The Lead-Cooled Fast Reactor Transition to Equilibrium Operating Conditions

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Abstract. Lead-cooled fast reactors are a promising direction in the development of nuclear energy because of broad opportunities to ensure their safety and security. Efforts are underway to develop lead-cooled fast reactors BREST-OD-300 and BREST-1200 in Russia. Nitride fuel is expected to be used at the core of these reactors. High density of this fuel gives an opportunity to create a core with the breeding ratio close to unity. This makes it possible to realize the equilibrium operating conditions with feeding of regenerated fuel only depleted uranium. The reactivity change between the refueling does not exceed the effective fraction of delayed neutrons in this case.

The use of plutonium accumulated earlier and newly emitted plutonium in the processing of thermal reactors irradiated fuel is assumed as initial core charge. In addition the initial charge of enriched uranium is considered. The stage of reactor operation, in which the transition of the fuel composition from start-up to equilibrium takes place, may be accompanied by a number of problems associated, in particular, with the need to ensure a minimum change of the reactivity between the refueling. The report summarizes the features of work in the reactor at this stage at different compositions of the initial charge. Criteria of reaching the equilibrium operating conditions are proposed, with due regard to which the transition time for all variants of the initial charge of plutonium obtained from VVER reactor spent fuel.

Key Words: Lead-cooled fast reactor, evolution of plutonium isotopic composition, equilibrium operating conditions.

1. Introduction

One of the promising directions in the development of fast reactors is lead-cooled reactors [1]. In Russia, efforts are underway to build the BREST-OD-300 demonstration lead-cooled fast reactor and to develop the BREST-1200 power reactor. A distinctive feature of the BREST type reactors is the principle of «natural safety», which, according to [2], imposes the following requirements to the nuclear power engineering of the future:

- the elimination of severe accidents at reactors and accidents at nuclear fuel cycle enterprises;

- radiation-equivalent disposal of radioactive waste;
- technological reinforcement of the nonproliferation regime.

Elimination of severe nuclear accidents is achieved, first, by the use of the dense nitride fuel, which makes it possible to construct a core with the breeding ratio close to unity and with the variation of reactivity $\Delta \rho$ between refueling (taking into account the neptunium effect) that does not exceed the effective fraction of the delayed neutrons β_{eff} . The condition $\Delta \rho \leq \beta_{eff}$ is

important for a reactor implementing the principle of natural safety and it must be fulfilled throughout the whole lifetime [2]. There are two main operation stages for reactor types of BREST. The first stage is transitional in which the isotopic and elemental composition of the fuel can vary significantly, and the second stage is equilibrium. The equilibrium operating condition is a condition at which the composition and mass of plutonium and minor actinides in the charged and discharged fuel remain practically constant. Let us assume that equilibrium operating conditions are achieved when the change of the isotopic fuel composition per micro-campaign results in the change of reactivity margin that does not exceed β_{eff} .

The initial charge composition strongly affects the duration of the transition stage and the necessary reactivity margin. High-energy plutonium contained in spent fuel of VVER reactors is perceived as the main source of fissile materials for fast neutron reactors [3]. In addition, a possibility to use plutonium from spent fuel of high-power pressure-tube reactors (RBMK) for initial charge is worthy of consideration. In papers [3,4], aside from plutonium, enriched uranium is considered as initial fuel for fast reactors. In particular, it is stated that the use of enriched uranium will accelerate the development of nuclear power engineering.

The present research focuses on the criteria of reaching the equilibrium operating conditions in reactors of BREST-1200 type at different compositions of initial fuel. To assess the characteristics of equilibrium operating conditions, initial charge with plutonium obtained from the VVER spent nuclear fuel with a cooling period of 20 years is considered. For all variants of initial fuel composition, time-to-equilibrium has been assessed. Changes of neutron physical parameters of a reactor are discussed.

