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FUEL CYCLES WITH HIGH FUEL BURN-UP: ANALYSIS OF REACTIVITY COEFFICIENTS*)

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Abstract

Fuel cycles of light-water reactors (LWR) with high fuel burn-up (above 100 MWd/kg), as a rule, involve large amounts of fissionable materials. It leads to forming the neutron spectrum harder than that in traditional LWR. Change of neutron spectrum and significant amount of non-traditional isotopes (for example, ^{237}Np, ^{238}Pu, ^{231}Pa, ^{232}U) in such fuel compositions can alter substantially reactivity coefficients as compared with traditional uranium-based fuel. The present work addresses the fuel cycles with high fuel burn-up which are based on Th-Pa-U and U-Np-Pu fuel compositions. Numerical analyses are carried out to determine effective neutron multiplication factor and void reactivity coefficient for different values of fuel burn-up and different lattice parameters. The algorithm is proposed for analysis of isotopes contribution to these coefficients. Various ways are considered to upgrade safety of nuclear fuel cycles with high fuel burn-up.

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Introduction

In development of nuclear fuel cycles (NFC) with high fuel burn-up, the fuel compositions involving non-traditional isotopes (^{237}Np, ^{238}Pu, ^{231}Pa, ^{232}U) and high concentration of fissionable materials are often analyzed [1-3]. Usually, calculations of NFC parameters are carried out for traditional LWR lattices, for example, VVER-lattice in Ref. 1 and PWR-lattice in Ref. 2.. Such a selection of traditional lattices is proved by technological reasons but it can be non-optimal from other points of view.

In the present work there are considered the NFC which are potentially able to reach high burn-up of fuel based on Th-Pa-U and U-Np-Pu compositions. Numerical analyses are carried out to determine neutron multiplication factor for infinite lattice of fuel elements (infinite neutron multiplication factor) under various fuel-to-moderator ratios and various values of fuel burn-up. The computations are performed with application of the computer code packages SCALE [4] and MCNP [5].

For the fuel compositions to be analyzed, dependence of neutron multiplication factor on moderator quantity differs substantially from that of traditional uranium fuel with enrichment below 5% ^{235}U. Under fixed lattice parameters, it is possible for positive values of void reactivity coefficient (VRC) to appear. Here and further, VRC is understood as a value $\Delta k_{void}=k(\rho)-k(\rho_0)$, where ρ - reduced coolant density, ρ_0 – coolant density in nominal operation conditions. It should be noted that presence of resonance absorbers in fuel compositions causes negative value of Doppler reactivity coefficient within full range of the working temperatures and for full campaign of the reactor core. So, safety of such systems is mainly determined by VRC. Positive values of reactivity coefficients are unacceptable from standpoint of safe reactor operation. Therefore, possible reasons for appearance of positive VRC are analysed, and the ways are proposed to overcome or remove these reasons.

The ways to reach high fuel burn-up

Problem of LWR fuel burn-up increasing was a high priority issue always. The last twenty years, mean fuel burn-up in PWR gradually increases, and maximal value of fuel burn-up has already reached 70-80 MWd/kg. Such an increase is reached now by higher initial fuel enrichment. To compensate large value of initial reactivity margin, gadolinium-based compounds are usually used as a burnable poison. Increase of initial fuel enrichment in traditional U-Pu fuel reduces fuel breeding ratio that adversely affect total efficiency of NFC. Introduction of burnable poisons (gadolinium, for instance) into fuel composition binds large initial margin of positive reactivity. However, after rapid burning of such an absorber, excess reactivity has to be bound by traditional methods: control rods and addition of boron into light-water coolant.

Let’s define neutronic and physical requirements to be satisfied for achievement of fuel burn-up above 150 MWd/kg (hereinafter, we call such a fuel burn-up as a “high” fuel burn-up):

- Significant share of fission reactions must occur with the isotopes generated in fuel from fertile nuclides. Ratio of fission reactions with these isotopes to total number of fission reactions for whole reactor campaign is used here to characterize this process.
- High value of fuel burn-up must not result in high reactivity of a fuel cell at any time moment.
- Reactivity coefficients (in respect of void, density, temperature, etc.) must satisfy conditions of safe reactor operation at any time moment, i.e. they have to be negative.

