces the efficiency of the controls (due to high absorption in plutonium, which shifts the absorption balance in the reactor in its favor);*
- *the fraction of delayed neutrons in plutonium is three times lower than in uranium (in plutonium $\beta_{ef} \approx 0,2\%$), this changes the properties of the reactor in power maneuvers to the more dangerous side, etc.*

The melting point of the compound UO_2 - PuO_2 decreases approximately in proportion to the content of PuO_2 from 2840 °C for pure UO_2 to 2390 °C for pure PuO_2 . From these data it can be calculated that the melting point of a typical MOX will be 20 — 40 degrees below the melting point of uranium oxide. With high degrees of burnout, the melting point may still decrease. MOX thermal conductivity also decreases monotonically as plutonium content increases. There are also some differences in physical and mechanical properties (Young's modulus, Poisson's ratio). At high rates of burnout, there is an increase in the yield of gaseous fission products from MOX compared to UO_2 .

In addition, there is a serious problem with dealing with irradiated MOX fuel. Irradiated in light water reactors, this fuel is different from the uranium complex isotope composition of fission products and actinides. Plutonium-containing fuels produce a much larger number of small actinides (americium, curium) due to greater neutron capture in the thermal part of the spectrum; an increase in the proportion of these nuclides results in increased activity and heat emission of the irradiated (U-Pu) O_2 fuel. The processing of such fuel is technologically more complicated than the processing of uranium. Due to the complex nuclide composition, the performance of regenerated 2nd generation plutonium is deteriorating and it will be expedient to use it only for loading in fast neutron reactors.

In the event of a serious accident at the reactor with a violation of the tightness of the core, the dose at a given distance from the reactor in the case of loading it by one third of the MOX fuel will be higher by 2,3 — 2,5 times. The effects of radioactivity will be amplified as many times. The use of MOX is able to exacerbate the negative environmental consequences of the accident by 3,2 — 4 times.

The issue of utilization of MOX fuel in the state of spent nuclear fuel remains urgent. The total amount of plutonium stored in the world at the beginning of the 21st century in all its forms is estimated at 1239 tonnes, of which two-thirds is in the nuclear fuel of the NPP. Already, more than 120,000 tonnes of spent nuclear fuel is in storage, and by 2020 it will be 450,000 tonnes.

A promising direction for the use and utilization of MOX fuels is the creation of reactors based on wave energy release in the core (wave reactors), as well as «fast» neutron reactors. In wave reactors, plutonium acts as an initiator and catalyst for the energy release wave. However, the introduction of wave reactors requires the following problems to be solved, taking into account the specific features of energy generation:

- *additional analysis of nuclear and radiation safety;*
- *determination of the maximum permissible safety levels of plutonium; additional analysis of the conditions of occurrence of neutron-thermal hydrodynamic instability in the core, etc.*

Based on the presented simplified methods of safety analysis, it is determined that the safety conditions for the maximum permissible temperatures of the fuel rod and nuclear fuel shells, as well as the thermal stability of nuclear fuel, are ensured at plutonium-239 concentrations not exceeding 3%.

The calculated analysis determined that the maximum permissible concentration of plutonium-239 for ensuring the conditions of nuclear safety of the wave reactor should also be no more than 3%. It is necessary to further substantiate the sufficiency of such concentration of plutonium-239 to initiate and maintain a stable wave of «burning» of nuclear fuel of wave reactors.

Keywords: MOX-fuel, wave nuclear reactor.

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I. INTRODUCTION

The problem of safe operation, disposal and practical application of plutonium-uranium nuclear fuel (MOX fuel) is one of the priority tasks of world nuclear energy. The particular relevance of solving these problems is determined by the fact that in fact MOX fuel can be the basis for nuclear weapons. Efforts to optimize resource conservation and security in modern world nuclear energy should be part of the concept of non-proliferation of nuclear weapons. Currently, these efforts are aimed at finding effective and safe technologies for reprocessing spent nuclear fuel (SNF), separating out fissile isotopes of uranium and plutonium and involving these nuclides, as well as plutonium-239 and highly enriched uranium-released fissile materials — into the fuel cycle of modern nuclear power reactors [1].

The utilization of weapons-grade highly enriched uranium (enrichment in uranium-235 is not less than 92%) is carried out by obtaining standard reactors from it by diluting depleted or natural uranium.

Particular problems are created by the artificial nuclide plutonium-239, which turned out to be the most suitable element (at a concentration of about 95%) for creating nuclear weapons. Plutonium isotopes are formed in any nuclear reactor that runs on uranium fuel. In order to obtain plutonium-239 (from uranium-238) to create nuclear weapons on its basis, the idea of reprocessing spent nuclear fuel arose and got real realization in the 1940s. Weapon-grade plutonium was obtained by processing irradiated uranium fuel with low burnup, which had been in specially designed reactors for only a few months, after a short cooling time. The approximate isotopic composition of plutonium produced in this mode (95% ^{239}Pu + 5% ^{240}Pu). During the arms race, a significant amount was gained.

At the present stage, in accordance with international agreements, weapons-grade uranium and plutonium extracted during disarmament from deactivated nuclear warheads should become inaccessible for re-involvement in the production of nuclear weapons.

The isotopic composition of plutonium produced in thermal neutron reactors (the so-called civil or energy plutonium) depends on the fuel burnup rate and differs significantly from the isotopic composition of weapons-grade plutonium. In a light-water reactor with an electric capacity of 1000 MW, about 200 kg of plutonium isotopes are formed annually. With deep fuel burnup ($\sim 60 \text{ MW} \cdot \text{day/kg U}$), energy plutonium has approximately the following composition: Pu^{239} 60%, Pu^{240} 25%, Pu^{241} 10%, Pu^{242} 3%, Pu^{238} 2% [2, 3].

The total amount of plutonium stored in the world at the beginning of the 21st century in various forms is estimated at 1239 tons, of which two-thirds are in SNF of the nuclear power plant. Already, more than 120 thousand tons of SNF is in storage, and by 2020 it will be 450 thousand tons. At the same time, 250 tons are weapons-grade plutonium, of which 150 tons belong to Russia, 85 to 95 tons belong to the USA, 7,6 t — Great Britain, 6-7 t — France, 1,7-2,8 t — China, 300-500 kg — Israel, 150-250 kg — India. 200 tons of energy plutonium were also produced: France — 70 tons, the UK — 50, Russia — 30, Japan — 21, Germany — 17, the USA — 14.5, Argentina — 6, India — 1, etc. According to 2001 data, Japan disposed of 48,2 tons of plutonium.