2. Problem setting and calculation procedure

Similarly to [5], a model scheme of a core with tetrahedral caseless fuel rod arrays and a lead reflector (*see FIG. 1.*) is considered. Uranium nitrides and plutonium are used for the initial charge. In the initial charge with plutonium obtained from the RBMK spent nuclear fuel, plutonium concentration in the fuel is 17.5% by heavy atoms, whereas in the charge with plutonium obtained from the VVER spent nuclear fuel, plutonium and minor actinides concentration is 14.2% by heavy atoms. Typical compositions of plutonium contained in spent nuclear fuel of VVER and RBMK reactors are provided, for example, in [6]. Fuel density, averaged over the fuel column, is assumed to be 12 g/cm³.

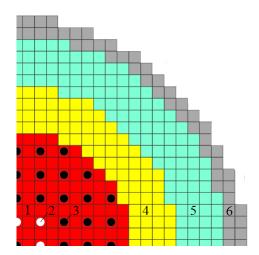


FIG. 1. Layout of core elements (1/4th): 1 – standard fuel rod array of the central region, 2 – automatic controller, 3 – emergency protection, reactivity compensator, 4 – standard fuel rod array of the fringe region, 5 – lead reflector, 6 – steel reflector

The main results of neutron physical calculations have been obtained using the MCM-based PRIZMA spectral code [7]. For initial charges with plutonium obtained from the RBMK spent nuclear fuel, the diffusion code "Arktika" [8] with the ROSFOND library [9] of assessed neutron data has been used. The kinetics of nuclide compositions has been calculated using the RISK [10] code that tracks the evolution of about 1,500 isotopes. After each micro-campaign, the change of isotopic fuel composition in the external fuel cycle has been calculated.

Calculations have been performed with the following assumptions:

• the reactor operates in the mode of partial refueling with the replacement of one-fifth of the fuel rod array with the deepest burnup by fresh fuel rods;

- the micro-campaign duration is one year (300 eff. days);
- the external fuel cycle duration is three years;
- the lifetime of a reactor is 60 years.

• during a micro-campaign, the reactor thermal capacity is constant and equals 2800 MW.

• during the first three refueling procedures the core us charged with the fuel of the same composition as at the start-up;

• there are no FM losses during processing; the fuel composition is cleared of 90% of fissile products during reprocessing, 90% of curium is removed, while all the remaining actinides are preserved;

• the NFC procedures allow fuel fabrication with arbitrary isotopic composition;

• neutron physical characteristics have been calculated with the control elements completely removed from the core;

• throughout the whole period of operation, the condition $k_{eff} \ge 1$ is maintained at the core. The deficiency of fissile materials, if any, is compensated by enriched uranium. In case of excessive FM, a part of regenerated fuel is replaced by depleted uranium;

• in calculations of the neutron physical characteristics, the temperature field in the core is stationary.

3. Initial charge with plutonium obtained from the VVER spent nuclear fuel. The criteria of reaching the equilibrium operating conditions

The considered core scheme is intended for a reactor with a fuel composition close to equilibrium. An example of such a composition is a mixture of uranium nitrides and plutonium obtained from spent nuclear fuel of VVER reactors after a cooling period of 20 years. For such a charge, throughout the whole period of operating life, the reactivity between two consecutive refueling procedures does not exceed $\beta_{eff} \approx 0.0035$. The breeding ratio, calculated as a relation of nucleation rate of fissile nuclei to their leak rate at a given moment of time averages 1.08 (*see FIG. 2.*).