In our opinion, a condition of principle is an active utilization of fertile nuclides in the process of fuel burn-up. Ideal NFC from standpoint of maximal fuel burn-up may be the breeding mode of fuel

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burn-up when neutron multiplication factor is kept constant with time. Saying by another words, positive reactivity related with appearance of new fissionable nuclei must compensate, at any time moment, negative reactivity related with disappearance of fissionable nuclei and accumulation of fission products. The conditions defined above can be written in form of formulae (1) and (2) in terms of one-group cross-sections and concentrations of heavy nuclides:

$$\frac{(n_f \cdot s_f)^1 \cdot r^1}{s_{\tilde{n}}^{\,2} \cdot r^2 + s_{\tilde{n}f}^{\,1} \cdot r^1} \geq 1 \qquad (1)$$

$$\frac{(n_f \cdot s_f)^1 \cdot (r^1 - (\frac{\Delta r^1}{\Delta t}) \cdot \Delta t) + (n_f \cdot s_f)^3 \cdot (\frac{\Delta r^3}{\Delta t}) \cdot \Delta t}{s_{\tilde{n}}^{\,2} \cdot (r^2 - (\frac{\Delta r^2}{\Delta t}) \cdot \Delta t) + s_{\tilde{n}f}^{\,1} \cdot (r^1 - (\frac{\Delta r^1}{\Delta t}) \cdot \Delta t) + s_{cf}^{\,3} \cdot (\frac{\Delta r^3}{\Delta t}) \cdot \Delta t + \tilde{s}_c^{\,fp} \cdot (\frac{\Delta r^{fp}}{\Delta t}) \cdot \Delta t} \geq 1 \qquad (2)$$

where the indices designate as follows: 1 – initial fissionable isotope, 2 – fertile nuclide, 3 – produced fissionable nuclide, fp – effective fission product.

These formulae can be used in selecting the fuel compositions with high burn-up properties. However, certain iterative process should be arranged because one-group cross-sections depend on neutron spectrum that, in its turn, depends on fuel composition.

The last years, several concepts of LWR fuel cycles have been proposed where introduction of some “exotic” isotopes into traditional fuel composition led to satisfaction of conditions for reaching high fuel burn-up. In these concepts, the introduced heavy nuclide simultaneously plays two roles: burnable poison and fertile nuclide. Burning together with main fissionable nuclide, the introduced nuclide provides generation of new fissionable nuclei and keeps neutron multiplication factor at constant level. In the proposed cycles, large concentrations of heavy nuclides with high neutron capture cross-sections decrease significantly a negative role of accumulated fission products.

The results obtained in the studies have demonstrated that regime of high fuel burn-up under LWR conditions could be realized if, for example, fuel containing ^{231}Pa or ^{237}Np would be used. In this case, the chains of isotopic transformations with some remarkable properties will be realized. The first of all, nuclides ^{231}Pa and ^{237}Np are neutron absorbers practically within full neutron energy range of LWR. So, in these chains they play the roles of original “fertile elements” and, at the same time, burnable poisons. For example, it has been shown in Ref. 1 that fuel compositions containing ^{231}Pa can be characterized by the stabilized multiplying properties up to very high fuel burn-up (30% HM and above) under conditions of LWR operation.

Numerical results for neutron multiplication factor and reactivity coefficients

The following parameters should be chosen to analyze real NFC of LWR:

1. fuel composition;
2. design of fuel rod (fuel diameter, thickness of cladding);
3. lattice parameters (type of lattice, pitch of lattice).

Basing upon the ideas listed above, the following fuel compositions are selected for further analysis: (^{232}Th (69%)+ ^{231}Pa (15%)+ ^{233}U(16%))O_2 and (^{238}U (70%) + ^{237}Np (10%) + ^{239}Pu (20%))O_2. Triangular lattice of fuel rods is considered. Inner diameter of fuel rod - 0,772 cm, outer diameter - 0,9164 cm. Lattice pitch - 1.275 cm. These data correspond to parameters of Russian reactor VVER-1000 [6].

To provide a possibility of reaching high fuel burn-up with the analysed fuel types, stainless steel is chosen as a cladding material. The computations are carried out with application of the computer code package SCALE (module SAS2H) which is especially developed for analyses of LWR fuel cycles [4]. The dependencies of infinite neutron multiplication factor on fuel burn-up are presented in Fig. 1.

