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NEW MATERIALS AND COATINGS FOR NUCLEAR TECHNOLOGY

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ABSTRACT

The possibility of using new materials in nuclear and thermonuclear reactors is being considered. The most preferred fuel for the fast reactor can be considered mixed mononitride (MN) with the addition of uranium nanopowder. The following fuels are of interest for thermal neutron reactors: $UO_{2.1}$, UN, U_2N_3, U_3Si_2 with the addition of nanopowder of slightly enriched uranium or Be (BeO). Ceramic fuel containing depleted uranium with nano-additives of metallic uranium can be used in the blanket of a thermonuclear reactor. As a neutron multiplier in blanket, the long-lived minor actinides (^{237}Np, ^{241}Am and ^{243}Am) can be used. There is a radioactive waste of nuclear reactors. Liquid lead and (or) its alloys have a number of advantages when cooling the core of a fast reactor or blanket of a thermonuclear reactor. The use of tungsten coatings of structural materials (stainless steel) contributes to solving the problem of corrosion and erosion in liquid lead. All innovations can be implemented within the framework of existing technologies.

INTRODUCTION. FORMULATION OF THE PROBLEM. SOLUTION STAGES

We single out two stages of solving optimization problems in the design of nuclear reactors. At the first, problems of optimal design (often multicriteria) with restrictions are solved. As control parameters, the geometric characteristics of the nuclear reactor and its components, the flow rate of the coolant, and others are chosen. Among the restrictions are the functionals characterizing the operation of the installation in normal operation and emergency modes. After completion of the conceptual studies, the tasks of optimal design lose their relevance. They are being replaced by other, including optimization, tasks (of the second stage). The solution to such problems allows you to choose new materials. It is believed that the installation parameters are known (accepted in the conceptual design). They can be adjusted. As control parameters, the properties of fuel, coolant, structural materials, and cost characteristics are considered. They analyze their technological development and compatibility. Actual solution of problems without a criterion for finding the sets of acceptable materials.

When choosing the optimal fuel for a nuclear reactor, the density, heat capacity and thermal conductivity of the fuel are considered among the control parameters, and the melting point or the beginning of fuel decomposition is among the constraints of the task. The maximum allowable temperature may vary during the solution of the task. If it decreases, additional (including technological) restrictions are needed to prevent this. In some cases (in the most successful cases), with a qualitative change in the composition of the fuel, the maximum allowable temperature can be kept constant.

The choice of preferred structural materials of nuclear and thermonuclear reactors involves the inclusion in the composition of the control parameters of density, thermal conductivity, heat capacity of structural materials, and neutron absorption cross sections. Among the limitations of the task, maximum temperatures, strength, and others are considered. For the practical implementation of the obtained solutions, it is possible to consider coatings of materials, two- and three-layer materials, and others.

THE PRACTICAL IMPLEMENTATION OF THE TASKS OF THE SECOND STAGE

FAST REACTORS

Software. The calculation and optimization complex "DRAGON-M" is used, improved and adapted by the author to solve new optimization problems. The program "FRISS" is used for modeling emergency conditions, including anticipated transients without scram (ATWS) [1]. The control parameters of the optimization complex "DRAGON-M" includes the properties of core materials (fuel, coolant, structural materials): density, thermal conductivity, heat capacity, porosity, viscosity. The codes "MCNP" [2] and "WIMS" [3] with modern constant software and a number of auxiliary programs developed by the author are also used.

Fuel composition optimization. The opposite role of the Doppler reactivity coefficient (DRC) in the safe termination of ATWS initiated by loss of flow or loss of forced circulation without scram (LOF WS) and transients overpower without scram (TOP WS) in high-power reactors with oxide fuel creates serious difficulties in the design of safe nuclear power plants. When switching to MN fuel, this opposite role is partially leveled, smoothed, but not excluded. The same role is achieved when using metallic fuel. But it has a low melting point.