Currently, 430 nuclear reactors are operating in the world, of which about 10 thousand tons of SNF containing 70 tons of plutonium are discharged annually. An estimated 92 tons of weapons-grade plutonium is enough to replace 11040 tons of natural uranium. 252 tons of energy plutonium can replace 30240 tons of natural uranium.

It should be noted that there is a significant limitation on the transition to MOX — MOX fuel can be produced from reactor plutonium, but it cannot be diluted with reactor uranium (i.e., uranium released from spent nuclear fuel in parallel with plutonium). Reactor uranium has an isotopic composition, making it unsuitable for further use. Therefore, in MOX fuel, reactor plutonium is diluted with uranium that has never been in the reactor, which, of course, increases the cost of mixed fuel and does not allow the disposal of post-reactor uranium.

One of the areas in the field of plutonium utilization is the conversion of the alloy to dioxide, which is suitable for the manufacture of tablets of vibro-compacted MOX fuel for VVER-1000 (and others) thermal neutron reactors. In its physicochemical characteristics, MOX is markedly different from uranium fuel. The difference in the melting points of MOX and UO_2 is significant — it is lower for MOX. The melting point of the $\text{UO}_2\text{-PuO}_2$ compound decreases approximately in proportion to the PuO_2 content from 2840 °C for pure UO_2 to 2390 °C for pure PuO_2 . From these data, it can be calculated that the melting point of a typical MOX will be 20 to 40 degrees below the melting point of uranium oxide. At high degrees of burnup, the melting temperature may still drop. The thermal conductivity of MOX also decreases monotonically as the plutonium content increases. There are also some differences in the physicochemical properties (Young's modulus, Poisson's ratio). At high degrees of burnout, an increase in the yield of gaseous fission products from MOX is observed in comparison with UO_2 .

The production of MOX is constrained by environmental problems and difficulties in transporting it over long distances.

Spent fuel from conventional light water reactors contains approximately 1% plutonium. Theoretically, one NPP unit can produce 5-6 tons of plutonium annually. After processing eight spent uranium fuel cells, one MOX fuel cell can be made, which will be accompanied by the formation of a huge amount of radioactive waste.

A promising area of application for MOX fuel is wave nuclear reactors, in which plutonium is used as an initiator and catalyst for a heat release wave in the reactor core. The use of MOX fuel for wave nuclear reactors determines the need to solve the following problems:

- determination of safety limits for plutonium concentrations in nuclear fuel;
- ensuring conditions for safe operation at the maximum allowable temperatures of the shells of fuel elements (fuel elements) and nuclear fuel.

The proposed work is devoted to the analysis of solutions to the above problems of safe operation and prospects for the use of MOX fuel.

II. LITERATURE DATA ANALYSIS AND PROBLEM STATEMENT

The experience of operating MOX fuel in thermal reactors has revealed a number of serious problems [1-31]:

- the high activity of the still unirradiated MOX fuel and the associated need for additional safety studies when using existing equipment for handling fresh fuel;
- due to the high specific activity of plutonium-239, which is several orders of magnitude higher than that of uranium-235, in order to maintain acceptable water activity in the reactor during operation of the nuclear power plant, it will be necessary to reduce the number of leaky fuel elements by orders of magnitude both in gas leakage and in direct contact of fuel with water;
- loading even part of the core with MOX fuel reduces the efficiency of regulatory bodies (due to the high absorption in plutonium, which shifts the balance of absorption in the reactor in its favor);
- the fraction of delayed neutrons in plutonium is three times less than in uranium (for plutonium $\beta_{\text{eff}} \approx 0,2\%$), this changes the properties of the reactor during power maneuvers in a more dangerous direction, etc.

Therefore, as shown by the estimates and experience of countries actively operating MOX cartridges in light-water reactors, without changing the characteristics of the control and protection system and / or corresponding modernization for safety reasons, it is most advisable to load no more than 1/3 of MOX fuel into the reactor with 2/3 ordinary uranium.

In addition, the handling of irradiated MOX fuel is a serious problem. Irradiated in light-water reactors, this fuel differs from uranium in the complex isotopic composition of fission products and actinides. In plutonium-containing fuel, a much larger amount of small actinides (americium, curium) is formed due to greater neutron capture in the thermal part of the spectrum; an increase in the fraction of these nuclides leads to increased activity and heat release of irradiated (U-Pu) O₂ fuel. Processing such fuel is technologically more complicated than processing uranium. Due to the complex nuclide composition, the operational parameters of regenerated 2nd generation plutonium are deteriorating, and it would be advisable to use it only for loading in fast neutron reactors.

In the event of a serious accident at the reactor with a violation of the tightness of the core, the dose at a predetermined distance from the reactor, if it is loaded by a third with MOX fuel, will be 2.3 to 2.5 times higher. The consequences of the release of radioactivity will worsen as many times. The use of MOX can aggravate the negative environmental consequences of the accident 3.2 to 4 times.

Therefore, the decision to use the «non-design» MOX fuel for VVER requires a thorough analysis of nuclear and radiation safety.

Prospects for the use of MOX fuel in the global nuclear energy industry after the accident at the Fukushima nuclear power plant (Japan) are considered in the well-known FIVEX ANALYTICS GROUP review based on the materials of the international environmental association Bellona, data from TVEL and USEC Inc.

Following the accident at the Fukushima-Daiichi nuclear power plant, the Japanese MOX fuel program is experiencing serious difficulties due to opposition from local authorities. Back in May 2004, in a referendum in Niigata Prefecture, residents voted against using MOX fuel at the Kashiwazaki-Kariva Nuclear Power Plant, one of the largest reactors in the world for burning this type of fuel. The protests of residents were associated primarily with the lack of guarantees regarding the safety level of using MOX fuel. Such fears are justified: from 1986 to 2011, over ten fairly serious accidents occurred at Japanese nuclear power plants, resulting in more than 500 injuries. However, despite protests regarding the use of MOX fuel, the government planned to scale up its use. If all fuel processing programs are completed, then in ten years Japan will become the first country in the world in terms of plutonium reserves — 80-90 tons (30 tons — export from Europe, 6 tons — as a result of reprocessing of spent nuclear fuel at the Tokai plant and 50 tons — at the factory in Rokkas).