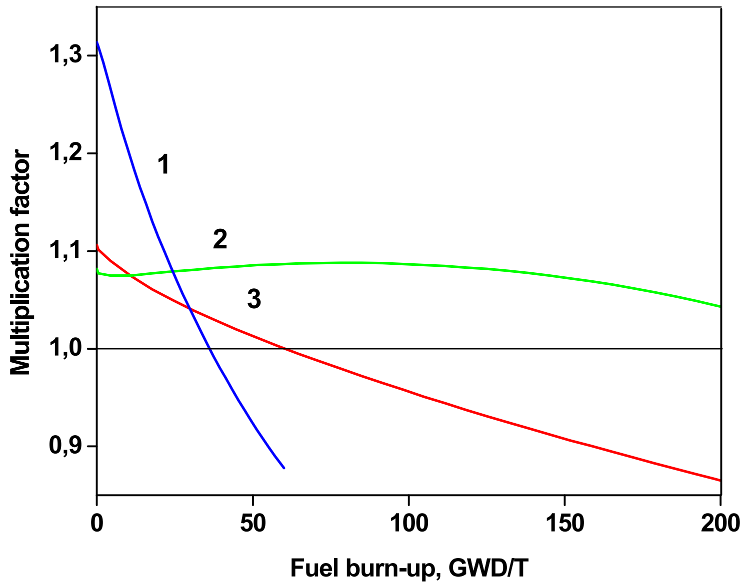

Fig. 1. Dependence of neutron multiplication factor in a fuel cell on fuel burn-up:
1 – UO_2 (4,4% ^{235}U);
2 – (^{232}Th (69%)+ ^{231}Pa (15%)+ ^{233}U(16%))O_2;
3 - (^{238}U (70%) + ^{237}Np (10%) + ^{239}Pu (20%))O_2

It can be seen that curve of neutron multiplication factor for the fuel cell containing protactinium and neptunium isotopes has a more gently sloping form than that for traditional fuel cell. This fact makes it possible to reach high fuel burn-up (150 MWd/kg and above). However, since the proposed fuel compositions involve large amounts of isotopes with high neutron capture cross-sections in thermal energy range, a question arises on proper choice of the lattice pitch and on assessment of reactivity coefficients (void reactivity coefficient, in the first turn) in such systems.

Dependencies of cell neutron multiplication factor on pitch of triangular lattice are presented in Figs. 2-4. The graph inside of pin region corresponds to water deprivation in the tightest lattice, point at the pitch equalled zero corresponds to full drying of the tightest lattice.

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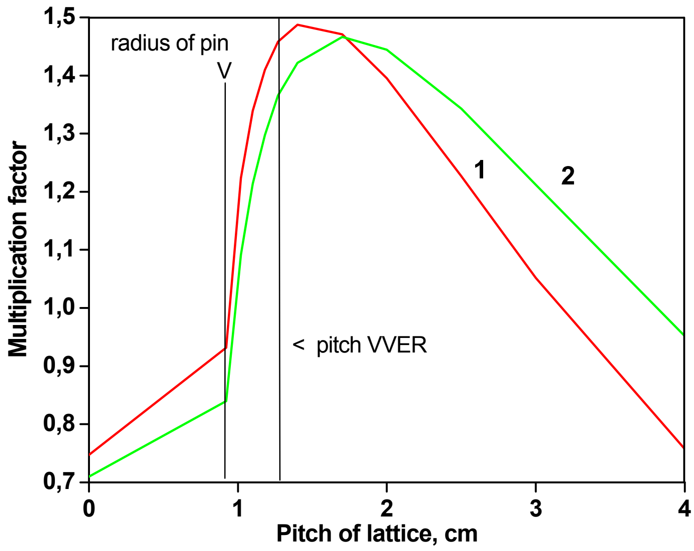

Fig. 2. Dependence of neutron multiplication factor for the cell containing UO_2-fuel (4,4% ^{235}U) on the lattice pitch: 1 – cold state; 2 – hot state.

