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High Fuel Burn-up and Nonproliferation in PWR-type Reactor on the Basis of Modified Th-fuel

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Neutronics-physical characteristics of the fuel lattice of a PWR-type reactor cooled by light water and by a mixture of light and heavy water have been analyzed. Th-fuel containing an essential amount of ^{231}Pa and ^{232}U is used, which allows an increase in fuel burn-up by a factor of 2-5 compared with that of traditional oxide uranium fuel with light water. It is important to underline that this is attained under the negative coolant density reactivity effect using cross sections of ^{231}Pa and ^{232}U from the updated JENDL-3.2 nuclear library. This radical increase of fuel burn-up is accompanied by a small change of reactivity during fuel irradiation (K_∞=1.1÷1.0), that favorably affects safety parameters of the reactor operation. A considerable percentage of ^{232}U in fuel, and consequently in U, is a strong barrier against the proliferation of such weapon nuclide as ^{233}U.

KEYWORDS: Coolant Density Reactivity Effect (CDRE), modified Th-fuel means Th-fuel containing an essential amount of ^{231}Pa and ^{232}U, high fuel burn-up, nonproliferation of ^{233}U weapon nuclide, JENDL-3.2 and ENDF/B-IV evaluated nuclear data libraries.

1. Introduction

Neutronics-physical characteristics of a fuel lattice of PWR-type reactor cooled by light water and by a mixture of light and heavy water has been analyzed. Here, Th-fuel is used that contains an essential amount of ^{231}Pa and ^{232}U. In this case, along with traditional transition chain:

$$^{232}\mathrm{Th} \rightarrow {}^{233}\mathrm{U} \rightarrow {}^{234}\mathrm{U} \rightarrow {}^{235}\mathrm{U} \rightarrow \ldots,$$

so called “non-traditional” (modified) transition chain:

$$^{231}\mathrm{Pa} \rightarrow {}^{232}\mathrm{U} \rightarrow {}^{233}\mathrm{U} \rightarrow {}^{234}\mathrm{U} \rightarrow {}^{235}\mathrm{U} \rightarrow \ldots$$

plays an essential role.

The following distinctions between the modified chain and traditional chain in terms of neutron balance should be mentioned. Firstly, in the case of a modified chain, neutron radiative capture cross-section of initial fertile nuclide (^{231}Pa) is significantly superior to that of initial fertile nuclide in a traditional chain (^{232}Th). This means that ^{231}Pa plays the role of a burning absorber more effectively than ^{232}Th, and reduction of ^{231}Pa concentration can release significant positive reactivity of fuel. Secondly, in case of a traditional chain, fertile nuclides (^{232}Th, ^{234}U) and well-fissionable nuclides (^{233}U, ^{235}U) alternate. Also, if a fissionable nuclide transforms into a fertile one, the next fission reaction can take place only after another neutron radiative capture reaction that worsens the neutron balance. Quite another situation is established in the modified chain. The isotope ^{232}U is a moderately fissionable nuclide and,

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even if ^{232}U does not undergo fission but absorbs neutron without fission, it converts into well-fissionable nuclide ^{233}U. Thus, there is a link between two fissionable nuclides that is absent in a traditional chain. All these effects explain the fact that the modified chain can support neutron multiplying properties of fuel over a substantially longer time than compared with a traditional chain [1, 2].

2. Mathematical model and adopted approximations

The computer code package SCALE-4.3 [3], developed for neutronics analysis of light water reactors, was used in numerical calculations. Using the code, a one-dimensional infinite lattice of fuel elements and time behavior of the fuel composition is analyzed. Neutron transport within a reactor cell was calculated in P_8-approximation with a number of internal iterations sufficient to assess value K_∞ to an accuracy of 10^{-5}. The fuel composition was re-calculated with a time interval of 3 years or less, to assess the value of K_∞ to an accuracy of 10^{-4}. Eighty-one fission products with a higher reactivity impact and most mass (97.5% HM of all fission products) were taken into account. The calculations were performed in 44-group energy approximation using evaluated nuclear data file ENDF/B-IV with AMPX computer code for preparation of cross sections [4], ^{231}Pa and ^{232}U cross sections were taken from evaluated nuclear data file JENDL-3.2 and the number of fission neutrons ν_f (^{233}U) was reduced from 3.13 to 2.456 under recommendations [1].