The task boils down to the choice of fuel properties that guarantee the same role of DRC in different ATWS and a high (acceptable) melting point. Initial data are reactor layout parameters obtained at the first stage, control parameters are the density, heat capacity and thermal conductivity of fuel. Constraints for the functionals determining reliability, safety, etc. are considered. The optimality criterion may be absent.

Solving the task with restrictions for different functionals (including functionals for which restrictions simulate the safety of the reactor from accidents of the ATWS type), considering the density and thermal conductivity of the fuel among the control parameters, we can obtain the range of acceptable values of the density and thermal conductivity of the fuel. The range of acceptable values must correspond to the same role of DRC in emergency modes LOF WS and TOP WS. The range of acceptable values of the density and thermal conductivity of the fuel is shown in Fig. 1.

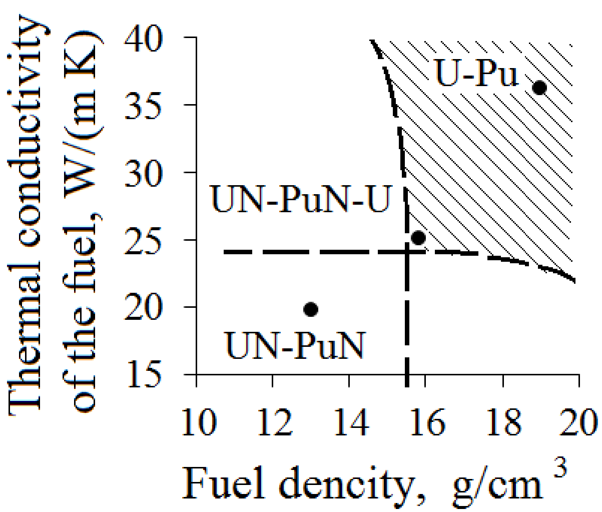

Fig. 1. The range of acceptable values of the density and thermal conductivity of the fuel (shaded). Inside the region there is a point corresponding to U-Pu fuel, outside the region there is a point corresponding to MN fuel, cermet fuel (UN-PuN-U) is located near the boundaries of the region.

A further increase in the density and thermal conductivity of the fuel compared to MN is needed. This is possible during the transition to cermet fuel based on MN and metallic uranium. The melting point of such a fuel decreases markedly with increasing proportion of metal. The task is to increase the density, thermal conductivity and heat capacity of the fuel (achieved by increasing the metal fraction) without significantly reducing (ideally, keeping it at a constant level) the maximum allowable temperature (achieved by increasing the ceramic fraction). An increase in the metal fraction leads to an increase in the safety of the reactor, including due to the achievement of the same DRC in different emergency situations. The task is clearly conflictual.

In the permissible region are metallic fuel, metallic, doped with zirconium and "traditional" cermet fuel. However, all these fuels are characterized by a relatively low melting point.

Nuclear fuels are usually made in the form of sintered pellets containing micrograins of ceramics (heavy metal oxide or nitride). The porosity of such fuel is about 25%. The porosity of a fuel made from UN nanoscale powder (nanopowder) can be reduced to 5% [4, 5]. This means that it is possible to use a mixture of ceramic micrograins and metal nanopowder. Uranium metal nanopowder can be placed in the pores of microgranular ceramic fuels. In this case, the volume fraction of nanopowder can be up to 20%. The characteristic grain size of a nanopowder of metallic uranium is tens of nanometers.

The ratio of the volumes of micrograins MN and nanopowder U, equal to **20/75**, corresponds to a mass ratio MN / U of 0.39. This is the maximum allowable mass content of metal nanopowder in the fuel, associated with the limitation of the maximum allowable temperature. If necessary, increase the proportion of metallic uranium can use larger grains of ceramics.

To preserve the effective multiplication factor during the transition from MN to MN-U fuel, enriched uranium nanopowder should be used. For example, for a BREST-OD-300 reactor [6] with MN fuel based on dump uranium and “civil” plutonium (extracted from WWER spent fuel) cooled by natural liquid lead, uranium metal must be enriched to 14.9%.

