

Minor actinides transmutation in pressurized water reactors. 1. Multiple recycling of minor actinides on the example of one VVER reactor*

Yury A. Kazansky¹, Gleb W. Karpovich¹

¹ Obninsk Institute for Nuclear Power Engineering, NRNU MEPhI, 1 Stugorodok, 249040 Obninsk, Kaluga Reg., Russia

Corresponding author: Gleb W. Karpovich (gleb.karpovich@yandex.ru)

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Abstract

This article explores the possibilities and conditions of combustion in a pressurized water reactor of its own accumulated minor actinides (MA). The simplest computational model is used: an infinitely extended medium with the distribution and composition of all materials of the fuel assembly of the reactor core, similar to VVER-1200, with uranium dioxide having an initial ²³⁵U enrichment of 4.95%. The burnup model is presented in the form of iterations, each of which simulates a fuel campaign lasting 4 years without refueling. At the start of the cycle, special fuel rods are loaded with minor actinides extracted from the reprocessed SNF of the VVER-1200 reactor. After the end of the fuel campaign, all the MAs are removed from the SNF and used in a new iteration.

As a result of calculations, it was found that the MA mass in the cycle after 3–7 iterations (depending on the number of fuel elements allocated for the placement and accumulation of MAs) tends to an equilibrium state (regardless of the MAs added every 4 years). In other words, the fuel rods allocated for loading MAs play the role of a kind of furnace, into which, in each iteration, MAs from the previous iteration accumulated in the given reactor are loaded. After several iterations, the burned MA mass converted into fission products is compared with the incoming one. The inclusion of MAs in this way into the fuel cycle converts at least 86% of MAs into fission products without affecting the power generation of the nuclear power plant. It is important that MAs are temporarily unloaded from the reactor after the next iteration in order to remove fission products and to add a new portion of MAs. After stopping the reactor operation, about 16% of the total amount of MAs generated for the entire history of the reactor's life is discharged into the storage facility. The initial fuel composition in the fuel rods allocated for loading MAs differs from the others only in the amount of MAs and the mass of ²³⁸U. The simplified computational model used in this work (without annual overloads of the reactor) influenced the burnup depth and, naturally, the duration of operation, i.e., the k_{∞} value becomes less than 1 after 1056 days instead of the actual 1460 days with annual fuel overloads. This affected the average fuel composition and, consequently, the neutron spectrum, and could affect the main result of the work, i.e., the number of burned-out MAs in different iterations. Additional calculations, taking into account the annual overloads of the reactor, showed that the change in the spectral composition had little effect on the amount of MAs at the end of the fuel campaign (within 2%). It turned out that the replacement of ²³⁸U with minor actinides in fuel rods, the number of which is less than 10, leads to a loss of reactivity. When the number of fuel rods for loading MAs is more than 10, the reactivity increases, giving hope for burning up MAs accumulated in several reactors.

Keywords

VVER, minor actinides, neptunium, americium, curium, transmutation, nuclear fuel, burnup, closed fuel cycle.

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Introduction and problem statement

A major complexity (and a natural increase in costs) involved in nuclear electricity generation is associated with the inevitably high radioactivity of spent nuclear fuel (SNF). After a pressurized water reactor of the VVER-1000 type stops to operate, the radioactivity of the fuel is $9 \cdot 10^{18}$ Bq/t (see, e.g., (Kolobashkin et al. 1983)), which is hundreds of millions of times as high as that of fresh uranium fuel.

The major contributors to radioactivity in the first hundreds of years after the SNF unloading from the reactor are fission fragments; the total radioactivity of uranium and plutonium is about 10%, and the rest of the heavy nuclides account for just a few percent.