3. Production of modified fuel and ability of fuel rods to work

A series of problems must be resolved to reach high fuel burn-up in a nuclear reactor. Some problems have been already analyzed in previous publications [1-2]. In brief, the following results have been obtained in these studies.

The problem of special fuel production, particularly artificial nuclides ^{231}Pa and ^{232}U (they are absent in nature because of short half-lives: 32,760 years and 69 years, respectively), can be resolved by fusion reactors [5]. The required power of fusion reactors will constitute a small fraction of the power produced by nuclear reactors loaded with special fuel. Therefore, dedicated fusion reactors may not be power producers but fuel producers, and this circumstance substantially simplifies the requirements to the design of these fusion reactors. The above artificial nuclides could be also produced by neutron irradiation of ^{230}Th in nuclear reactors.

The paper [2] considers the problem of the ability of fuel rods work under conditions of high fuel burn-up. It was evaluated that, even if fuel burn-up reaches a level of 30% HM, a damage dose of stainless steel is only about 65 displacements per atom (DPA). The paper [6] demonstrates that, for some types of stainless steels used as a construction material, maximal damage dose till loss of steel strength properties can reach a value of 180 DPA. Since, in the present paper, we propose to use not zirconium-based alloys (typical cladding material in thermal nuclear reactors) but stainless steel (typical cladding material for fast breeder reactors) as a construction material, these fuel rods will be able to work till fuel burn-up at a analyzed level of 12-20% HM without renewal of cladding. Pursuant to the data presented in the paper [7], steel claddings are able to withstand neutron fluence up to $4 \cdot 10^{23}$ n/cm^2 for neutrons with energy above 0.1 MeV. It was evaluated in the paper [2] that the fluence of high-energy neutrons at fuel burn-up of about 30% HM is lower by a factor about 3. So, at analyzed levels of fuel burn-up (12-20% HM), neutron fluence will be still lower. Vibro-packed oxide fuel elements were also developed. They were irradiated in a fast neutron

spectrum up to high burn-up as much as 32% HM without failure [8]. In the present study it was assumed that the fuel applied is a porous nitride fuel with reduced density: 9.8 g/cm^3, i.e. 31% lower than the density of uranium nitride fuel and 18% lower than the density of thorium nitride. There are no published data on the properties of protactinium nitride. Furthermore, there is no information on the swelling of mixed nitride fuel (Th,Pa,U)N. However, it is known that uranium mono-nitride is characterized by a substantially lower value of swelling than that of uranium dioxide, owing to better structural integrity [9].

The proposed fuel contains a substantial fraction of ^{232}U, the initial nuclide for the chain of radioactive decays, culminating in stable ^{208}Pb. This chain includes six α-decays and results in helium accumulation in fuel. If the helium amount is a substantial fraction of the amount of gaseous fission products, the problem of fuel cladding integrity will be more complicated. It was evaluated [2], that the helium addition, generated in α-decays to a total amount of gaseous fission products generated, may be 25% for fuel burn-up - 30% HM. It can be concluded that under less fuel burn-up the protection of the fuel cladding integrity will require proper selection of fuel porosity.