Note that the uranium nanopowder fills the pores between the MN grains, not replacing the mononitride. As a result, the thermal power of the reactor increases by approximately 20%. Uranium metal nanopowder is a good getter (absorber) of free nitrogen released from MN fuel during reactor operation and migrating to the cladding. In the presence of free nitrogen, the corrosion rate of the inner surface of the cladding of the fuel elements increases. The addition of uranium metal nanopowder to MN fuel helps minimize the corrosion rate of the inner surface of cladding, increasing the reliability of fuel pins, and, therefore, increasing the reliability of the reactor as a whole.

The transition from ceramic to ceramic-metal fuel leads to an increase in the average neutron energy in the reactor. This leads to a decrease in the absolute value of negative DRC: from minus $1.75169 \cdot 10^{-6}$ to minus $1.31367 \cdot 10^{-6}$ during the transition from MN fuel to cermet fuel at a 20% content of uranium filling the pores between MN micrograins. Fig. 2 illustrates the dependence of the Doppler reactivity effect when changing the temperature of the fuel from 0 to ± 100 K from the nominal when changing the volume fraction of metallic uranium from 0 to 20%.

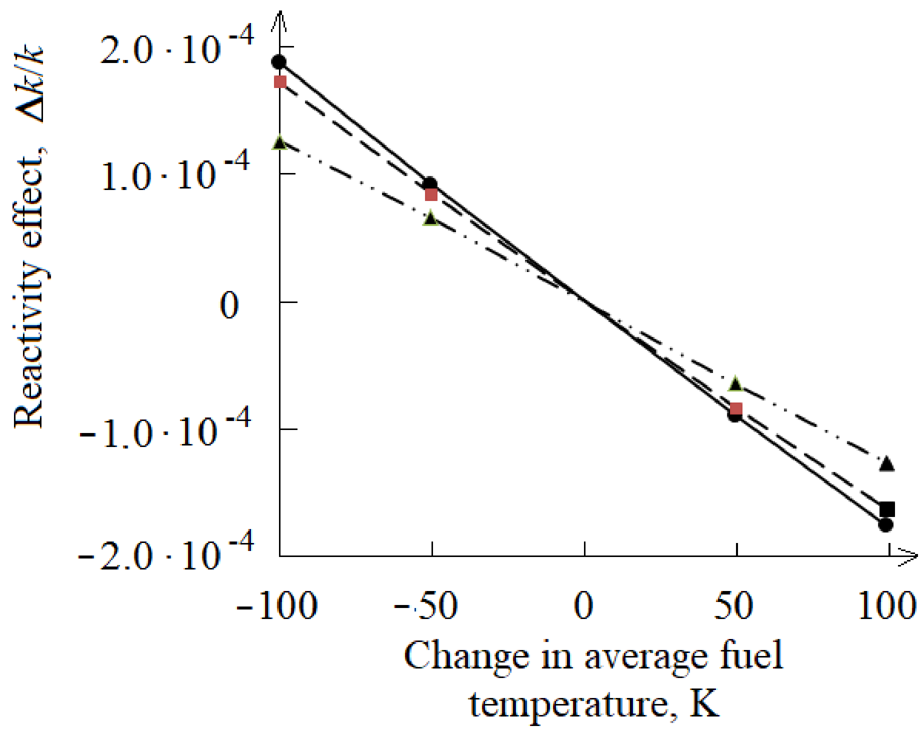

Fig. 2. The Doppler reactivity effect when using fuel (% of volume are indicated):
MN (●), *MN*-5%U (■), *MN*-20%U (▲)

Calculations show that the same role of DRC in different emergency conditions (LOF WS, TOP WS) can be achieved not only with the use of metallic (including doped) fuel, but also for cermets fuel with a volume fraction of metallic uranium in mixed mononitride fuel of about 15 ... 20%. At the same time, the void reactivity effect (VRE) is reduced, mainly due to a decrease in its spectral component (Fig. 3) and the neutron balance is improved. To compensate (absorption) excess in relation to the implementation of the chain reaction of neutron fission, you can use an additional absorber in the core. If the removal of this absorber from the core during loss or decrease in the density of the coolant is impossible, then the presence of an additional absorber will significantly reduce the VRE.

