

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

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ABSTRACT

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 materials²³⁵ — into the fuel cycle of modern nuclear

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: Pu²³⁹ 60%, Pu²⁴⁰ 25%, Pu²⁴¹ 10%, Pu²⁴² 3%, Pu²³⁸ 2% [2, 3].

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 UO₂ is significant — it is lower for MOX. The melting point of the UO₂-PuO₂ compound decreases approximately in proportion to the PuO₂ content from 2840 °C for pure UO₂ to 2390 °C for pure PuO₂. 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 physicomachanical 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 UO₂.

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

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.

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 UO₂ and alternative MOX fuel is determined by the following modernization parameters:

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

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

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

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 UO₂ 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 UO₂ 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 UO₂ fuels at the initial moment of the accident/transient.

In this case, the current differences in the maximum temperature of the MOX and UO₂ fuels

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

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

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 UO_2 fuel rods at $Nu \gg 1$:

$$\Delta T_{06} = T_{06}(\text{MOX}) - T_{06}(\text{UO}_2) = \frac{\Delta T_{\text{tm}}}{Nu} \quad (9)$$

Nuclear safety conditions when replacing a design UO_2 fuel with MOX fuel at the maximum allowable temperatures of the fuel $T_{\text{tm}}^{\text{n}}(\text{MOX})$ and fuel cladding:

$$T_{\text{tm}}(\text{UO}_2) + \Delta T_{\text{tm}} < T_{\text{tm}}^{\text{n}}(\text{MOX}), T_{06}(\text{UO}_2) + \frac{\Delta T_{\text{tm}}}{Nu} < T_{06}^{\text{n}}(\text{MOX}) \quad (10)$$

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_{tm} has the form

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

Solution (11):

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

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

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

At $K_{\text{HT}} \leq 0$, temperature perturbations will «decay» in time (the fuel temperature is stable), and at $K_{\text{HT}} > 0$, 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.

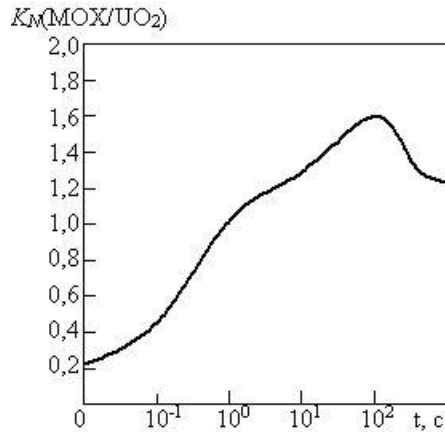


Fig. 3. The indicator of modernization K_N on the relative power of residual heat release of MOX and UO_2 fuel during an accident (at maximum increase).

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_T 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_T .

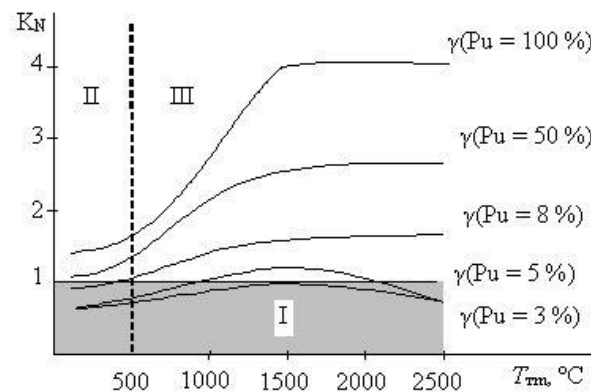


Fig. 4. Nuclear safety limits of MOX fuel by fuel temperature T_T and plutonium concentration γ (Pu): I — area of permissible adaptation of MOX fuel; II — temperature ranges of nuclear fuel at a stopped reactor and/or in a spent fuel pool; III — temperature ranges of nuclear fuel in workers and emergency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

$$\frac{dK_2(\text{UO}_2)}{dT_{tm}} < 0$$

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:

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

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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