When in storage, the activity of SNF decreases as the result of radioactive nuclei transformations. It will take however at least 200 thousand years for the SNF radioactivity to go down to the radioactivity level of the uranium ore extracted from the Earth's interior for production of nuclear fuel. Besides, the radioactivity in SNF storage facilities will not only decrease via nuclear transformations, but will also increase due to new batches of SNF unloaded from reactors. Finally, the relations between the components of the unloaded SNF depend on the fuel cycle scenario and the power level of a particular unit and of nuclear power industry as the whole. Dozens of publications are available concerned with the quantitative and qualitative radioactivity indicators of SNF unloaded from the reactor depending on the implemented fuel cycle. We shall refer to a number of papers that describe in brief the concepts of nuclear power radioactivity depending on fuel cycle (Use of Fast Reactors 1992, IAEA-TECDOC-693 1993, Shmelev et al. 1993, Kazansky et al. 1993a, 1993b, 2016, Slesarev et al. 1994, Adamov et al. 1996, Ganev et al. 2000a, 2000b, Kazansky and Klinov 2000, Bergelson et al. 2003, 2009, Wenchao et al. 2005, 2017, Khorasanov and Blokhin 2013, Kazansky and Romanov 2014, Ivanov et al. 2017, Washington and King 2017, Korobeynikov et al. 2019). Normally, the following components of SNF are taken into account: fission fragments (FF), uranium isotopes (the key component of the nuclear fuel cycle in the overwhelming majority of nuclear reactors), plutonium isotopes, which, when in a closed uranium-plutonium fuel cycle and if there are fast neutron reactors, can provide the planet with fuel resources for at least thousands of years, and, finally, neptunium, americium and curium isotopes called minor actinides (MA).

As shown by the available estimates (Kazansky et al. 1993a, 1993b), the radioactivity level of SNF unloaded from the reactor will become nearly stable in the storage facility simultaneously with the increase in the weight of the unloaded SNF. In this approximation, FFs and MAs turn out to be the major sources of radioactivity in the event of a closed fuel cycle, this being explained by a short time of residence in uranium and plutonium storage facilities. Radiation equivalence, that is the equality of the radioactivity withdrawn from the Earth's interior and

returned to it in the form of long-term geological repositories, can be achieved for a closed fuel cycle (Adamov et al. 1996).

If nuclear power stops to operate, the flow of SNF from the reactors will also stop. The maximum period of nuclear power operation is approximately one to two millennia (taking into account today's concepts of the nuclear fuel resources and the scale of the global energy consumption). The minimum period is one to two centuries (the key possible reasons are the emergence of new more efficient ways of obtaining and using energy, a change in the paradigm of technological advancement, and deindustrialization). Then the level of radioactivity will decrease rather quickly (hundreds of years), by one or two orders of magnitude, largely as a result of the FF radioactive decay. And this level will depend on the scenario of the nuclear power operation: the total radioactivity level of accumulated SNF will be the highest with an open fuel cycle, and it will be lowest with a closed fuel cycle for uranium and plutonium, while the SNF radioactivity level can be reduced further due to minor actinides being added to the fuel cycle (Kazansky et al. 1993a, 1993b). Apparently, concerns about the distant future and the "purity" of nuclear power have become the motivation for many studies on minor actinide handling (Kazansky et al. 1993a, 1993b).

Transmutation is used for the forced change in the radioactivity of unstable nuclides (a change in the composition of nuclei as the result of nuclear reactions under the action of neutrons, which is possible, basically, in power reactors). The ideology of transmutation and its key characteristics were the major issues addressed at the International Conference in Obninsk in 1992 (Shmelev et al. 1993). Specifically, the reports delivered at the conference and later papers confirm that MA irradiation involves an increase in radioactivity and then its decline the rate of which depends on the irradiation time. For 30 years of irradiation in a thermal neutron reactor, the radioactivity declines by about 10 times, and the number of MA nuclei decreases by about 15 to 25 times (Kazansky and Romanov 2014, Kazansky et al. 2016).