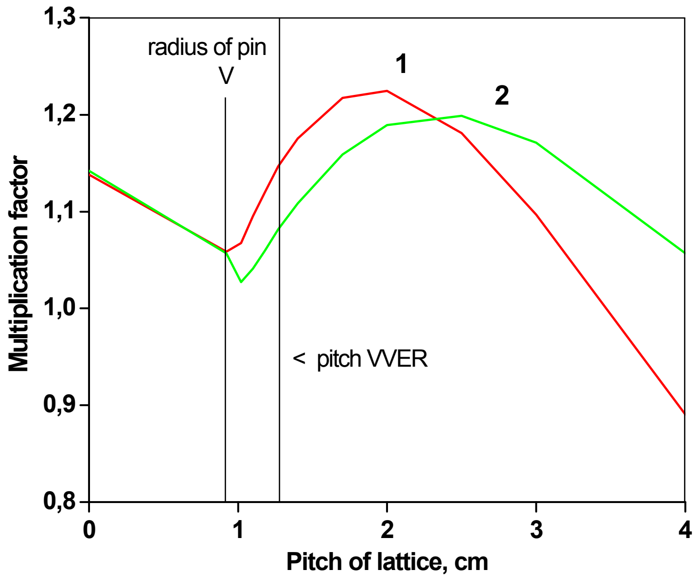

Fig. 3. Dependence of neutron multiplication factor for the cell containing (^{232}Th (69%)+ ^{231}Pa (15%)+ ^{233}U(16%))O_2-fuel on the lattice pitch: 1 – cold state; 2 – hot state.

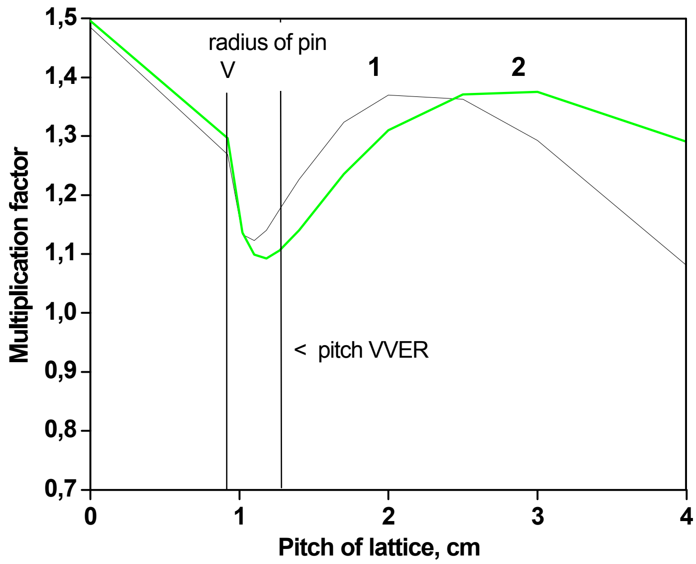

Fig. 4. Dependence of neutron multiplication factor for the cell containing (^{238}U (70%) + ^{237}Np (10%) + ^{239}Pu (20%))O_2-fuel on the lattice pitch: 1 – cold state; 2 – hot state.

It can be seen from Fig. 2 that, in case of traditional fuel composition:

1. pitch of VVER lattice meets the requirement on VRC negativity; pitch is near to optimal value in respect of initial reactivity margin;
2. increase of uranium-water ratio reduces initial reactivity margin while decrease of this ratio can lead to appearance of positive VRC;
3. drying of the tightest lattice leads to negative VRC.

Values of neutron multiplication factor for pitch of VVER-lattice are calculated with application of both computer codes: MCNP and SCALE. Good agreement of the values obtained in these two computations and good agreement with the values published in Ref. 6 demonstrate, in our opinion, an adequacy of the used mathematical models.

Analysis of Figs. 3, 4 and comparison with Fig. 2 lead to the following conclusions:

1. pitch of VVER-lattice is not optimal for these fuel compositions because reduction of uranium-water ratio in wide range leads to increase of initial reactivity margin while VRC remains negative;
2. insignificant increase of uranium-water ratio leads to appearance of positive VRC;
3. drying of the tightest lattice leads to appearance of positive VRC.

Further, contributions of different isotopes to VRC are analyzed, and possible ways to provide safe operation of nuclear reactors containing large amounts of heavy isotopes with high neutron capture and fission cross-sections are considered.