Before the events in Fukushima, many energy companies in Japan planned to use MOX fuel in their reactors. So, in 2006, Shikoku Electric Power Co. and Kyushu Electric Power Co. received approval from local authorities for the use of MOX fuel at one of their units since 2011. At the same time, the load of MOX fuel should be equal to 25% of the reactor core.

Japan began decommissioning the first Fugen MOX fuel reactor, which was shut down in March 2003. It is planned that it will be completely decommissioned and dismantled by 2028 with a total decommissioning cost of about 70 billion yen (about 700 million euro).

The delay in the return to Japan of high-level waste obtained from the reprocessing of spent nuclear fuel in the UK at the Sellafield nuclear complex will also be delayed. According to the British, scheduled for 2011, the shipment of high-level waste can be canceled. The accident at the Japanese Fukushima nuclear power plant affected the UK's MOX plans, where the Nuclear Decommissioning Authority (NDA) announced the closure of the Sellafield MOX fuel plant as soon as possible.

In the United States, where there has long been an ambiguous attitude towards plutonium even before the events in Fukushima, the only major potential customer was interested in MOX fuel - TVA, which has long-standing ties with federal authorities. The South Carolina MOX Plant is building a Shaw-Areva MOX Services joint venture. Commercial operation of the plant is scheduled for 2018. Prior to the accident at the Fukushima nuclear power plant, TVA and Energy Northwest announced their agreement to discuss the possibility of using MOX fuel. One of the documents to be revised will be the EIA (Environmental Impact Assessment). To expand its customer base, the US Department of Energy (DOE) is proposing changes to the design documentation for the MOX plant and expanding its capabilities. If previously it was believed that it would produce fuel only for PWR reactors, today it is a question of fabricating fuel for BWR reactors and modern projects of light water reactors, such as AP-1000.

In France and Belgium, as well as throughout the world, MOX fuel producers face a number of technical difficulties in its production and storage, which primarily worsens the economy of the MOX industry. These are factors such as:

- the presence of high alpha radiation, as well as americium-241, which is a highly radioactive source of gamma radiation;
- restriction on the duration of plutonium storage in the production of MOX fuel (2-3 years);
- to increase the time during which the fuel rods can work in the reactor, it is necessary to increase fuel enrichment. With an enrichment of 4.2% for uranium fuel and 8% for MOX fuel and the currently allowed maximum concentration of plutonium in MOX fuel in France, 5.3%, the equivalent energy production is 30,000 MW-day per 1 ton of heavy metal, while when using uranium fuel, 47,000 MW-days are produced;
- during experiments on the reprocessing of spent MOX fuel, plutonium, which had a large amount of impurities, was isolated, and therefore it was less likely to be split than plutonium released from uranium spent nuclear fuel. Further purification of plutonium was associated with an increase in the cost of reprocessing. When reprocessing SNF of MOX fuel, higher concentrations of transuranium elements and, consequently, more radioactivity are formed, compared with reprocessed uranium fuel. This was one of the reasons that in August 1996, Electricity de France (EDF) announced its intention to store spent MOX fuel without reprocessing it.

Thus, it is not yet clear what the policy should be regarding what to do with spent MOX fuel in the future. From the foregoing, we can conclude that the proposed direction for the use of MOX fuel is considered as a way to reduce plutonium reserves. However, the MOX direction can bring a lot more problems than plutonium itself. If we add to the economic disadvantages of MOX fuel the existing environmental consequences of the functioning of the nuclear, including plutonium, industry, it makes sense to classify and consider plutonium as waste. In this case, there will be a hope that future generations will have to handle less plutonium than it could be as a result of the implementation of the current French reprocessing projects of COGEMA.

The possibility of using MOX fuel in VVER reactors has not yet been confirmed. Today, such an opportunity is only being explored as part of a calculation and experimental justification. The purpose of this work is to understand the advantages and disadvantages of this direction of plutonium utilization, as well as to adopt the experience of Western Europe. At the Physics and Energy Institute. A.I. Leipunsky (Obninsk) began construction of a SUPR stand (mixed uranium-plutonium gratings) for experimental study of the safety characteristics of light-water reactors using plutonium, including weapons-grade, in fuel. In order to decide whether it is possible or not to use MOX fuel in VVER reactors, it is necessary to determine the conditions for combining the design of reactors and fuel assemblies with MOX fuel, to study at least about a dozen more important characteristics that affect safety and efficiency.

Thus, after the Fukushima accident, the world nuclear community was seriously "thinking" about the further prospects of using «non-projected» MOX fuel at nuclear power plants.

The countries with the most developed plutonium technology in the "post-Fukushima" period are France, Belgium, and the United Kingdom. For example, in France, which is a monopolist in the supply of

MOX fuel and a leader in its use, about 20 reactors are loaded with mixed fuel and an increase in their number is planned. At the same time, France in the near future does not plan to process irradiated MOX cassettes.

Partially loaded with MOX fuel produced in France, and several power units in Japan. Moreover, Japan plans to build an advanced pressurized water reactor, which is supposed to be the first reactor in the world to operate exclusively on MOX fuel, as well as the construction of a commercial factory manufacturing MOX fuel for thermal reactors.

Despite the existing problems with the operation of mixed fuel in light-water reactors and the difficulties encountered when converting reactors to its use, research in this direction continues. For example, specialists from the French AREVA group continue to study the possibilities of completely transferring EPR reactors to work with MOX cassettes. At the same time, the first block with EPR, which is now being built in Finland, will be operated on uranium fuel - at least in the first years after launch. The Flamanville-3 block, being built in France, will begin work with a standard zone containing 30% of MOX assemblies. For its UK projects, the AREVA group hopes to bring the content of mixed assemblies in the zone to 50%.

Currently, China is showing great interest in the field of MOX fuel production by attempting to acquire MOX fuel fabrication technology from Belgian companies and research centers. Belgian experts have extensive experience in this area - a total of 670 tons of SNF were processed in Belgium with the return of the separated plutonium to the fuel cycle. But a few years ago, Brussels announced the completion of the MOX program, after which the achievements of the Belgian nuclear scientists remained unclaimed in their own country.