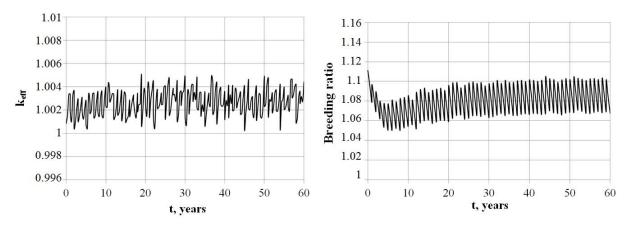


FIG. 2. The change of eigenvalue for the conventionally critical problem, and the breeding ratio during the reactor operation

Variation of the breeding properties of the fuel is predominantly determined by variation of plutonium isotope concentrations in the fuel. ²³⁹Pu concentration in the core after the fourth micro-campaign reveals only a slight change, reaching by the 17th micro-campaign its maximum value of ~9.25% by heavy atoms and then gradually decreases up to ~9.19% by heavy atoms. by the end of the operating life. The evolution of the plutonium isotopic composition for the most part pertains to ²⁴⁰Pu accumulation (*see FIG. 3.*), whose concentration after the first micro-campaign is 3.14% increases by the end of the operating life up to 4.04%. Variation of ²³⁸Pu, ²⁴¹Pu, and ²⁴²Pu concentrations and their respective breeding ratio are provided in *FIG. 4*.

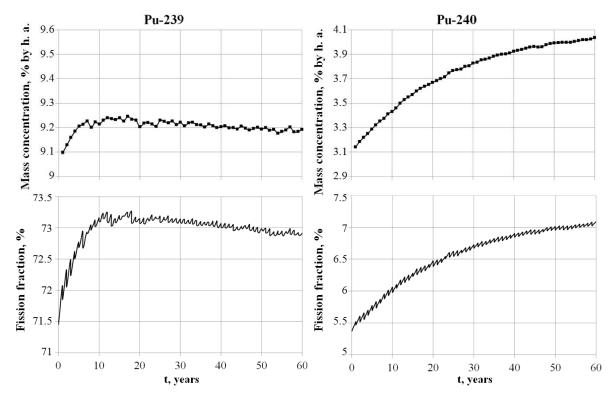


FIG. 3. Variation of 239Pu and 240Pu concentrations and their fission fractions

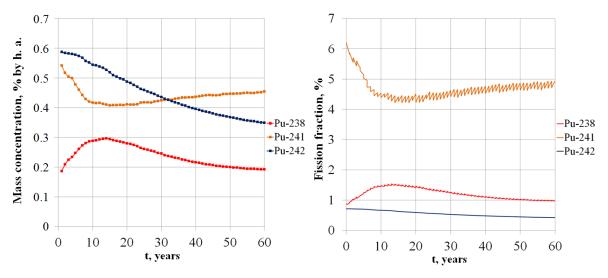


FIG. 4. Variation of 238Pu, 241Pu, and 242Pu concentrations and their fission fractions

The evolution of plutonium isotopic composition has a significant influence on its breeding properties. Plutonium mass concentration (without minor actinides) at the initial moment of time is 13.3%, whereas at the end of operating life it is 14.2%. The reactor criticality and relatively small reactivity excursions during a micro-campaign are reached via adjustment of charged fuel composition at each refueling. If the fuel composition is not adjusted and if the fuel with plutonium mass concentration of 14.2% is loaded into the core at the beginning of each micro-campaign, then at the startup its breeding properties will differ from the equilibrium composition by $\sim 7\beta_{eff}$, and will become comparable with β_{eff} after the 26th micro-campaign (*see FIG. 5.*).

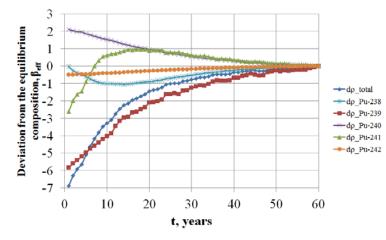


FIG. 5. Deviation of plutonium breeding properties from the equilibrium composition and contributions of specific isotopes into this deviation

Note that for all the variants of initial charges under consideration plutonium isotope concentrations in the fuel in the operating reactor tend to the same stationary values. Consequently, let us assume that for the fuel cycle under consideration and the chosen core configuration, the equilibrium fuel composition does not depend on the initial charge composition. Let us take the core properties at the end of the reactor lifetime as equilibrium operating conditions.