Algorithm for assessment of isotopic contributions to VRC

Two direct computations (for nominal and perturbed states of a fuel cell) are often used for determination of reactivity coefficients. When a perturbation is introduced into a system, neutron

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spectrum is changed and, as a consequence, rates of neutron reactions with fuel isotopes are changed too. Total effect of all these changes on neutron multiplication factor is defined by a sensitivity coefficient. Similar approach is used in the present work.

The following simple formula can be used for analysis of isotopic contributions to VRC value:

$$\Delta K_i = \frac{\nu\Sigma_f \cdot \Phi + \nu\Sigma_f{}^i \cdot \Delta\Phi}{\Sigma_a \cdot \Phi + \Sigma_a{}^i \cdot \Delta\Phi} - \frac{\nu\Sigma_f \cdot \Phi}{\Sigma_a \cdot \Phi}$$

where $\Sigma^i \cdot \Delta\Phi = \rho_i \int \sigma^i(E) \cdot (\Phi_2(E) - \Phi(E)) \cdot dE$ - variation of neutron reaction rate with isotope i; Φ_2(Å) – perturbed neutron spectrum.

Numerical algorithm based on the formula written above has been developed within the frames of the computer code package SCALE. Some results obtained in computations are presented in Table 1 and Table 2.

Table 1. Variation of neutron multiplication factor for 5%-reduction of water density in VVER cell: fuel - UO_2 (4,4% ^{235}U).

Isotope	ΔK_i
All isotopes	-0,015
^{238}U	0,004
^{235}U	-0,019

Table 2. Variation of neutron multiplication factor for 5%-reduction of water density in VVER cell: fuel - (^{232}Th (69%)+ ^{231}Pa (15%)+ ^{233}U(16%))O_2

Isotope	ΔK_i
All isotopes	-0,005
^{232}Th	0,002
^{231}Pa	0,013
^{233}U	-0,020

Table 3. Variation of neutron multiplication factor for 5%-reduction of water density in VVER cell: fuel - (^{238}U (70%) + ^{237}Np (10%) + ^{239}Pu (20%))O_2

Isotope	ΔK_i
All isotopes	0,006
^{238}U	0,004
^{237}Np	0,014
^{239}Pu	-0,013
^{240}Pu	0,001

As it is seen from the presented results, different isotopes give different contributions to the variation of neutron multiplication factor caused by change of coolant density. Comparison of these contributions enables us to predict variation of sensitivity coefficient in the process of fuel burn-up. The most «dangerous» isotopes for VRC are fertile nuclides with large radioactive capture cross-sections for thermal neutrons. For the fuel compositions under analysis these are isotopes ^{237}Np and ^{231}Pa. In the course of fuel burn-up, concentrations of these isotopes will decrease and, hence, their

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negative effect on VRC will decrease also. At the same time, accumulation of fission products and heavy transuranium isotopes can cause an adverse effect. To determine total VRC variations in the process of fuel burn-up, these coefficients are computed for different values of fuel burn-up. The results obtained in these computations are presented in Figs. 5-6: dependencies of neutron multiplication factor variations (in relative units) - $\Delta k(\rho)=k(\rho-\delta\rho)-k(\rho)$ for three values of fuel burn-up on coolant density (in relative units). Here, $\delta\rho$ - variation of coolant density around ρ value. In these computations, variation of coolant density is adopted as $0.1\rho_0$, where ρ_0 – coolant density under the working conditions.

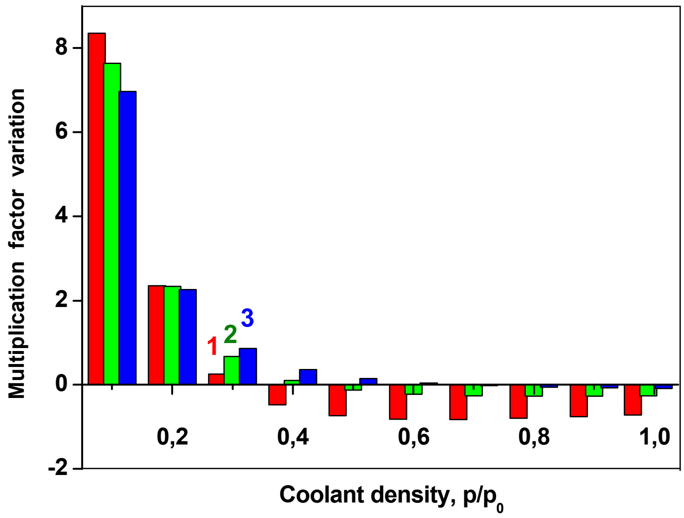

Fig. 5. Dependence of neutron multiplication factor variation on coolant density variation.
Fuel - (^{232}Th (69%)+ ^{231}Pa (15%)+ ^{233}U(16%))O_2 .
1 – fresh fuel; 2 – 50 MWd/kg; 3 – 100 MWd/kg.