4. Coolant density reactivity effect

Here, we analyzed the modified fuel compositions which can provide the utmost possible values of fuel burn-up in light-water reactor lattice of the VVER-1000 type under the following constraints. Firstly, initial neutron multiplication factor in infinite medium K_∞ must be at a level of 1.1 for compensation of neutron leakage and for reliable reactor control during reactor operation. Secondly, the time-integrated coolant density reactivity effect (CDRE) must be non-positive for the entire fuel campaign. Thirdly, CDRE at fresh fuel must be equal to zero for safe start-up of the reactor. The first constraint really limits content of ^{233}U in the modified fuel while the second and third constraints restrict content of ^{231}Pa and ^{232}U in the fuel. In general, these constraints lead to a limitation on achievable fuel burn-up. Observance of these constraints, that are so attractive for safe reactor operation yet extremely stringent (K_∞ of fresh fuel in typical light-water power reactor is about 1.37), appeared impossible in case of light-water coolant. Therefore, we analyzed the possible use of a mixture of light and heavy water as a reactor coolant with heavy water fraction above 25%.

Figure 1 shows the dependencies of neutron multiplication factor on fuel burn-up for typical light-water reactor lattice (uranium dioxide fuel enriched up to 4.4%^{235}U) and for the same lattices of (^{231}Pa-^{232}U-^{233}U-Th)N fuel rods with a mixture of light and heavy water as a coolant (heavy water fraction in the mixture is a variable with values of 25%, 50%, 75% and 100%). It can be seen that burn-up of special thorium-based fuel reaches 12-20% HM, several times higher than that for traditional uranium dioxide fuel. In addition, the multiplying properties of the modified thorium fuel can vary within substantially narrower ranges, that favorably affects safety parameters of the reactor operation. Special notice should be given to the fact that the larger the heavy water fraction in the reactor coolant, the higher the burn-up of special thorium-based fuel. This can be explained by a shift of neutron spectrum into resonance energy range where the neutron multiplying properties of such a fuel are better than those within the thermal energy range [1].

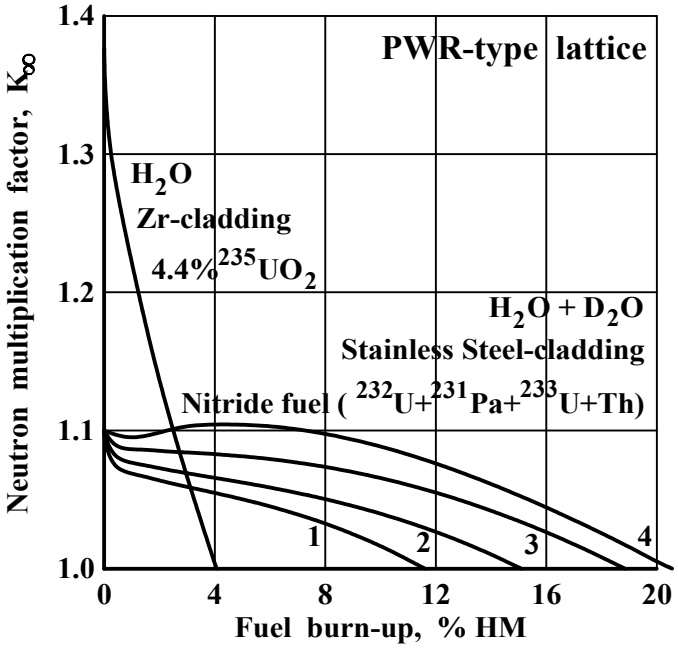

Fig.1 Dependence of neutron multiplication factor on fuel burn-up for conventional light-water power reactor of the VVER-1000 type for different fuel and coolant.

1-coolant: 25%D_2O + 75%H_2O, fuel: 7.1%^{232}U + 4.0%^{231}Pa + 4.6%^{233}U+ 84.3%^{232}Th;
2-coolant: 50%D_2O + 50%H_2O, fuel: 7.0%^{232}U + 5.8%^{231}Pa + 5.8%^{233}U+ 81.4%^{232}Th;
3-coolant: 75%D_2O + 25%H_2O, fuel: 7.8%^{232}U + 6.9%^{231}Pa + 6.5%^{233}U+ 78.8%^{232}Th;
4-coolant: 100%D_2O fuel: 10.0%^{232}U + 4.0%^{231}Pa + 5.0%^{233}U + 81.0%^{232}Th