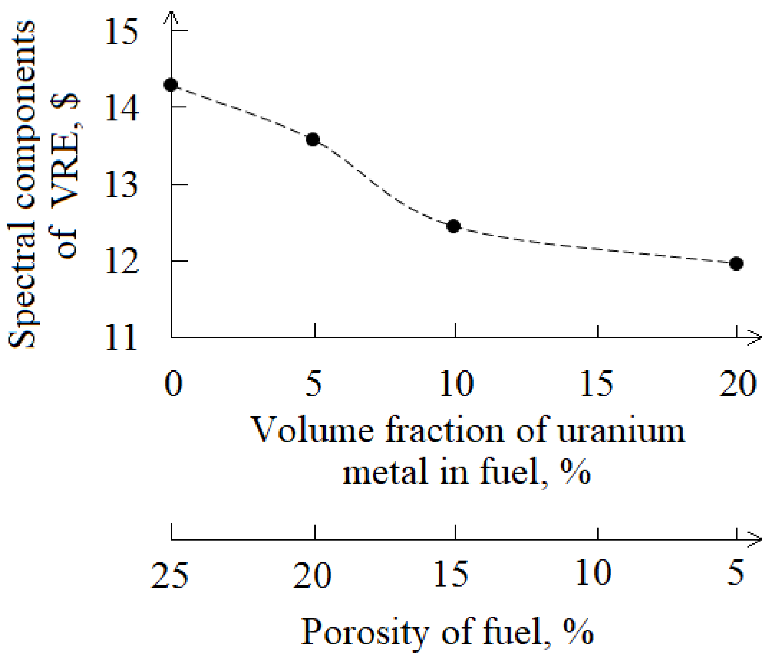

Fig. 3. Dependence of the spectral component of VRE on the volume fraction of uranium metal in fuel and porosity

European projects of lead-cooled fast reactors are focused on the use of mixed oxide (MOX) fuel [7]. Similar innovations can be proposed for such a fuel by placing a uranium metal nanopowder (a good getter of free oxygen coming out of plutonium oxide) between the oxide micrograins. In Fig. 4 shows the values of the DRC for different fuel compositions. Ceramic (MOX or MN) is a micrograin, metal uranium is a nanopowder. From Fig. 4 it follows that the DRC decreases in absolute value with increasing fuel density.