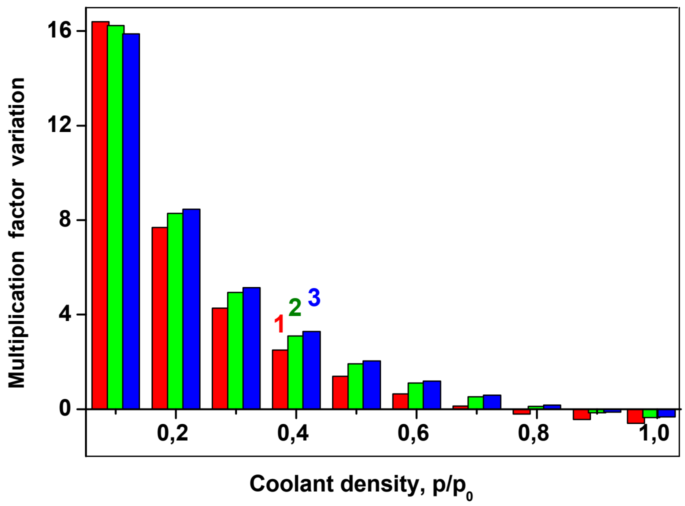

Fig. 6. Dependence of neutron multiplication factor variation on coolant density variation.
Fuel - (^{238}U (70%) + ^{237}Np (10%) + ^{239}Pu (20%))O_2 .
1 – fresh fuel; 2 – 50 MWd/kg; 3 – 100 MWd/kg.

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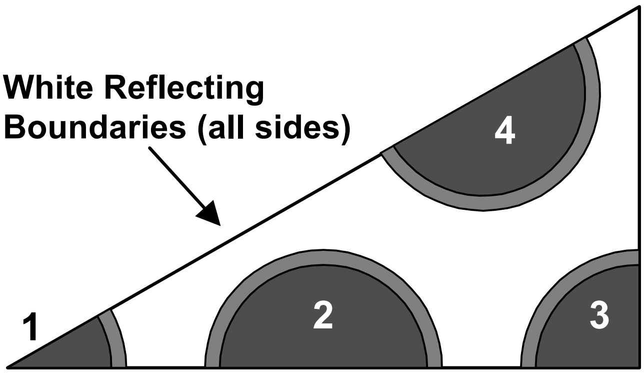
It is seen from Figs. 5-6 that VRC changes insignificantly in the process of fuel burn-up. The worst value of VRC, for a majority of density variation ranges, corresponds to fresh fuel. So, the approaches, we'll propose below for suppression of positive VRC in case of fresh fuel, will be even more effective for the partially burnt-up fuel.

Some ways to suppress positive VRC

Two ways to suppress positive VRC are considered in the present work. The first way implies reduction of the reactor core height for increasing axial neutron leakage in case of coolant density decreasing. The second way implies introduction of additional moderating material into lattice of fuel rods, and moderator remains in the reactor core under any values of coolant density. Introduction of various heterogeneous insertions into fuel assemblies is a contemporary tendency in improving fuel utilization and reaching desirable values of reactivity coefficients. For example, fuel rods containing gadolinium compounds are planned to use in new fuel assemblies of VVER-type reactors [7].

All the computations are carried out with application of the computer code MCNP for the model depicted in Fig. 7. The model represents a triangular prism of finite height containing four fuel rods with a coolant in between. Boundary conditions are as follows: conditions of symmetry are set on lateral sides of the prism, and vacuum conditions are set on axial sides. Sizes of fuel rods and compositions of materials are the same we used previously in the cell calculations. Centers of fuel rods are located in nodes of triangular mesh. So, if distance between centers of fuel rods is equal to VVER pitch, then the model completely corresponds to a fuel cell, if all fuel rods are identical ones.