In the paper [2], significantly higher values of fuel burn-up are presented for light-water coolant. These calculations are performed with application of evaluated nuclear data file ENDF/B-IV. Application of the updated library JENDL-3.2 for generation of ^{231}Pa and ^{232}U cross-sections library led to lower fuel burn-up and a positive CDRE value. The condition of non-negative CDRE for fuel life-time limited the content of ^{231}Pa and ^{232}U in fresh fuel, i.e. it is precisely these nuclides that contribute most to high fuel burn-up.

It can be also seen from Figure 1 that CDRE becomes positive in the first days of reactor operation. This is caused by the poisoning effect of ^{135}Xe. A further increase of CDRE is related to the poisoning effect of ^{149}Sm. The growth of CDRE value, induced by accumulation of ^{135}Xe and ^{149}Sm, can be explained by the fact that these fission products have large cross-sections of neutron radiative capture within the thermal energy range from 0.01 eV to 0.1 eV (about $3 \cdot 10^6$ barns and $5 \cdot 10^4$ barns, respectively). Then, these cross-sections substantially decrease within the higher energy range from 0.1 eV to 100 eV (about 0.4 barn and 100 barns, respectively). If coolant density decreases, neutron spectrum shifts from the range of large neutron capture cross-sections to the range of lower neutron absorption by way of these fission products. It improves the neutron multiplying properties of fuel, i.e. it leads to the growth of CDRE because the thermal energy range (0.01-100 eV) plays a decisive role in the neutron balance of the thermal reactor. All the effects are demonstrated in Figure 2.

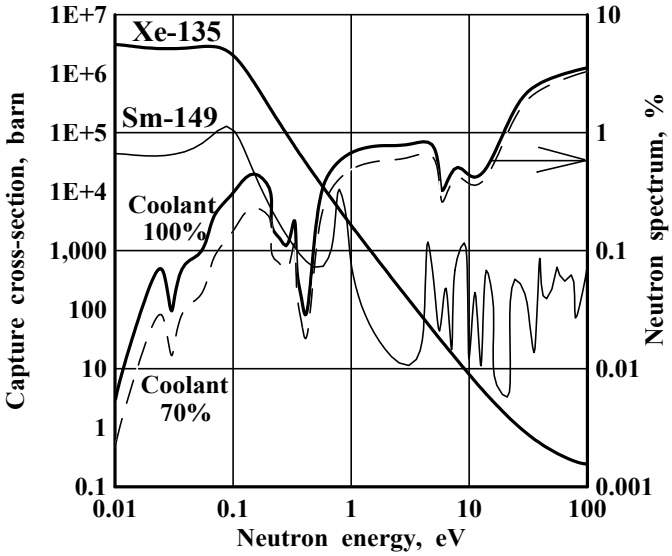

Fig.2 Energy dependencies of neutron radiative capture cross-sections for ^{135}Xe and ^{149}Sm; neutron spectrum of water thermal reactor at nominal and reduced density of coolant. Coolant: 25%D_2O+75%H_2O, fuel: 7.1%^{232}U+4%^{231}Pa+4.6%^{233}U+84.3%^{232}Th, time: in 70 days after the reactor start-up.

Similar growth of CDRE is observed in a light-water reactor loaded with traditional uranium dioxide fuel but it does not play a decisive role here because CDRE remains significant in value, and negative (see Figure 3).