Transmutation of some fission fragments is often discussed in scientific literature. However, transmutation of all fission fragments (repeated placement in nuclear reactors) does not lead to a noticeable decrease in radioactivity (Kazansky and Klinov 2000, Ivanov et al. 2017). Papers are known which deal with special-purpose reactors for burning minor actinides (MA) (Khorasanov and Blokhin 2013, Korobeynikov et al. 2019), including those using americium as fuel. Finally, there are papers on the feasibility of the MA disposal and transmutation in existing (Shmelev et al. 1993, Slesarev et al. 1994, Bergelson et al. 2003, 2009, Wenchao et al. 2005, 2017, Washington and King 2017) and future nuclear power reactors (Ganev et al. 2000a, 2000b).

The purpose of this paper is to confirm or disprove the feasibility of MA burning in VVER reactors without affecting the electricity generation, the lifetime and the

nuclear fuel consumption. This approach makes it possible to use the existing reactors to address the problem of MA accumulation. The investigation results presented in this paper have a focus (use of effective and future power reactors for MA burning) similar, e.g., to those in (Bergelson et al. 2003, 2009, Wenchao et al. 2005, 2017, Washington and King 2017). The major difference is the composition of burnt fuel elements with MAs and replacement of enriched uranium with minor actinides, provided the level of electricity generation remains the same.

The use of effective power reactors will lead to difficulties in operation (the radioactivity of fresh fuel assemblies with minor actinides will exceed greatly the radioactivity of VVER fuel assemblies). Therefore, the number of reactors with minor actinides shall be as small as possible. Such calculations have been undertaken and the results will be published.

Computational model

It is proposed that MAs are added to the closed fuel cycle. The reactor is simulated as a single fuel assembly. The paper considers the fuel cycle, the flowchart of which is shown in Fig. 1.

The minor actinides produced in the VVER-1200 reactor are recycled in the above fuel cycle: the withdrawn SNF is cooled in the pool being thereafter delivered for reprocessing for the Np, Am and Cm extraction. Minor actinides are further used to be added to enriched uranium based on which fresh nuclear fuel is made. The period of the nuclear fuel irradiation is four years. For the convenience of calculations, the SNF storage and reprocessing time in the simulated fuel cycle is also assumed to be equal to four years.

The dynamics of the heavy nuclei accumulation and burnup depends on the geometry of the nuclei distribution in the reactor core. To understand the effects of the MA distribution on the MA burnup results, uranium is replaced with MAs in a different number of fuel elements in one fuel assembly: 312 (all), 39, 19, 10, 5, 4. It needs to be taken into account that one fuel element contains only 1.5 kg of fuel in the form of a heavy metal oxide mixture (Faghihi et al. 2019).

The nuclear fuel burnup is calculated as follows:

- instead of simulating the entire core of a nuclear reactor (using FAs with different burnups), a model of one VVER-1200 FA with 312 fuel elements is used, placed in an infinite multiplying medium of itself (a mirror boundary condition is defined for the FA facets); the burnup for the entire fuel life (four years) is calculated in the same model;
- MAs are added to a different number of fuel elements, this making it possible to select the optimal number of fuel elements to contain MAs, and to investigate the effects of the MA cross-section blocking and, therefore, the minor actinide burning rate;
- the burnup is normalized based on the thermal power of the VVER-1200 reactor (3220 MW) assuming that power density is distributed uniformly among the FAs (in this case, one FA accounts for 19.75 MW);
- after a four-year cycle (1460 days of irradiation) of the fuel irradiation, the entire mass of the accumulated MAs replaces ^{238}U .