At the equilibrium operating conditions, about 86% of all fissions pertain to plutonium isotopes. At that, the contribution of ²³⁹Pu isotope is about 73%. ²³⁹Pu concentration in the

equilibrium fuel composition is approximately 9.2% by heavy atoms; ²³⁹Pu mass fraction in plutonium is 64.6%. The relative deviation of ²³⁹Pu mass concentration in the core after the fourth micro-campaign does not exceed 0.6% of the assumed equilibrium value. Such a deviation introduces a perturbation into the fuel breeding properties at the level of β_{eff} . Let us conventionally assume that for the remaining variants of initial charges, the fuel composition becomes equilibrium when ²³⁹Pu concentration in the fuel deviates from the stationary (equilibrium) value by no more than 0.6%. The average fuel burnup depth by heavy atoms is ~ 7.5%.

4. Initial charge with plutonium obtained from the RBMK spent nuclear fuel

Plutonium from the RBMK spent nuclear fuel contains a great number of even-even isotopes, whereupon it is characterized by considerable background radiation and high power density. Fuel production from such plutonium will be the most complex from the technical viewpoint, and, accordingly, the most expensive. In this connection, fuel loading with plutonium of RBMK spent nuclear fuel is not considered as the basic one, although it has certain advantages as compared to the other variants. As a source fissile material for the initial fuel charge we have selected plutonium obtained from the RBMK spent nuclear fuel with the burnup of 30 GW·day/ton of U and with a cooling period of 7 years.

Due to high concentration ²⁴⁰Pu isotope in plutonium, the initial breeding ratio is ~1.3, which is considerably higher than at the equilibrium operating conditions. Hence, the difference of reactivity margin during the first micro-campaign is $\Delta \rho \approx 1.17\beta_{eff}$ ($\beta_{eff} \approx 0.38\%$). During the three following micro-campaigns the value of k_{eff} decreases and from this point on it yields ($0.3 \div 0.6$) β_{eff} . The breeding ratio gradually decreases, and approximately in 40 years it averages 1.08, just like in the equilibrium fuel composition (*see FIG. 6.*).

Thus, the reactor operation can be conventionally considered as equilibrium, except for the first micro-campaign. At that, the core us characterized by a relatively high breeding ratio.

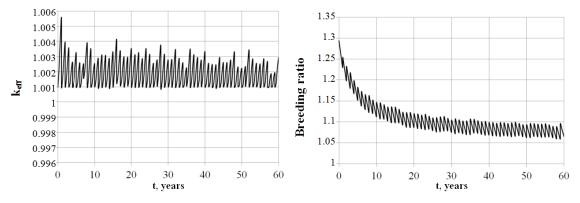


FIG. 6. The change of eigenvalue for the conventionally critical problem, and the breeding ratio during the reactor operation

The plutonium isotopic composition changes considerably during almost the whole period of operation. The ²³⁹Pu concentration reaches its equilibrium value approximately in 50 years (*see FIG. 7.*). If the core is loaded with the fuel with the equilibrium plutonium concentration of 14.2% before each micro-campaign, then the difference in the fuel breeding properties at the start-up will be ~27.5 β_{eff} , whereas in 50 years it will be about $2\beta_{eff}$. The charged fuel composition should be adjusted during the whole period of reactor operation. The most significant effect on breeding properties is made by variation of ²³⁹Pu and ²⁴¹Pu concentrations.

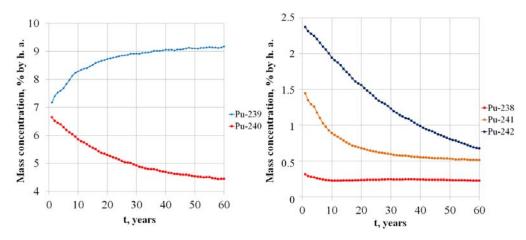


FIG. 7. Variation of plutonium isotope concentrations in the fuel during reactor operation

Operation of a reactor with core variable composition is accompanied by additional problems during fuel fabrication from regenerated materials due to changes in nuclide composition. Besides, it complicates control of the reactor. This is also typical for initial charge with enriched uranium.