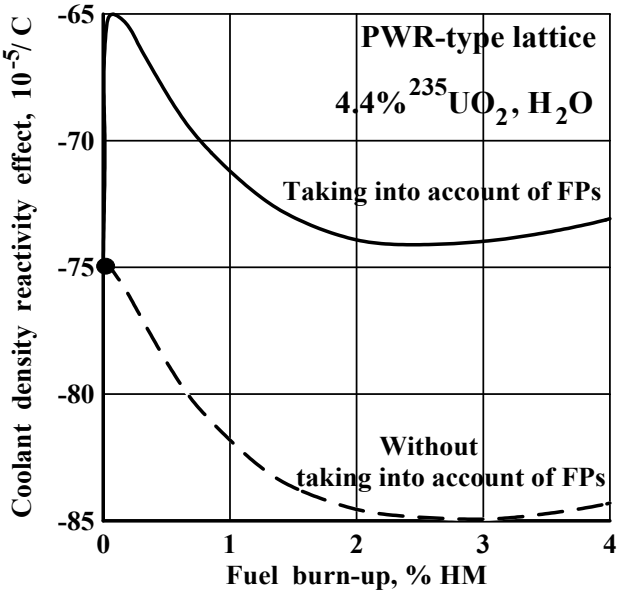

Fig.3 Coolant density reactivity effect with and without account of fission products as a function of fuel burn-up in conventional VVER-1000-type light-water power reactor.

Figure 4 demonstrates the dependencies of CDRE on fuel burn-up for different contents of heavy water in the water coolant and for the fuel composition which is able to ensure zero values for initial CDRE and time-integrated CDRE over the period of the fuel campaign. It is seen that if heavy water fraction in the reactor coolant increases, maximal values of positive

CDRE become lower. This can be explained by hardening the neutron spectrum, where the poisoning effect of ^{135}Xe and ^{149}Sm appears in a lower degree.

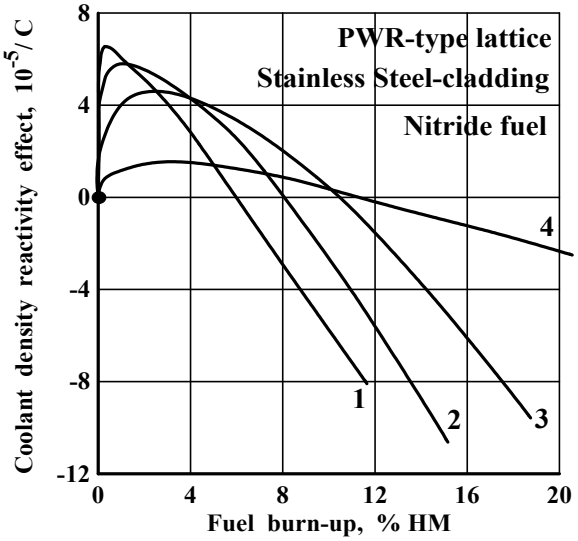

Fig.4 Dependencies of coolant density reactivity effect on fuel burn-up for different fuel and coolant. (Meanings of 1, 2, 3, 4 – see in Figure 1)

Fresh fuel compositions, ensuring the utmost possible fuel burn-up and these values of fuel burn-up are presented in Figure 5 as functions of heavy water fraction in the reactor coolant. It is seen that fuel burn-up increases linearly along with growth of heavy water fraction in the reactor coolant up to 75%, then fuel burn-up increases slower.

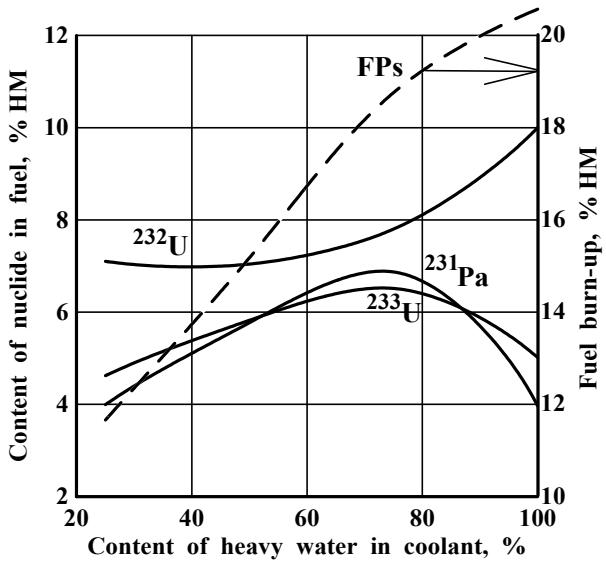

Fig.5 Fresh fuel composition (Th is not shown) that provides the following values: neutron multiplication factor in infinite medium $K_\infty = 1.1$; zero values of initial coolant density reactivity effect and time-integrated (over the reactor campaign) coolant density reactivity effect for different heavy water contents in water coolant; maximal value of fuel burn-up.