The MA weight and isotopic composition, as well as the fuel burnup both with and without MA addition were monitored in the process of the calculations, this making it pos-

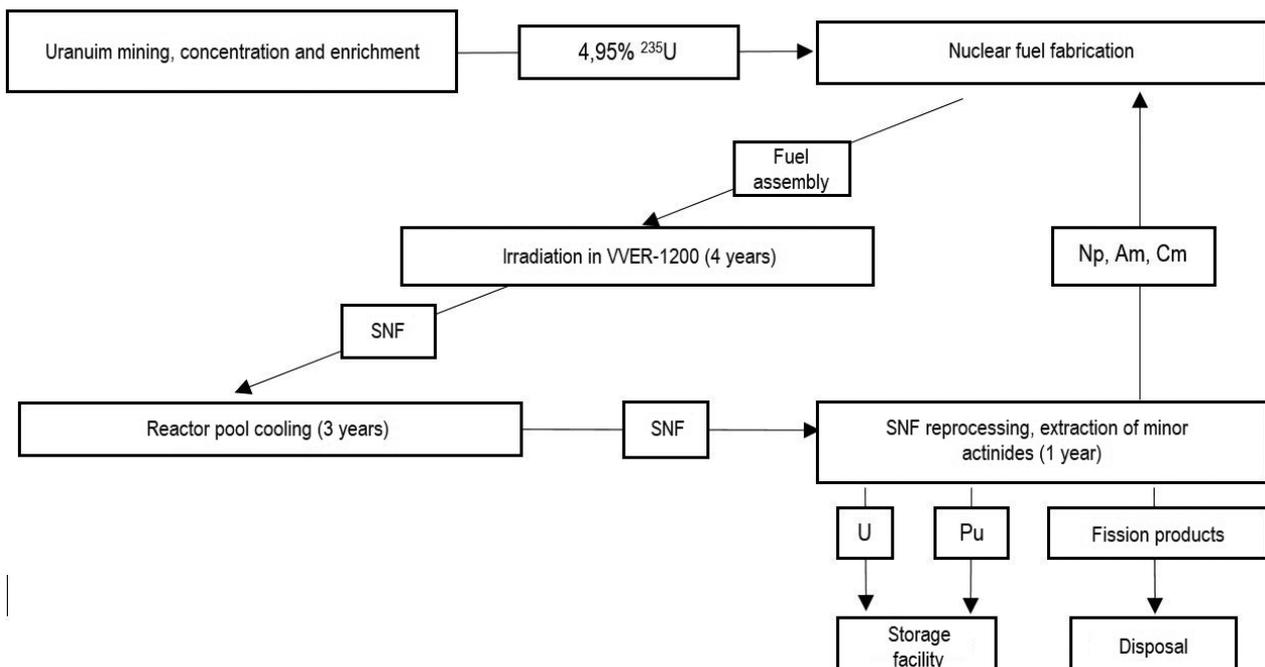


Figure 1. Flowchart of the fuel cycle for a thermal neutron reactor with minor actinide recycling.

sible to evaluate the MA effects on the irregularity of the power density distribution as compared with standard fuel.

The calculations were carried out using the Serpent 2.1.30 neutronic simulation code (VTT, Finland) (Leppänen 2015) with the JEFF 3.1.1 evaluated nuclear database supplied by the code manufacturer (Leppänen and Viitanen 2013).

Fuel burnup with a fixed initial amount of ^{235}U in each fuel element

Initially, the nuclear fuel burnup was simulated in the case when the initial amount of ^{235}U in each fuel element was fixed. The first calculation with a reference assembly with an enrichment of $X = 4.95\%$ was taken as the zero iteration. The burnup of such FA leads to 776 g of accumulated minor actinides (the weights of the key isotopes are as follows: 410 g of ^{237}Np ; 195 g of ^{241}Am ; 118 g of ^{243}Am ; 47 g of ^{244}Cm ; 4 g of ^{245}Cm). Then, the produced minor actinides are returned to the FA, and the iteration number shows the number of the MA returns to the reactor.

As shown by the calculation results, a dynamic equilibrium is reached after a certain value the MA quantity in the fuel cycle tends to. The equilibrium quantity of MAs as such contained in the fuel cycle depends on the number of the FAs with MAs: the larger is the number of such FAs, the faster the equilibrium is achieved and the smaller is the MA load in the reactor. The fuel weight in one fuel element is 1.5 kg. Taking into account the data presented in Fig. 2, the maximum fraction of the minor actinide nuclei in the fuel elements with MAs is 26%.