However, the attitude towards the use of MOX fuel in light-water reactors is ambiguous. For example, Ukraine does not plan to use mixed fuel in light-water reactors, postponing the study of this problem until the transition to the construction of fast reactors.

The most devastating explosions of the Fukushima accident in 2011 with disastrous environmental consequences occurred at the 3rd unit of the Fukushima-Daiichi NPP, which, according to the official information of the operating organization TERSO, was loaded with MOX fuel by one third, and in the premises of the spent fuel storage unit of the 4th unit (the most discussed version is the flow of hydrogen through ventilation from the 3rd block). A possible reason for such consequences of a severe accident at the 3rd block could be directly MOX fuel due to the known «negative» differences from the design uranium fuel (which was used on the 1st and 2nd blocks): lower values of the melting temperature (up to 100 °C) and thermal conductivity, which determines the higher temperature (also up to 100 °C) of nuclear fuel in the center of a fuel rod, *ceteris paribus*; increased radiotoxicity and other neutron and thermophysical characteristics. But at the present time there are no sufficient grounds for these statements, since there is still no sufficiently substantiated and realistic comparative analysis of the scenarios of the development of severe accidents and the conditions for the occurrence of gas-vapor explosions at the power units of the Fukushima-Daiichi NPP. Therefore, there are no sufficient grounds for «curtailing» national programs for the exploitation of MOX fuel, especially since plutonium utilization issues remain extremely relevant.

At the same time, one of the unambiguous lessons of the Fukushima accident is the need for additional safety analysis when using «non-design» MOX fuel in BWR, PWR, VVER and other types of reactors. In particular, such issues, in our opinion, include the assessment of permissible concentrations of plutonium and the proportion of fuel elements with MOX fuel in the reactor core to ensure the design level of safety.

At the present stage, both energy and weapons-grade plutonium-239 are disposed of as part of mixed uranium-plutonium fuel based on UO₂ and PuO₂dioxides (MOX fuel) in the core of power reactors. Natural or depleted uranium is used as the uranium component, and weapon or energy plutonium as the plutonium component. The plutonium content in MOX fuel loaded into light-water reactors is 3–5%, and in fast-neutron reactors — 10% and higher.

For domestic VVER, MOX fuel (even with partial loads of the reactor core and/or spent fuel storage pool) is a «non-project» nuclear fuel that can be used both for the forced disposal of energy and weapons-grade plutonium, and for the modernization of nuclear reactors (for example, promising nuclear reactors of a new generation of safety based on MOX fuel for the formation of a self-regulating stable «neutron-fission wave»— the so-called wave reactors).

A promising direction for improving the safety of nuclear energy is the development and implementation of «wave» nuclear reactors with the organization of self-regulation of energy emissions in nuclear fuel. The introduction of self-regulating modes of energy release in nuclear fuel based on the peculiarities of the development of wave neutron-physical processes of fission of «heavy» uranium and transuranium elements determines the possibility of increasing nuclear safety, since the management of reactive accidents significantly reduces the impact of possible erroneous operator actions.

The theoretical foundations of the neutron-fission wave of "burning" were considered in the famous works of L. P. Feoktistov and were further developed in works on the so-called "wave" reactor of the scientific school of V. D. Rusov [1]. In a simplified form, the model of self-regulation of energy releases of the "wave" reactor can be represented as follows:

- 1) a source with a primary critical mass creates a chain reaction of a neutron flux, which is absorbed by the relatively «inert» uranium U-238;
- 2) upon neutron absorption, U-238 turns into U-239, which, due to two β -decays with a certain half-life, passes into the fission-active plutonium isotope Pu-239;
- 3) with sufficient energy release in neighboring regions, the concentration of plutonium will become more critical and the amplitude of the energy release will shift — a neutron-fission energy propagation wave occurs;
- 4) with certain ratios of the number of «nucleated» atoms to the number of “disappeared” (reproduction coefficient), regardless of the initial conditions, a steady-state self-regulating wave of “burning” of nuclear fuel with a characteristic propagation velocity is formed:

$$D \approx L / \tau_{1/2},$$

where L is the mean free path of the neutron in the medium, $\tau_{1/2}$ — is the characteristic half-life of the isotopes. Preliminary calculations revealed that the decisive parameter for the discrepancy in the relative power of internal energy releases for plutonium and uranium fuel is a significant difference in a certain temperature range between the values of the cross section for fission of nuclides. To find the cross sections of nuclear reactions averaged over the neutron spectrum, for example, it is necessary to know the energy spectrum of neutrons. As is known, the energy spectrum of neutrons in thermal reactors differs from the Maxwell spectrum because of its nonequilibrium state, due to the fact that in the core there are continuous processes of neutron formation during fission of nuclei by fission spectra for fissile nuclides, processes of neutron deceleration and their absorption. The most widely used approach is based on the neutron gas model, which assumes that thermalized neutrons are distributed over the energies of the Maxwell distribution, but not at the temperature of the fissile medium of the core, but at the temperature of the neutron gas. In this case, the influence of absorption or insufficient deceleration in the medium on the spectrum shift from the «pure» Maxwell is estimated by the input temperature of the neutron gas, which is given by the following semi-empirical expression:

$$T_{\text{nr}} = T \left[1 + 1,4 \Sigma_a(kT) / \xi \Sigma_s(1 \text{ eV}) \right]$$

where T — temperature of the fissile medium, Σ_a — macroscopic cross section for the absorption of the medium (thermal), ξ — average logarithmic decrement of energy during deceleration, Σ_s — macroscopic cross section for the scattering of the medium, $\xi \Sigma_s$ — slowing capacity of the medium (at 1 eV).

The cross sections are averaged over the neutron spectrum according to the following expression:

$$\langle \sigma(E_{\text{rp}}, T) \rangle = \frac{\int_0^{E_{\text{rp}}} E^{1/2} e^{-E/kT_{\text{nr}}} \sigma(E, T) dE}{\int_0^{E_{\text{rp}}} E^{1/2} e^{-E/kT_{\text{nr}}} dE}$$

where σ — micro section of the nuclear reaction, E — energy of neutrons, E_{rp} — boundary energy of thermal neutrons.