5. Advantages of modified fuel

The following benefits may be derived from using fuel with stabilized neutron multiplying properties up to high fuel burn-up. Primarily, it drastically decreases the technological procedures related to fuel fabrication, fuel transportation and refueling (per 1 MW*day energy produced). So, for example, the number of procedures, as mentioned above, will be less by a factor of 2-5, if traditional fuel of light water reactor with a burn-up of 4-6% HM is replaced with a fuel with a burn-up of 12-20% HM. In addition to economical benefits, a reduction in the number of fuel reloads during reactor life-time will result in a substantial reduction of the potential possibility of switching fissile materials from civilian to military purposes. It enhances proliferation resistance of the nuclear fuel cycle. If, in addition, power density in the reactor core is reduced by a factor of 2-3, then the reactor with high fuel burn-up is able to operate a full reactor life-time (15-40 years) without refueling. Such reactors may be supplied to developing countries as "black boxes" with minimal danger of nuclear proliferation, because all operations with nuclear fuel may be concentrated just at the fuel regeneration plants. Also, such reactors will be simpler in manufacture and operation because they are equipped with tools for operative reactivity control only. The equipment needed for fuel reloading, cooling and transportation will not be installed at the nuclear power plant.

Another factor related to improvement of fuel proliferation protection is the build-up of ^{232}U in fuel under irradiation, which can prevent nuclear fuel from switching to military purposes. This is explained by increased and long-term heat generation due to α-decay of ^{232}U (830 W/kg, $T_{1/2}$=69 years) as well as by hard γ-radiation of daughter products of its decay. Figure 5 shows that fresh fuel contains even more ^{232}U than ^{233}U. This means that the specific decay heat generation of uranium, containing the proliferation-attractive fissile nuclide ^{233}U, is at a level of 450-500 W/kg, i.e. greater by a factor of 45 than that of reactor-grade plutonium (10.5 W/kg) and by a factor of 200 than that of weapons-grade plutonium (2.3 W/kg) [10]. Such a level of decay heat generation in uranium makes it substantially complicated (or impossible) to apply uranium for military purposes. At that fuel heat generation is about 60 W/kg. This level of heat generation and hard γ–radiation demand using remote technology for fuel processing. Contents of main fuel nuclides as functions of fuel burn-up are presented in Figure 6 for a 25%-fraction of heavy water in the reactor coolant. It is seen that the concentration ratio of ^{232}U and ^{233}U varies insignificantly over the full life-time of the reactor fuel. This means that the level of uranium heat generation is kept high at any time moment of reactor operation, including heat generation of spent fuel.

In the case of a closed nuclear fuel cycle, the use of fuel with high burn-up also leads to a substantial reduction in chemical reprocessing activities. So, for example, with fuel burn-up in light water reactor at a level of 5% HM, about 95% of non-burnt fuel is directed to chemical reprocessing, i.e. a 20-fold recycling is required for burn-up of a fuel load. If fuel burn-up is at a level of 12-20% HM, then only a 5-8-fold recycling is required, i.e. demands for radiochemical reprocessing activity can be reduced by a factor of about 3-4.