It should be noted that the time the dynamic equilibrium is reached at is different for each isotope. It is reached the fastest for ^{237}Np , Am and ^{244}Cm . Further, the mass of minor actinides is enriched with heavy curium isotopes, and more californium isotopes are accumulated. Plutonium recovered from the fuel with MAs added earlier for burning contains up to 8.5% of ^{238}Pu . This isotope is accumulated due to the radiative capture of ^{237}Np neutrons. ^{238}Pu is

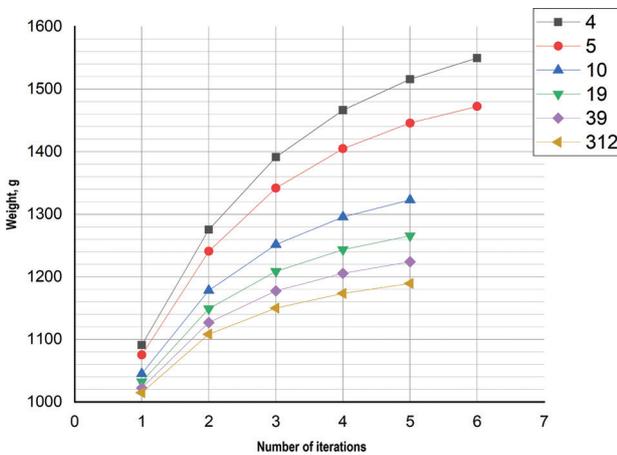


Figure 2. Weight of minor actinides in FAs at the end of life as a function of the number of recycles for different numbers of fuel elements containing MAs.

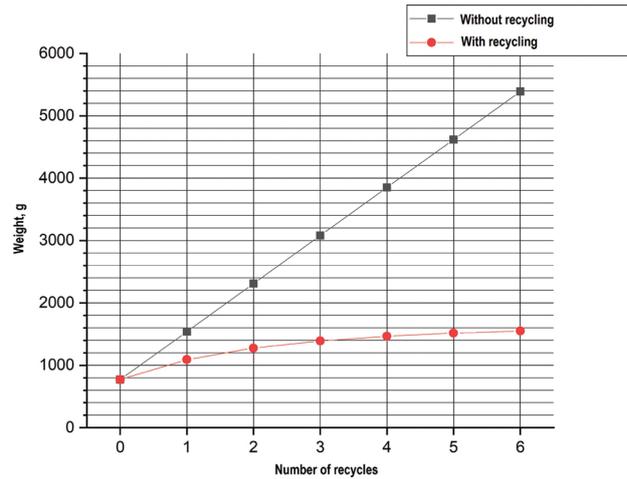


Figure 3. Comparison of MA weights in the fuel cycle with or without recycling.

characterized by a high heat release per volume unit due to radioactive decay (0.57 W/g) and makes it difficult to handle the plutonium extracted from SNF (specifically when used to manufacture nuclear munitions). However, contaminated plutonium does not really make a problem as the concentration of ^{238}Pu can be easily reduced to 2% or less by mixing it with plutonium from standard spent fuel.

But is it possible that recycling is not continuous but rather limited to several MA returns to the reactor? Reducing the number of recycles makes it possible to reduce the amount of radiation-hazardous activities for the fuel cycle. To answer this question, it is necessary to compare the quantity of minor actinides in the fuel cycle in two cases: when MAs are placed in storage without being reprocessed and when these are returned to the reactor after each SNF reprocessing. With no recycling, a fixed quantity of minor actinides is placed in storage each time, and no decay of these is taken into account in the overall dynamics since the MAs in storage are not cleaned at a regular basis of their radioactive decay products.