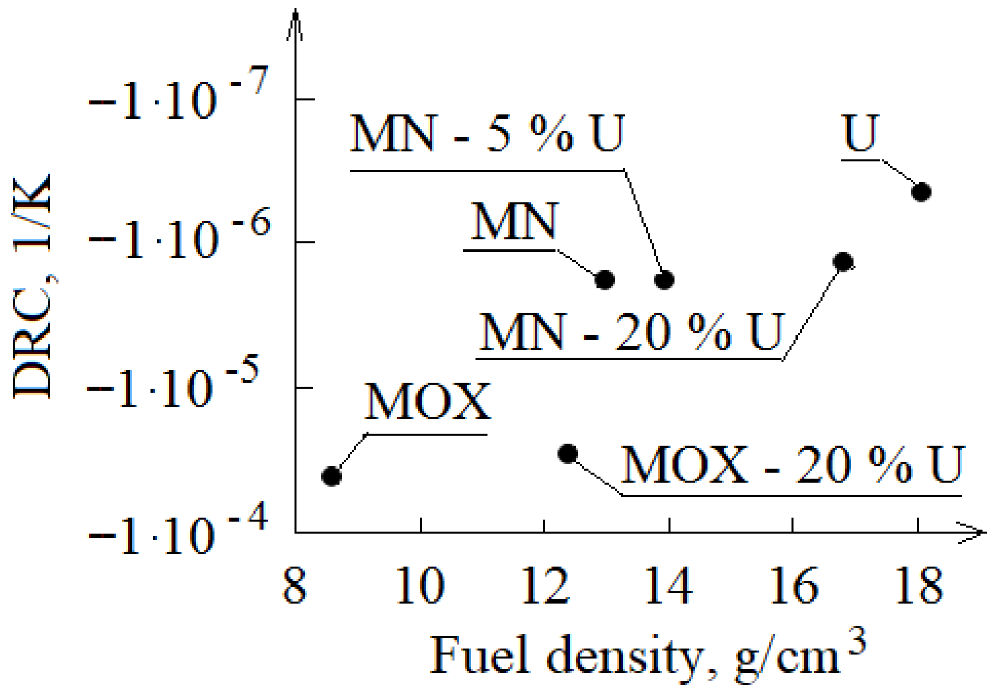

Fig. 4. DRC values for different types of fuel (% of volume are indicated)

The role of tungsten coatings of structural materials. The absorber can be placed in the form of fuel pins coatings. In [8], the possibility of using fuel rods with tungsten coatings deposited using low-temperature plasma spraying is considered. The purpose of using such coatings was to minimize the rate of corrosion and erosion of structural materials in liquid lead. Coatings contribute to a marked reduction of VRE.

When using lead with a content of ^{208}Pb of about 98%, even in the case of a complete transition to tungsten claddings, the neutron balance is better than when using natural lead in the absence of tungsten in the core.

Coolant. At the first stage of solving optimization problems, it was found that, according to a set of criteria, liquid lead is one of the most preferred coolants for fast reactors [9]. The second stage requires solving the local problem of optimization of lead coolant in the framework of the reactor concept being developed. Technological additives (alkali metals) to lead coolant can solve some problems, but also create a number of additional problems [9]. The safety of the installation depends on the isotopic composition of lead. Optimization of the composition of lead coolant does not require isotope separation. The maximum content of ^{208}Pb is characteristic of lead of thorium ores (^{208}Pb is the final decay product of ^{232}Th), ^{206}Pb is typical of lead of uranium ores (^{206}Pb is the final decay product of ^{238}U). By mixing lead from different deposits and (or) different samples of one deposit, we can obtain a continuous spectrum of changes in the concentrations of stable isotopes of lead, i.e., lead of a given isotopic composition.

For low power reactors, a lead-206 coolant is preferred. Its use allows to minimize the polonium-210 production. In high power reactors, a coolant based on the double-magic lead-208 isotope should be used. This will significantly reduce the VRE and reduce the risk of reactive accidents.

The MCNP calculations show that for BREST reactors of infinitely high power with MN fuel, the content of ^{208}Pb in the lead coolant should be at least 80 ... 85%. When using tungsten coatings of cladding of fuel pins and fuel based on MN micrograins and a uranium metal nanopowder, lead with a ^{208}Pb isotope concentration of more than 70 ... 75% is suitable, which corresponds to the minimum concentration of this isotope in known samples of lead extracted from thorium ores. Thus, lead extracted from any thorium deposit can be used as a coolant.

THERMAL NEUTRON REACTORS

The following fuels can be considered promising for thermal neutron reactors: $UO_{2,1}$, UN, U_2N_3, U_3Si_2 (with a much lower concentration of fissile nuclides) with the addition of nanopowder of slightly enriched uranium or Be (BeO). A preliminary analysis of the use of such fuels in the WWER-1200 reactor is presented in [1].

([1] N.V. Bulgakov, V.S. Okunev, "Possibilities for improving WWER parameters during the transition to cermet fuel based on uranium metal nanopowder", Nuclear Physics and Engineering, 2018, vol. 9, No. 5, pp. 418-424, DOI: 10.1134/ S2079562918050044.)

Optimization of the geometry of the lattice of fuel pins when using fuels based on U_3Si, U_3Si_2 and cermet UO_2 with the addition of U nanopowder improves the neutron balance. This helps to increase the power of the reactor and the campaign, and also opens up opportunities for the use of structural materials with a large neutron absorption cross section, for example, stainless steel.