The result of averaging according to this expression for capture, fission, and absorption microcross sections with typical dependences of microcross sections σ on neutrons v_n exothermic nuclear reactions, has a general form (the expression for the micro section of the fission reaction is presented here).

$$\langle \sigma_f(T) \rangle = \frac{\sqrt{\pi}}{2} \sigma_f^T \sqrt{\frac{300}{T_{\text{nr}}}} g_f(T_{\text{nr}}) F(z_{\text{nr}})$$

where σ_f^T — thermal fission micro section at a neutron velocity of 2200 m/s, $z_{\text{nr}} = E_{\text{rp}} / kT$ — dimensionless boundary energy for the Maxwell spectrum, i.e. The «cross-linking energy» of the Maxwell and Fermi neutron spectra, $F(z)$ is the correction function, T_{ng} — temperature of the neutron gas, $g_f(T_{\text{ng}})$ — Westcott factor for the fission reaction.

Using the above formulas, we calculated the temperature dependences of the cross sections for nuclear fission and radiation capture averaged over the neutron thermal spectrum for plutonium-239 and uranium-235. For uranium-238, the temperature dependences of the radiation capture cross sections averaged over the thermal neutron spectrum were calculated.

The obtained calculated temperature dependences of the cross sections of nuclear fission and radiation capture reactions averaged over the thermal neutron spectrum for plutonium-239 and uranium-235 are shown in Fig. 1 and 2, respectively. In fig. Figure 2 also presents the calculated temperature dependence of the radiation capture cross sections averaged over the thermal neutron spectrum for uranium-238.

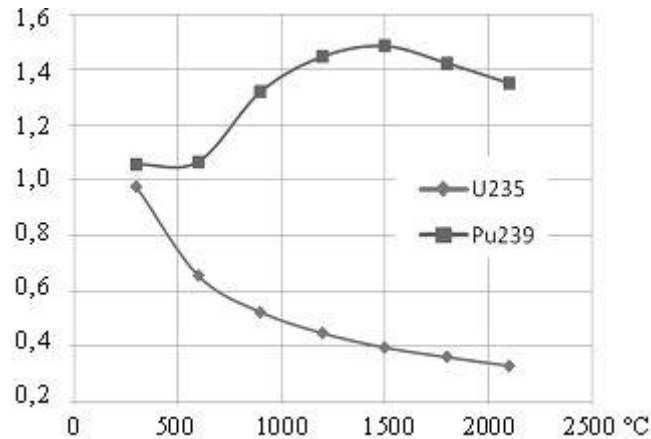


Fig. 1. The dependence of the fission cross sections of uranium-235 and plutonium-239, averaged over the thermal spectrum of neutrons, from the temperature of the fissile fuel medium (values of cross sections are normalized to σ_f^0) [1]

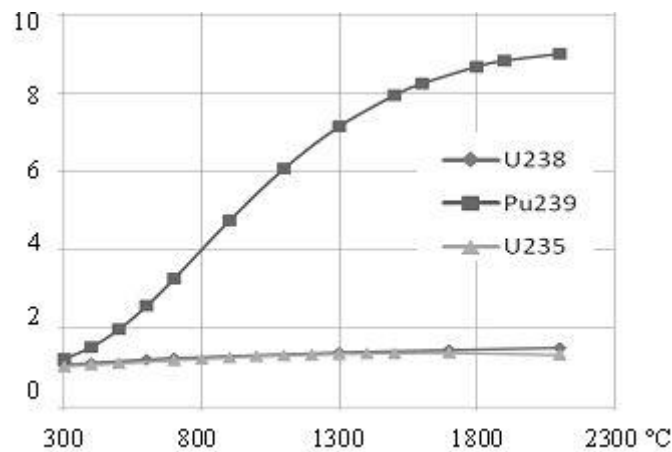


Fig. 2. The dependence of the radiation capture cross sections of uranium-235, uranium-238 and plutonium-239, averaged over the thermal spectrum of neutrons, from the temperature of the fissile fuel medium (values of cross sections are normalized to σ_f^0) [1]

The main advantage of a «wave» reactor is the formation of a self-regulation mode of energy release without operator intervention, which, *ceteris paribus*, provides increased «internal» nuclear safety (prevention and / or management of reactive accidents).

The main limitation of the prospects of a «wave» reactor is technical difficulties in the formation of a steady neutron-fission wave, as well as the need to justify the criteria of both «internal» (associated with reactive accidents) and «external» (associated with failures of equipment / systems of the reactor installation) safety taking into account the features of the modes of wave «burning» of nuclear fuel.

The introduction of wave reactors requires the solution of the following main problems, taking into account the features of wave energy release:

- development and maintenance of stability criteria for wave energy release.
- additional analysis of nuclear and radiation safety.
- analysis of the conditions of occurrence and consequences of thermohydrodynamic instability of the coolant, etc.

For operating, transient and emergency modes of wave reactors, the most dangerous are the conditions for the occurrence of thermoacoustic instability of the coolant. Thermoacoustic instability is characterized by high-amplitude pressure pulsations (up to 50% of the average pressure in the core) with a fundamental frequency inversely proportional to the speed of sound in the coolant. Thermoacoustic instability of the coolant determines the possibility of occurrence of critical hydrodynamic shock for reliability on the structure of the reactor core and equipment of the reactor loop [26 — 31].

III. THE PURPOSE AND OBJECTIVES OF THE STUDY

- 3.1. Analysis of operating experience and utilization of MOX fuel.
- 3.2. Analysis of the prospects for using MOX fuel for wave nuclear reactors.

IV. MATERIALS AND RESEARCH METHODS

In the general case, the difference between the neutron and thermophysical properties of the design UO_2 and alternative MOX fuel is determined by the following modernization parameters:

$$\tilde{K}_1 = \frac{\left[\frac{R_T^{-1}}{C_T M_T} \left(1 - \frac{1}{\text{Nu} + 1} \right) \right] (\text{MOX})}{\left[\frac{R_T^{-1}}{C_T M_T} \left(1 - \frac{1}{\text{Nu} + 1} \right) \right] (\text{UO}_2)}, \tilde{K}_2 = \frac{\left[\frac{N_T}{C_T} + \frac{R_T^{-1} T_{\text{TH}} \text{Nu}}{(\text{Nu} + 1) C_T M_T} \right] (\text{MOX})}{\left[\frac{N_T}{C_T} + \frac{R_T^{-1} T_{\text{TH}} \text{Nu}}{(\text{Nu} + 1) C_T M_T} \right] (\text{UO}_2)} \quad (1)$$