Fig. 3 shows that recycling of minor actinides needs to be continuous since the gain in the number of the MAs burnt grows with each MA return to the reactor.

Fuel is fully replaced 15 times in the course of the VVER-1200 reactor design lifetime (60 years). If no MAs are recycled, the reactor will produce nearly 1900 kg of minor actinides throughout the fuel life. With recycling (worst-case scenario), this amount is reduced to 261 kg; 86% of MAs transform, therefore, into fission fragments.

To clarify the reasons for the equilibrium quantity of MAs growing with the increase in the MA density, it is necessary to calculate the average ratio of the radiative capture and fission macrosections for all heavy nuclei for fuel with MAs in the first iteration when the composition of minor actinides in all distribution options is the same. The average ratio is calculated using the formula

$$\langle \alpha \rangle = \frac{\sum_{i=1}^n \Sigma_c^i}{\sum_{i=1}^n \Sigma_f^i},$$

where Σ_c^i is the radiation capture macrosection; Σ_f^i is the fission macrosection of the i^{th} isotope.

Table 1. Ratio of the radiation capture and fission macrosections for all heavy nuclei for fuel elements with Mas

Number of fuel elements with MAs	4	5	10	19	39	78	156	312
MA concentration, wt. %	12.78	10.23	5.11	2.69	1.31	0.66	0.33	0.16
$\langle\alpha\rangle$ for fuel with MAs	3.39	2.93	1.94	1.40	1.05	0.87	0.77	0.72

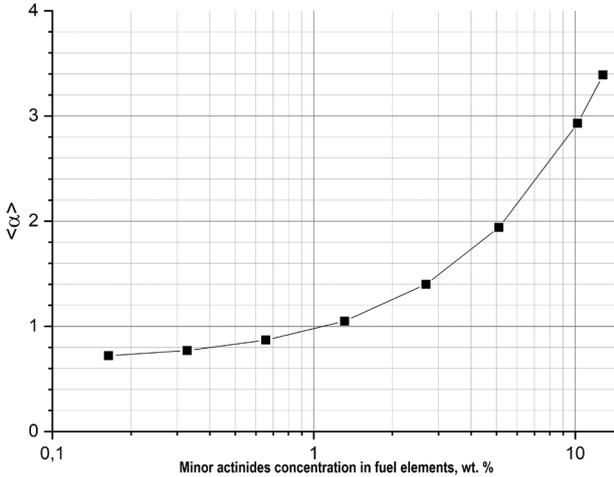


Figure 4. Ratio of the radiation capture and fission cross-section for all heavy nuclei for fuel with MAs.

For comparison, $\langle\alpha\rangle = 0.67$ for the VVER-1200 uranium oxide fuel with the enrichment of 4.95% at the beginning of the fuel life.

As it can be seen from Fig. 4, an increase in the concentration of minor actinides in fuel leads to the fission cross-sections being blocked more intensely than the radiation capture cross-sections. Therefore, high MA density in the reactor is not practical from the standpoint of the rate of the MA transmutation into fission fragments and the observance of the required reactor BOL reactivity margin.

MA recycling shall not lead to a shorter fuel life (and, so, to smaller nuclear power generation). This requires calculating the time for a fuel assembly with MAs to reach $k_\infty = 1$ and the criticality at its fuel life end and comparing it with the indicators of an FA in operation with no MAs ($T(k_\infty = 1) = 1083$ days).

As it can be seen from the table, adding MAs into an FA somewhat worsens its multiplication properties: k_∞ decreases to 1 in a shorter time. This means that FAs with minor actinides lose their reactivity margin earlier than FAs with no MAs. To understand if it is necessary to add more fissile nuclei (increase the uranium enrichment, add plutonium, etc.) to keep the reactor critical, we shall check the effects of minor actinides on the reactor reactivity at the fuel life end.