The current stage of development of thermal neutron reactors in the direction of improving safety largely repeats the development of new concepts of fast reactors from the beginning of the 1990s. For light-water reactors, the possibilities of increasing inherent safety through the use of new types of fuel (Accident Tolerant Fuel) with a higher concentration of heavy atoms, density and thermal conductivity are considered. Such fuels have previously been considered for fast reactors. The problem of increasing power in the framework of the given dimensions of the core is considered. And no less, light-water reactors are significantly inferior to fast reactors in safety.

The use of fuel with a higher concentration of uranium (as compared to UO_2) can improve the neutron balance in the core, i.e. effective multiplication factor k. Excess neutrons (with respect to the criticality condition $k = 1$) can be used to increase the fuel breeding ratio in the core and to increase the reactor campaign.

The thermal power of the reactor can be estimated as

$$W \approx \Sigma_f \, \varphi \, E_f \, V, \quad (1)$$

where Σ_f is the macroscopic fission cross section, is the average neutron flux density, E_f is the energy released during one fission of the nucleus, V is the volume of the core. In turn,

$$\Sigma_f = \sigma_f^{(5)} N_5 + \sigma_f^{(8)} N_8,$$

where $\sigma_f^{(5)}$ and $\sigma_f^{(8)}$ are the microscopic fission cross sections for ^{235}U and ^{238}U, respectively, N_5 and N_8 are the concentration of nuclei ^{235}U and ^{238}U, respectively.

When using fuel with a higher uranium content and a lower content of light nuclei, the average neutron energy in the core increases. On the one hand, this leads to an increase in the number of neutrons produced per nuclear fission, and, as a result, leads to an increase in the breeding ratio in the core and in the campaign of the reactor. On the other hand, this leads to a decrease in the ^{235}U microscopic fission cross section. (An increase in the intensity of ^{238}U fission in the spectrum of a thermal neutron reactor does not play a decisive role.) As a result, the neutron balance worsens and the fuel breeding ratio in the core decreases. As a result, the simultaneous action of these two factors may not lead to a noticeable increase in the campaign.

As follows from relation (1), an increase in power with an increase in the uranium content in the fuel can occur due to an increase in Σ_f due to an increase in the concentration of ^{235}U and an increase in the contribution to the fission neutron balance of ^{238}U (an increase in the fast fission factor). The fast fission factor is defined as ratio of the next number of fast neutrons produced by all fission to the number of fast neutron produced by thermal fissions. An increase in the neutron flux density upon transition to a more rigid spectrum in this case plays a secondary role.

In a first approximation, we can assume that

$$W / N_5 \approx \text{const}_1 \text{ or } W / \gamma \approx \text{const}_2,$$

where γ is the density of the fuel. If the power of a reactor with UO_2 fuel is taken as a unit, then with a constant fuel rod spacing (corresponding to WWER-1200) and an average neutron flux density when using fuel based on UO_2 and nanopowder of metallic uranium, the power can be increased to 1.38. When using USi, USi_2, U_3Si, U_3Si_2 and UN - 0.91, 0.78 ... 0.81, 1.36, 1.06 and 1.51, respectively. However, intensive absorption of neutrons by ^{14}N nuclei in the composition of mononitride fuel reduces the neutron density (and, accordingly, the neutron flux density) in the core, the power of 1.51 (relative to the reactor with UO_2 fuel) is not achievable.

Calculations for a WWER-1200 type reactor show that an increase in the campaign at the fuel rod spacing optimal for a given fuel occurs mainly due to an increase in the infinity multiplication factor k_{inf} (Fig. 5). Other factors play a secondary role. Thus, the maximum campaign corresponds to the fuel at which the highest k_{inf} value is achieved when using "fresh" fuel. Fig. 6 shows the dependence of k_{inf} on the step h of the fuel rod array. The calculations were carried out according to the WIMS program. The numbers indicate the types of fuel used: *1* - UO_2, *2* - UO_2-U (U nanopowder), *3* (dotted line) - USi, *4* - USi_2, *5* (dotted line) - U_3Si, *6* - U_3Si_2, *7* - UN.