Assuming an insignificant effect of changes in thermophysical properties and conditions of external heat transfer, the relevant modernization parameters:

$$\tilde{K}_1 \approx 1, \tilde{K}_2 = \frac{\frac{K_N N_T (\text{UO}_2)}{C_T} + \frac{R_T^{-1} T_{\text{TH}} \text{Nu}}{(\text{Nu} + 1) C_T M_T}}{\frac{N_T (\text{UO}_2)}{C_T} + \frac{R_T^{-1} T_{\text{TH}} \text{Nu}}{(\text{Nu} + 1) C_T M_T}} \quad (2)$$

In this case, the determining parameter of modernization is the indicator

$$K_N = \frac{N_T (\text{MOX})}{N_T (\text{UO}_2)} \quad (3)$$

which actually reflects the ratio of the internal energy release capacities of MOX and UO_2 fuel. The power of the internal energy release of nuclear fuel is determined by the conditions of the ongoing neutron-physical processes and depends on the density of the neutron flux of the j_{th} nuclide in the composition of the nuclear fuel Φ_j , the capture cross section σ_j and the concentration of the j_t nuclide γ_j . MOX fuel is characterized by an increased plutonium content relative to the design UO_2 fuel (not more than 1% Pu). The temperature dependences of the capture cross section significantly differ both on a qualitative and a quantitative level in the temperature range of nuclear fuel of more than 500 °C. Therefore, the modernization parameter for the difference in heat dissipation power can be represented as

$$K_N = \frac{\sigma(\text{Pu})\gamma(\text{Pu})}{\sigma(\text{UO}_2)\gamma(\text{UO}_2)} \quad (4)$$

It is also necessary to take into account possible differences in the maximum temperatures of MOX and UO_2 fuels at the initial moment of the accident/transient.

In this case, the current differences in the maximum temperature of the MOX and UO_2 fuels

$$\begin{aligned} \Delta T_{\text{tm0}}(K_N, t) &= T_{\text{tm0}}(\text{MOX}) \exp \left[- \int_0^t K_1(\tau) d\tau \right] + \\ &+ \exp \left[- \int_0^t K_1(\tau) d\tau \right] \int_0^t \tilde{K}_2 K_2(\text{UO}_2, \tau) \exp \left[\int_0^\tau K_1(\xi) d\xi \right] d\tau \\ &- T_{\text{tm0}}(\text{UO}_2) \exp \left[- \int_0^t K_1(\tau) d\tau \right] - \\ &- \exp \left[- \int_0^t K_1(\tau) d\tau \right] \int_0^t K_2(\text{UO}_2, \tau) \exp \left[\int_0^\tau K_1(\xi) d\xi \right] d\tau = \\ &= \exp \left[- \int_0^t K_1(\tau) d\tau \right] \left\{ \Delta T_{\text{tm0}} + \int_0^t (\tilde{K}_2 - 1) K_2(\text{UO}_2, \tau) \exp \left[\int_0^\tau K_1(\xi) d\xi \right] d\tau \right\} \end{aligned} \quad (5)$$

where $\Delta T_{\text{tm0}} = T_{\text{tm0}}(\text{MOX}, t = 0) - T_{\text{tm0}}(\text{UO}_2, t = 0)$.

An analysis of equation (5) showed that in the considered approximation, the current discrepancies in the values of the maximum temperature of the MOX and UO₂ fuel during the accident are determined by the K_N indicator:

$$\Delta T_{tm} \leq 0 \quad \text{при} \quad K_N \leq 1 \quad (6)$$

The K_N indicator for the relative residual heat of MOX and UO₂ fuel can be estimated from the well-known semi-empirical dependences of the Way-Wigner-Way-Finger for UO₂ fuel:

$$N_T(\text{UO}_2, t) = N_{T0}(\text{UO}_2, t_0) \cdot 6,6 \cdot 10^{-2} \left[t^{-0,2} - (t+t_0)^{-0,2} \right] \quad (7)$$

and Untermaier-Wells for MOX fuel

$$N_T(\text{MOX}, t) = N_{T0}(\text{MOX}, t_0) \cdot 10^{-2} \left[(t+10)^{-0,2} - (t+t_0+10)^{-0,2} + (t+2 \cdot 10^{-1,2})^{-0,2} - (t+t_0+2 \cdot 10^{-1,2})^{-0,2} \right] \quad (8)$$

Where $N_{T0}(t_0)$ — heat release power at the initial moment of reactor shutdown during the working campaign period t_0 , t — current time of the accident after shutdown of the reactor.

The difference in the current values of the maximum temperature of the cladding of the MOX and UO₂ fuel rods at $Nu \gg 1$:

$$\Delta T_{o6} = T_{o6}(\text{MOX}) - T_{o6}(\text{UO}_2) = \frac{\Delta T_{tm}}{Nu} \quad (9)$$

Nuclear safety conditions when replacing a design UO₂ fuel with MOX fuel at the maximum allowable temperatures of the fuel $T_{tm}^n(\text{MOX})$ and fuel cladding:

$$T_{tm}(\text{UO}_2) + \Delta T_{tm} < T_{tm}^n(\text{MOX}), T_{o6}(\text{UO}_2) + \frac{\Delta T_{tm}}{Nu} < T_{o6}^n(\text{MOX}) \quad (10)$$

When upgrading the properties of nuclear fuel (including MOX fuel), it is necessary to take into account the dependence of the determining neutron and thermophysical parameters on the temperature of the fuel.

Under certain conditions and conditions of nuclear fuel, such a significant dependence can lead to aperiodic (spontaneous change of state) or periodic (oscillatory process) instability. In accordance with the general theory of instability, any system can be subject to random (fluctuation) perturbations (effects and / or changes in the determining parameters of the state of the system). Depending on the current state of the system, these perturbations can either «decay» in time (the system is stable) or lead to a spontaneous change in the determining parameters (the system is aperiodically unstable) or oscillatory processes (periodic instability). The energy «source» of unstable processes is the conversion of the internal energy of the system.

The main determining parameter of the state of nuclear fuel is its temperature T_T . Fluctuation perturbations of the fuel temperature δT_T depending on the current state under certain conditions can lead to temperature instabilities of an aperiodic or periodic nature.