Fuel parameter
Fuel pin radius = 0.386 cm
Pin pitch = 1.275 cm
Average Linear Power = 152 W/cm
Fuel density = 8,79 g/cm^3

Fig. 7. Geometrical model for three-dimensional calculations.

The results obtained in calculations for dependence of neutron multiplication factor on coolant density for the model containing different fuel types are presented in Figs. 8-9. Two values of the reactor core height (3 m and 1 m) and two values of the lattice pitch (1,275 cm and 2 cm) are used in these computations. Designation of the curves shown in Figs. 8-9 is presented in Table 4. Three-meter height corresponds to height of VVER core while one-meter height is considered as an extreme option. Pitch of 1,275 cm corresponds to pitch of VVER core while 2-cm pitch was selected after analysis of neutron multiplication factor dependence on the lattice pitch (these dependences are shown in Figs. 3-4). The only purpose of these computations was to reveal tendencies in VRC variation under variations of height and the lattice pitch, not optimization of the lattice parameters for each fuel type.

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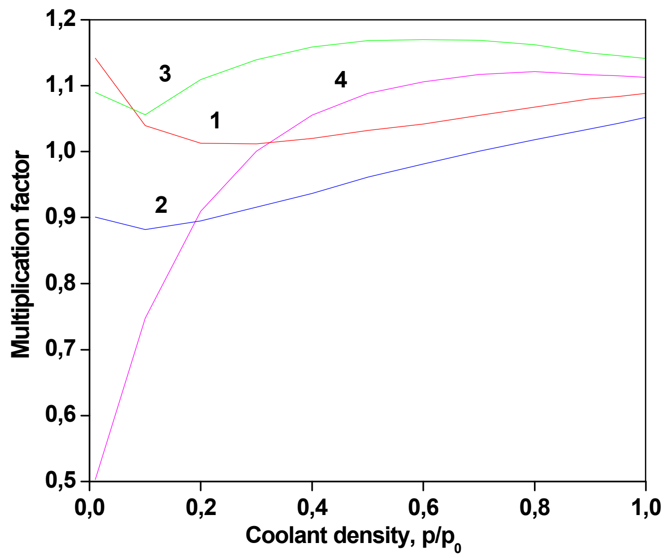

Fig. 8. Dependence of neutron multiplication factor on coolant density for the cell with different heights and the lattice pitches. Fuel - (^{232}Th (69%)+ ^{231}Pa (15%)+ ^{233}U(16%))O_2 .

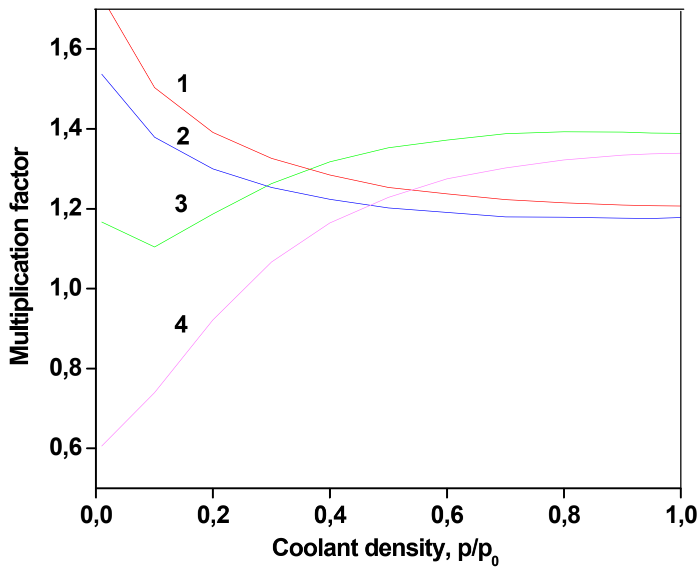

Fig. 9. Dependence of neutron multiplication factor on coolant density for the cell with different heights and the lattice pitches. Fuel - (^{238}U (70%) + ^{237}Np (10%) + ^{239}Pu (20%))O_2 .

Table 4. Designation of the curves presented in Figs. 8-9.