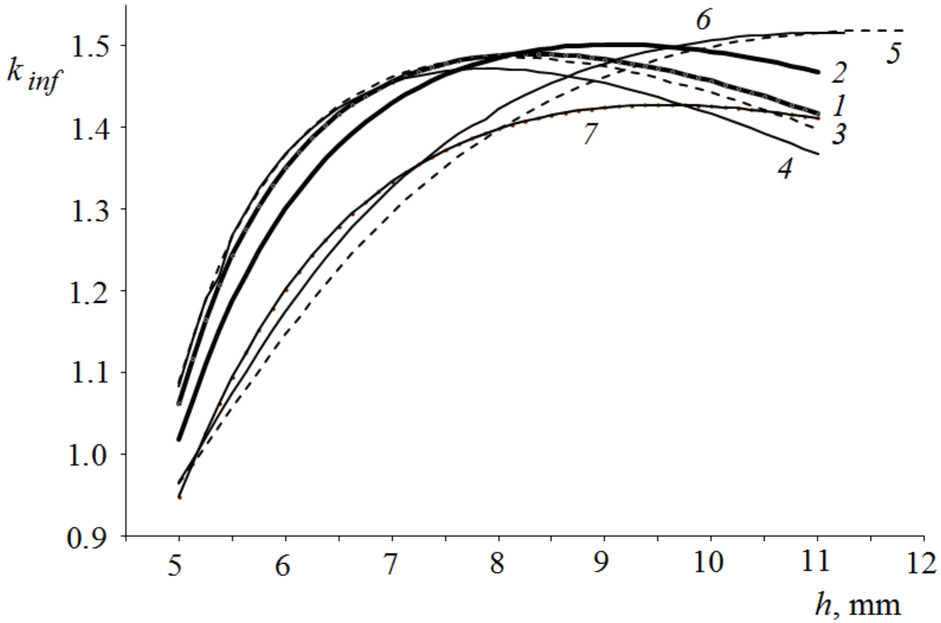

Fig. 5. The dependence of k_{inf} on the step of the lattice of the fuel rods for different types of fuel

(N.V. Bulgakov, V.S. Okunev, "Possibilities for improving WWER parameters during the transition to cermet fuel based on uranium metal nanopowder", Nuclear Physics and Engineering, 2018, vol. 9, No. 5, pp. 418-424, DOI: 10.1134/ S2079562918050044.)

Silicides can compete with ceramic-metal fuel based on UO_2 and nanopowder of uranium metal. In this case, the variation of the properties of cermet fuel is possible due to the difference in the content of metallic uranium, silicide - due to the different contents of uranium and silicon atoms in the fuel. At small lattice spacings, USi fuel has clear advantages over UN.

The dependences of k_{inf} on the time t of reactor operation are similar for all types of fuel. The highest rate of fuel burnout is observed in the first few days. The $k_{inf}(t)$ dependences for all types of fuel have the form of approximately parallel straight lines at each calculated burnup step (30 days). We can assume that

$$T / \Delta k_{inf} \approx \text{const}_3,$$

where T is the campaign of the reactor, Δk_{inf} is the excess of k_{inf} over the value ensuring the criticality of the reactor.

As expected, the highest k_{inf} value over the entire time interval of the reactor operation is achieved using fuels with the highest uranium content and corresponds to U_3Si. It is followed by U_3Si_2, UO_2 with the addition of uranium nanopowder and USi. Conventional UO_2 fuel is inferior to them. The worst case scenario is mononitride fuel due to the more intense absorption of neutrons by ^{14}N nuclei (99% of natural nitrogen), which worsens the neutron balance. The idea of enriching fuel nitrogen with the ^{15}N isotope, expressed by V.V. Orlov at the dawn of the development of the concept of fast lead-cooled reactors with mononitride fuel, which later turned out to be irrelevant for fast reactors, is important for thermal neutron reactors.