Negative consequences for the thermal instability of nuclear fuel for nuclear safety may include:

- exceeding safety limits for the temperature of nuclear fuel and the cladding of a fuel rod;
- instability of neutron-physical processes, which under certain conditions leads to the loss of controlled regulation of the power of a nuclear reactor (unregulated "acceleration" or shutdown);
- spontaneous increase in the temperature of nuclear fuel in a stopped reactor or spent fuel pool, etc.

Let us consider the conditions for the occurrence of thermal instability of nuclear fuel in the linear approximation ($\delta T_T \ll T_T$). In this case, the equation of the heat balance of nuclear fuel in the format of fluctuation perturbations of the temperature of nuclear fuel δT_m has the form

$$\frac{d\delta T_{tm}}{dt} = \left(-K_1 + \frac{dK_2}{dT_{tm}} T_{tm} \right) \delta T_{tm} \quad (11)$$

Solution (11):

$$\delta T_{tm} \approx \exp \left[\left(-K_1 + \frac{dK_2}{dT_{tm}} T_{tm} \right) t \right] \quad (12)$$

Solution (12) implies the criterion and condition for the thermal stability of nuclear fuel

$$K_{HT} = -\tilde{K}_1 + \frac{d\tilde{K}_2}{dT_{tm}} T_{tm} \leq 0$$

At $K_{NT} \leq 0$, temperature perturbations will «decay» in time (the fuel temperature is stable), and at $K_{NT} > 0$, it will spontaneously increase (temperature instability of nuclear fuel).

V. ANALYSIS OF THE RESULTS

The corresponding calculated values $K_N = N_T(\text{MOX}, t) / N_T(\text{UO}_2, t)$ of the indicator for the relative power of the residual heat are shown in Fig. 3.

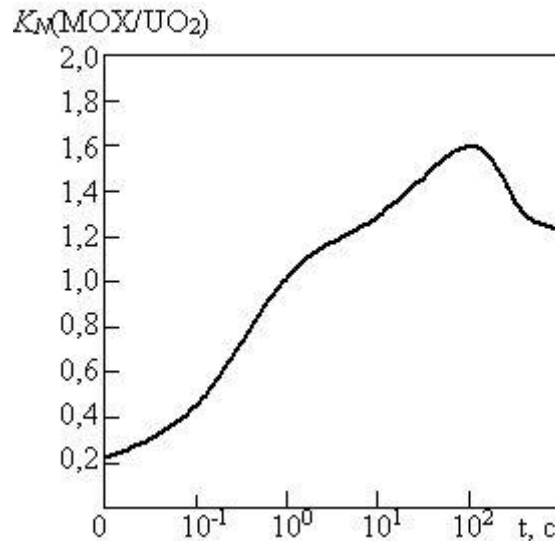


Fig. 3. The indicator of modernization K_N on the relative power of residual heat release of MOX and UO_2 fuel during an accident (at maximum % increase).

The analysis determines an insufficiently substantiated result: at the initial stages of the accident, the power of residual heat release of MOX fuel is significantly less than the corresponding values for UO_2 fuel with the same reactor operating time at power. In addition, the Untermayer-Wells relationship for MOX fuel does not take into account the concentration of plutonium-239. According to well-known studies, the concentration of plutonium in fuel has a significant effect (both qualitatively and quantitatively) on the temperature dependence of the neutron capture cross-sectional area, and, accordingly, on the power of heat release.

To simplify the analysis, the «operating time» of Pu in the design UO_2 fuel was conservatively neglected. The results of an express analysis of nuclear safety allow us to conclude that in the region of relatively low temperatures of nuclear fuel characteristic of a shutdown reactor and spent nuclear fuel storage pools, the permissible concentration of plutonium is no more than 8%, and for higher temperatures no more than 3% (Fig. 4).

At higher Pu concentrations, an additional analysis of the nuclear safety of MOX fuel is necessary.

The interpretation of the mechanism of occurrence of thermal instability of nuclear fuel is as follows. A fluctuation increase in the temperature of nuclear fuel δT_T determines a corresponding increase in the power of internal heat release δN_i and a further increase in the temperature of nuclear fuel, ceteris paribus. However, an increase in T_T also determines an increase in the temperature difference between nuclear fuel and the external environment, an increase in the density of heat removal from nuclear fuel q_{HT} and a corresponding decrease in T_T . If the influence of the growth effect of N_T under the influence of a perturbation δT_T exceeds the effect of increasing the density of heat removal from nuclear fuel, thermal instability of nuclear fuel occurs. Otherwise, the growth of T_T , determined by the increase in the power of internal heat release of nuclear fuel, is «compensated» by the increase in heat removal from nuclear fuel — the system is stable against fluctuation disturbances δT_i .

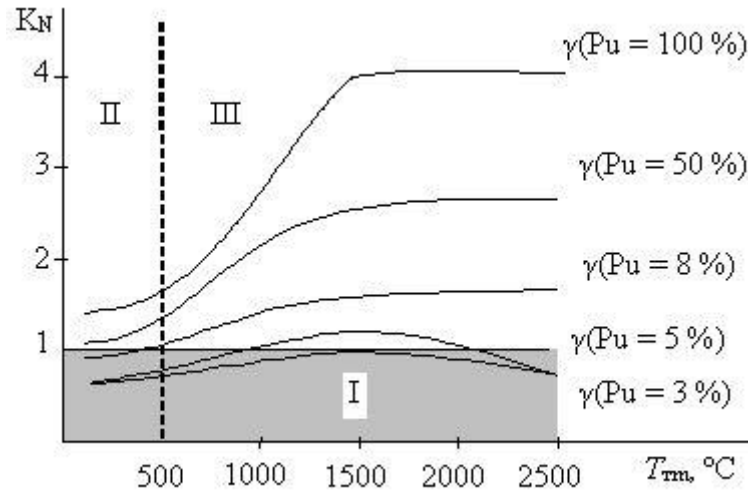


Fig. 4. Nuclear safety limits of MOX fuel by fuel temperature T_T and plutonium concentration γ (Pu): I — area of permissible adaptation of MOX fuel; II — temperature ranges of nuclear fuel at a stopped reactor and/or in a spent fuel pool; III — temperature ranges of nuclear fuel in workers and emergency operation of the reactor

In the approximation $K_2 \sim K_N \sim \sigma$ and for a known temperature dependence of the capture cross-sectional area σ for the design UO_2 fuel (see Figs. 1 and 2):

$$\frac{dK_2(UO_2)}{dT_{tm}} < 0$$

and the thermal stability of the UO_2 fuel is satisfied over the entire temperature range.