Lines	Core height, m	Pitch, cm
1	3	1,275
2	1	1,275
3	3	2
4	1	2

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The results obtained in calculations of neutron multiplication factor for the modified model (fuel rod 2 is replaced by graphite rod with radius RC) are presented in Figs. 10-11. Two values of graphite rod radius RC (0,386 cm and 0,55 cm) are used in these calculations. Such a modification of fuel assembly is carried out for 3-m height of the reactor core. Two values of the lattice pitch (1,275 cm and 2 cm, like in previous calculations) are used here. Designation of the curves shown in these figures is presented in Table 5.

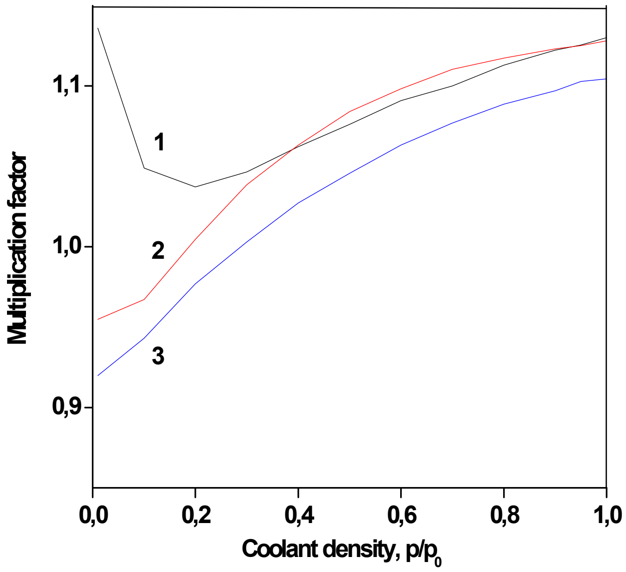

Fig. 10. Dependence of neutron multiplication factor on coolant density for the cell with a graphite rod. Fuel - (^{232}Th (69%)+ ^{231}Pa (15%)+ ^{233}U(16%))O_2 .

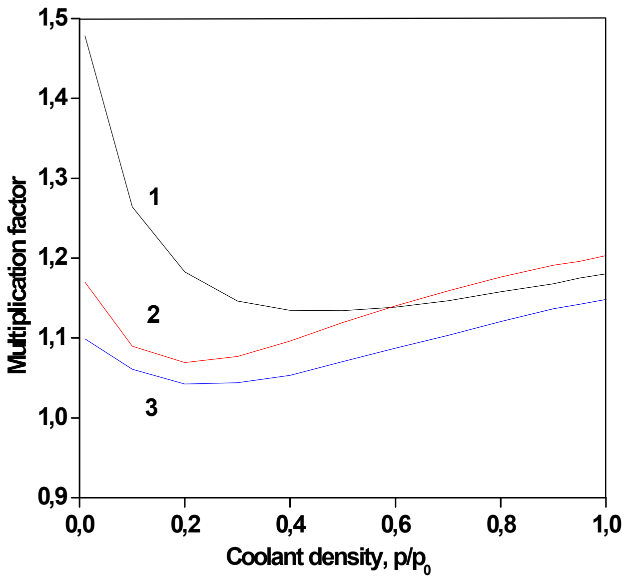

Fig. 11. Dependence of neutron multiplication factor on coolant density for the cell with a graphite rod. Fuel - (^{238}U (70%) + ^{237}Np (10%) + ^{239}Pu (20%))O_2 .

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Table 5. Designation of the curves presented in Figs. 10-11.

Lines	Core height, m	RC, cm
1	3	-
2	3	0,386
3	3	0,55

It is seen from Figs. 8-11 that VRC values for both fuel types considered here can be made negative for any ranges of coolant density variations.

Conclusions

So, the results obtained in this study have demonstrated that:

1. Non-traditional fuel compositions developed for achievement of high fuel burn-up in LWR can possess positive values of reactivity coefficients that is unacceptable from the reactor operation safety point of view.
2. The lattice pitch of traditional LWR is not optimal for non-traditional fuel compositions. The increased value of the lattice pitch leads to larger value of initial reactivity margin and provides negative VRC within sufficiently broad range of coolant density.
3. Fuel burn-up has an insignificant effect on VRC dependence on coolant density. So, the measures undertaken to suppress positive VRC of fresh fuel will be effective for partially burnt-up fuel compositions also.
4. Increase of LWR core height and introduction of additional moderators into the fuel lattice can be used as the ways to reach negative VRC values for full range of possible coolant density variations.

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