5. Initial charge with enriched uranium

Initial charge with enriched uranium is considered in the framework of large-scale nuclear power development [3]. In particular, the use of enriched uranium makes it possible to lift restrictions related to terms of commissioning of the BREST-type reactors. In paper [5] it is shown that at such initial charge and constant core geometry, the reactivity excursion per micro-campaign will exceed β_{eff} during ~38 years. The reactivity variation in the process of fuel burnup greatly depends on the breeding rate of fissile isotopes in the core. Initial charge with enriched uranium is characterized by a low conversion ratio at the start-up (~0.69) due to smaller, as compared to plutonium, neutron yield v during ²³⁵U fission. With plutonium accumulation, v increases from 2.5 at the initial moment of time up to 2.9 by the end of reactor lifetime, resulting in the decrease of the cumulative fraction of fissions and the increase of the absorption fraction on source isotopes. The conversion ratio gradually grows and reaches a unity in approximately 38 years of operation (*see FIG. 8.*).

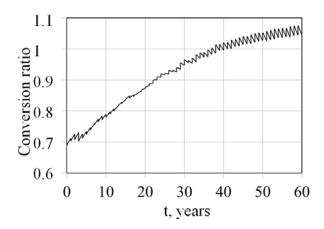


FIG. 8. Variation of conversion ratio during reactor operation

If at the initial moment of time for the uranium charge the main fissile isotope is ²³⁵U (~90% of fissions), then at the equilibrium operating conditions the majority of fissions will occur on fissile plutonium isotopes. Fgure 9 shows the change of mass fraction in the discharged ²³⁵U, ²³⁹Pu, ²⁴⁰Pu fuel. Approximately in 40 years of reactor operation ²³⁹Pu concentration in the core reaches its equilibrium value. At that, concentrations of the remaining plutonium isotopes continue to change (*see FIG. 10.*).

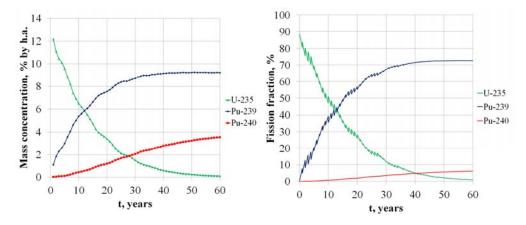


FIG. 9. Variation of 235U, 239Pu, and 240Pu concentrations and their respective fission fractions

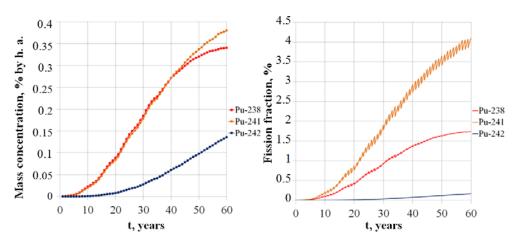


FIG. 10. Variation of 238Pu, 241Pu, and 242Pu concentrations and their respective fission fractions

At these assumptions, in approximately 40 years the reactor will operate with a practically constant 239 Pu concentration in the fuel in a self-sustained mode, with a reactivity margin less than β_{eff} . Thus, the reactor conventionally reaches the equilibrium operating conditions approximately in 40 years.

Different variants that involve changes in both the fuel composition and the core geometry during the transition operation period are studied at present in order to decrease the reactivity variation per micro-campaign. An efficient method to enhance fuel breeding and decrease the excursion reactivity is the use of high-power plutonium from the RBMK spent nuclear fuel at the initial charge in addition to enriched uranium. High ²⁴⁰Pu isotope concentration in RBMK high-power plutonium allow obtaining a core with a conversion ratio close to unity and with reactivity excursions per micro-campaign not exceeding the effective fraction of delayed neutrons. Such an approach does not require any changes of the core geometry in the process of transition to the equilibrium operating conditions.