In the considered approximation, the determining effect of differences in the heat dissipation power of the design and alternative MOX fuel on the feasibility of nuclear safety conditions, the criterion of thermal instability:

$$K_{HT} = -\tilde{K}_1 + \frac{dK_2}{dT_{tm}} \tilde{K}_2 T_{tm} \leq 0, \text{ where } \tilde{K}_2 \sim K_N, \frac{d\tilde{K}_2}{dT_{tm}} \sim \frac{dK_N}{dT_{tm}}.$$

Taking into account the results obtained, the dependences of K_N on the temperature of nuclear fuel and plutonium concentration ($K_N \approx 1$ and $dK_N/dT_{tm} \approx 0$ — see Figs. 1 and 2), the thermal stability conditions when using MOX fuel do not change at a plutonium concentration of not more than 3%.

VI. DISCUSSION OF THE RESULTS OF THE STUDY

6.1. The use of MOX fuel as an alternative to UO_2 fuel using «slow» neutrons in WWER / PWR / BWR type nuclear reactors is impractical, since it significantly reduces the level of nuclear and radiation safety.

6.2. Based on the presented simplified safety analysis methods, it was determined that the safety conditions for the maximum allowable temperatures of the claddings of fuel rods and nuclear fuel, as well as the thermal stability of nuclear fuel are provided at a plutonium concentration of 239 no more than 3%.

The refinement of the obtained value of the maximum permissible concentration of plutonium 239 can be carried out on the basis of more adequate methods of nuclear safety analysis.

6.3. The use of MOX fuel is a promising direction for creating wave nuclear reactors based on the initiation and maintenance of a neutron-fission wave of «burning» nuclear fuel. The calculation analysis carried out in the work determined that the maximum permissible concentration of plutonium 239 to ensure the nuclear safety conditions of the wave reactor should also be no more than 3%. Further, it is necessary to further substantiate the sufficiency of such a concentration of plutonium 239 for initiating and maintaining a stable wave of «burning» nuclear fuel of wave reactors.

VII. CONCLUSIONS

1. Operational experience and scientific and technical studies determine the restrictions on the production and introduction of MOX fuel for WWER / PWR / BWR reactors for the following main reasons:

— the high activity of the still unirradiated MOX fuel and the associated need for additional safety studies when using existing equipment for handling fresh fuel;

— due to the high specific activity of plutonium-239, which is several orders of magnitude higher than that of uranium-235, in order to maintain acceptable water activity in the reactor during operation of the nuclear

power plant, it will be necessary to reduce the number of leaky fuel elements by orders of magnitude both in gas leakage and in direct contact of fuel with water;

— loading even part of the core with MOX fuel reduces the efficiency of regulatory bodies (due to the high absorption in plutonium, which shifts the balance of absorption in the reactor in its favor);

— the fraction of delayed neutrons in plutonium is three times less than in uranium (for plutonium $\beta_{\text{eff}} \approx 0,2\%$), this changes the properties of the reactor during power maneuvers in a more dangerous direction, etc.

The melting point of the $\text{UO}_2\text{-PuO}_2$ compound decreases approximately in proportion to the PuO_2 content from 2840 °C for pure UO_2 to 2390 °C for pure PuO_2 . From these data, it can be calculated that the melting point of a typical MOX will be 20 to 40 degrees below the melting point of uranium oxide. At high degrees of burnup, the melting temperature may still drop. The thermal conductivity of MOX also decreases monotonically as the plutonium content increases. There are also some differences in the physico-mechanical properties (Young's modulus, Poisson's ratio). At high degrees of burnout, an increase in the yield of gaseous fission products from MOX is observed in comparison with UO_2 .

In addition, the handling of irradiated MOX fuel is a serious problem. Irradiated in light-water reactors, this fuel differs from uranium in the complex isotopic composition of fission products and actinides. In plutonium-containing fuel, a much larger amount of small actinides (americium, curium) is formed due to greater neutron capture in the thermal part of the spectrum; an increase in the fraction of these nuclides leads to increased activity and heat release of irradiated (U-Pu) O_2 fuel. Processing such fuel is technologically more complicated than processing uranium. Due to the complex nuclide composition, the operational parameters of regenerated 2nd generation plutonium are deteriorating, and it would be advisable to use it only for loading in fast neutron reactors.

In the event of a serious accident at the reactor with a violation of the tightness of the core, the dose at a predetermined distance from the reactor, if it is loaded by a third with MOX fuel, will be 2.3 to 2.5 times higher. The consequences of the release of radioactivity will worsen as many times. The use of MOX can aggravate the negative environmental consequences of the accident 3.2 to 4 times.

2. The burning issue remains the disposal of MOX fuel in the state of spent nuclear fuel. The total amount of plutonium stored in the world at the beginning of the 21st century in various forms is estimated at 1239 tons, of which two-thirds are in SNF of the nuclear power plant. Already, more than 120 thousand tons of spent fuel is in storage, and by 2020 it will be 450 thousand tons.

3. A promising direction for the use and disposal of MOX fuel is the creation of reactors based on wave energy release in the core (wave reactors), as well as reactors with fast neutrons. In wave reactors, plutonium acts as the initiator and catalyst of the energy release wave. However, the introduction of wave reactors requires solving the following problems, taking into account the peculiarities of energy release:

— additional analysis of nuclear and radiation safety;

— determination of maximum permissible concentrations of plutonium for safety;

— additional analysis of the conditions for the occurrence of neutron-thermo-hydrodynamic instability in the core, etc.

4. Based on the presented simplified safety analysis methods, it was determined that the safety conditions for the maximum allowable temperatures of the claddings of fuel rods and nuclear fuel, as well as the thermal stability of nuclear fuel, are ensured at a plutonium-239 concentration of not more than 3%.

5. The calculation analysis carried out in the work determined that the maximum permissible concentration of plutonium-239 to ensure the nuclear safety conditions of the wave reactor should also be no more than 3%. Further, it is necessary to further substantiate the sufficiency of such a concentration of plutonium-239 for initiating and maintaining a stable wave of "burning" nuclear fuel of wave reactors.

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