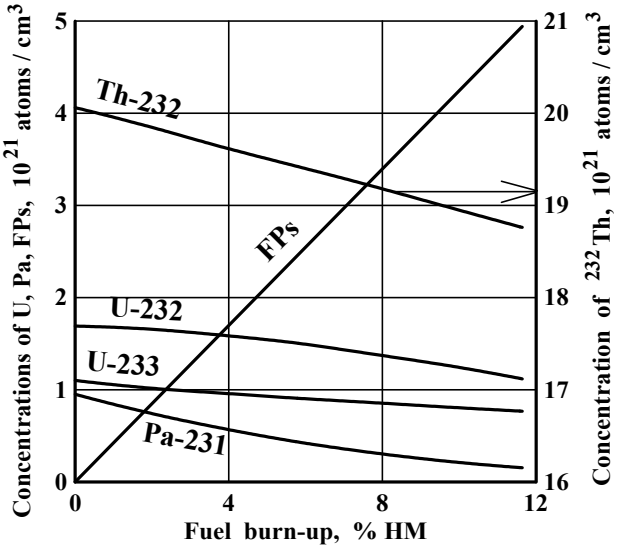

Fig.6 Variation of fuel composition as a function of fuel burn-up when coolant: 25%D_2O+75%H_2O, fuel: 7.1%^{232}U+4%^{231}Pa+4.6%^{233}U+84.3%^{232}Th.

Another positive aspect of high fuel burn-up should be noted. Under neutron irradiation of fuel, not only the processes of fission product generation take place; other processes include partial neutron transmutation caused by natural radioactive decay and radiative neutron capture reactions of fission products. Of course, the higher the fuel burn-up, the larger the content of fission products in fuel. However, it seems reasonable to compare amounts of fission products per unit of generated energy. The evaluations presented in Table 1 show that specific accumulation of the most dangerous long-lived fission products in fuel burnt up to 15% HM ($m_{15\%}$) is lower by 5-15% than that in the fuel burnt up to 4.2% HM ($m_{4.2\%}$)

Table 1 Accumulation ratio of fission products (per unit of generated power) for fuel burn-up of 15% HM and 4.2% HM

Nuclide	^{90}Sr	^{135}Cs	^{137}Cs	^{129}I	^{99}Tc	^{107}Pd	^{126}Sn	^{79}Se	^{93}Zr
$m_{15\%}/m_{4.2\%}$	0.90	0.86	0.91	0.90	0.84	0.90	0.94	0.94	0.92

6. Conclusion

The paper considers the neutron-physical characteristics of a thermal power reactor with prolonged life-time of thorium-based fuel containing isotopes ^{231}Pa, ^{232}U and ^{233}U. It is demonstrated that, in such a reactor, a possibility exists, in principle, to reach high (12-20% HM) fuel burn-up both from the point of view of stable neutron multiplying properties and from that of safe reactor operation in terms of slightly negative coolant density reactivity effect. It was assumed that the reactor coolant consists of light water and heavy water with contents of the latter from 25% and above.

The results are obtained from calculations based on application of the Japanese evaluated nuclear data library JENDL-3.2 for generation of micro cross-sections for isotopes ^{231}Pa and ^{232}U. The values of fuel burn-up obtained with application of JENDL-3.2 are significantly lower than those obtained with application of the American evaluated nuclear data file ENDF/B-IV. Therefore, it seems reasonable to re-evaluate nuclear data of ^{232}U and the ill-studied ^{231}Pa.

It has been demonstrated that, if heavy water fraction in reactor coolant increases, burn-up of thorium-based fuel containing essential amounts of ^{231}Pa and ^{232}U will increase also. This fact can be explained by the following considerations. Introduction of heavy water in light water coolant of the reactor shifts the neutron spectrum towards a range of higher energies where neutron multiplying properties of fuel are better. The harder neutron spectra can also be attained by other ways, such as in tighter lattices of fuel rods and the application of super-critical water parameters with reduced coolant density. Introduction of enriched uranium into fuel composition could favorably alter the CDRE value and, thus, additionally increase fuel burn-up. Effectiveness of the above ways will be evaluated in further studies. A combination of these methods could make it possible to reach even higher fuel burn-up.

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