BLANKET OF A THERMONUCLEAR REACTOR

Ceramic fuel with additives of a depleted U nanopowder can be used in the blanket of a thermonuclear reactor. As a neutron multiplier, the minor actinides ^{237}Np, ^{241}Am and ^{243}Am (long-lived radioactive waste from nuclear power) can be used. In the spectrum of thermonuclear neutrons, they have significant advantages compared with ^{9}Be. The cross section for their fission in the spectrum of thermonuclear neutrons is approximately 10 times higher than the cross section for the reaction (n, 2n) by ^{9}Be (Fig. 6).

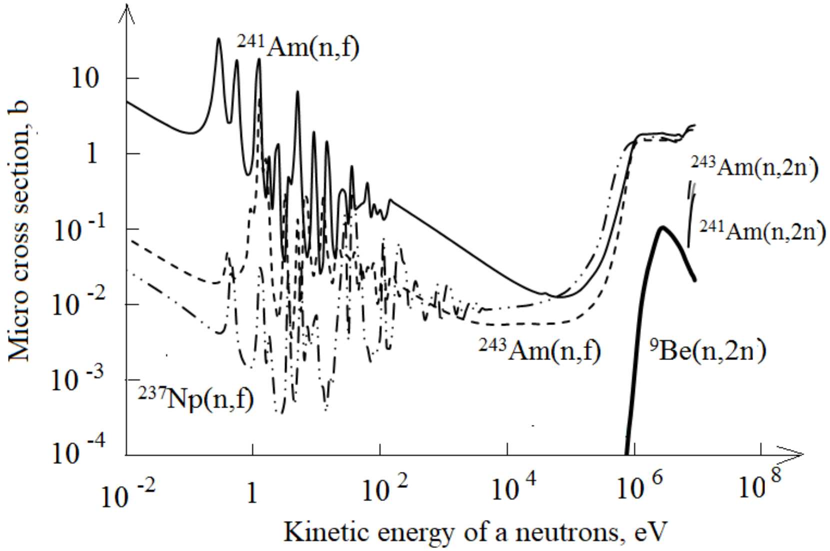

Fig. 6. Energy dependence of the cross sections (according to [2]),

$1 \text{ b} = 10^{-28} \text{ m}^2$.

([2] M.B. Chadwick, et. al., "ENDF/B-VII.1 Nuclear Data for Science and Technology: Cross Sections, Covariances, Fission Product Yields and Decay Data", Nuclear Data Sheets, Los Alamos National Laboratory Unclassified Report LA-UR 11-05121, **112-12**, 2887-2996 (Dec. 2011), htpps://t2.lanl.gov/nis/data.shtml.)

At neutron energies above 6 MeV, the reactions (n, 2n) are realized on Np and Am nuclei. If 2 neutrons can be obtained per beryllium-9 nucleus, then by fission of one neptunium nucleus or americium by a 14 MeV neutron, 4 or 5 neutrons can be obtained. In addition, at energies above 6 MeV, reactions of (n, 2n) are realized on the nuclei of minor actinides.

CONCLUSION

After determining the main characteristics of nuclear or thermonuclear reactors at the conceptual development stage, the formulation and solution of new optimization problems is relevant. This will allow you to adjust design decisions, which does not require a change in the basic layout parameters. The results of solving local correction problems may not have the possibility of practical implementation. However, if the practical implementation is feasible, one can expect not just improvements in nuclear technology, but also technological breakthroughs, and within the framework of existing technologies developed earlier.

When using fuel based on MN micrograins and U nanopowder, the task of increasing the power, reliability, and safety of a fast reactor is not in conflict. The greatest success should be expected with the integrated use of such fuel, lead coolant based on ^{208}Pb and tungsten coatings of structural materials. Such coatings are also of interest for blanket materials of a thermonuclear reactor. They will open up the possibility of using cheap lead, which is more contaminated with impurities. Tungsten can be used in the coating wall of the first wall of a thermonuclear reactor.

All the proposed innovations can be implemented within the framework of existing technologies.

THANKS FOR YOUR ATTENTION