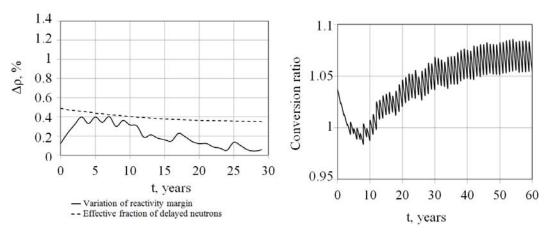


FIG. 11. Change of reactivity excursions and conversion ratio

Calculations show that for the reactor configuration under consideration, with a fuel column 110 cm high, the requirements to reactivity excursion are met at plutonium mass concentration ~12%, $^{235}U - 4.4\%$ by heavy atoms (the maximum reactivity excursion is approximately 0.95 β_{eff} , see FIG. 11.).

Throughout the whole period of operating life, the reactor will operate in a self-sustained mode by fissile isotopes. The total 235 U consumption during the whole operation period is ~4.8 tons, the consumption of plutonium from the RBMK spent nuclear fuel is 11.3 tons (with the startup load of 7 tons plus 4.3 tons per each of the three following refueling procedures). According to the ATEK [11] software complex assessments, plutonium stock after decommissioning of the last RBMK equals 140 tons. This quantity of plutonium is sufficient for startup of about 12 reactors of the BREST-1200 type, which makes it impossible to consider this variant of decreasing the reactivity excursion as the basic one.

A possible way to decrease reactivity excursions is the decrease of neutron leaks that results in plutonium buildup. However, this approach may result in security breaches for fast reactors (compensation of void effect of reactivity by coolant), which requires a more detailed research.

6. Conclusion

The initial charge with plutonium obtained from the VVER spent nuclear fuel with a cooling period of 20 years appears to be the closest to equilibrium. During the whole reactor lifetime, reactivity between two consecutive refueling procedures does not exceed $\beta_{eff} \approx 0.0035$, thus one can conventionally state that the reactor operates in equilibrium operating conditions from the first micro-campaign.

As for the initial charge with plutonium obtained from the RBMK spent nuclear fuel, the reactor core is characterized by high values of breeding ratio at the initial period of operation $(1.2\div1.3)$. The excessive plutonium buildup results in the change of the reactivity margin during the first micro-campaign that exceeds β_{eff} . From this point on, reactivity excursions equal $(0.3\div0.6)\beta_{eff}$. The ²³⁹Pu concentration reaches its equilibrium value in about 50 years. Thus, most part of its operating life the reactor will operate with a variable fuel composition. This is accompanied by additional problems during fuel fabrication from regenerated materials due to changes in nuclide composition. Besides, it complicates control of the reactor.

For initial charge with enriched uranium at constant geometry, the transition stage is approximately 40 years. The core is characterized by a low conversion ratio that results in the

deficiency of fissile materials in recycling products. The initial conversion ratio is ~0.69. The reactor reaches the self-sustained mode by fissile materials in about 38 years. The maximum reactivity change per micro-campaign during this period may be up to $2\beta_{eff}$. Different variants that involve changes in both the fuel composition and the core geometry during the transition stage of operation are studied at present in order to decrease the reactivity variation. An efficient method to enhance fuel breeding and decrease the excursion reactivity is the use of high-power plutonium from the RBMK spent nuclear fuel at the initial charge in addition to enriched uranium. Besides, such loading makes it possible to decrease the time required to reach a stationary plutonium isotopic composition. However, the number of reactors that can be started with such fuel is limited by the quantity of plutonium contained in spent fuel of RBMK reactors. To lift restrictions related to terms of commissioning of reactors, reactivity excursions can be compensated by decreasing neutron leaks from the core. Such an approach requires additional research, in particular, to ensure the negative void effect of reactivity.

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