Fig. 5 shows two major relationships:

- the smaller is the number of fuel elements with MAs, the smaller is k_∞ at the end of life, this being explained by the effects of the heavy nuclei cross-section blocking (the fission-cross sections

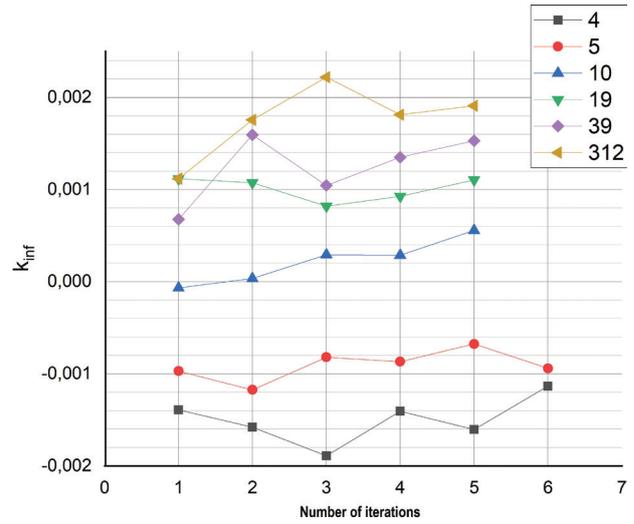


Figure 5. Effects of minor actinides on the reactivity of the reactor with a different number of fuel elements with MAs. The EOL reactivity of the reactor with no MAs is assumed to be equal to zero.

are blocked more intensely than the radiation capture cross-sections);

- for several scenarios, k_∞ at the fuel life end is larger than for FAs with no minor actinides: this does not contradict earlier conclusions based on the data in Table 2 since the FAs with added MAs are characterized by a slower reactivity loss rate than the reference assembly with no minor actinides due to the generation of fissile nuclides from MAs which are not fissionable in a thermal spectrum. This gives hope for achieving a fuel cycle with the enriched uranium replacement for minor actinides at the beginning of the next iteration.

Table 2. Time needed for an FA with MAs to reach $k_\infty = 1$

Number of fuel elements with MAs	4	5	10	19	39	312
$T(k_\infty = 1)$, days	1052	1052	1053	1055	1056	1058

One more important parameter is fuel burnup in fuel elements with an MA load.

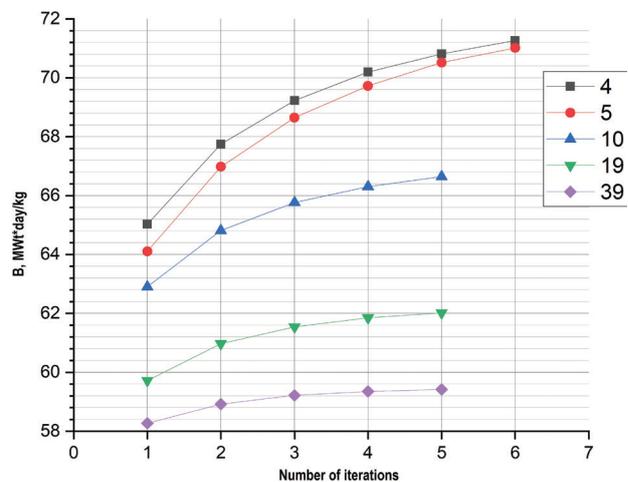


Figure 6. Burnup of fuel with added minor actinides at the fuel life end as a function of the number of recycles (for different scenarios of MA distribution in FAs).

For comparison, 4.95% uranium oxide fuel in a VVER-1200 burns up to 57 MW·day/kg in four years.

The calculated burnup of fuel with MAs shows that the burnup increases with the growth in the MA concentration in the fuel. This is explained by the additional minor actinide contribution to power density due to fission.

Taking into account the spectral component in MA accumulation

The above results were obtained using a model of one FA in an infinite breeding medium with an estimated fuel life of four years. In reality, the reactor is shut down for refueling each 12 to 18 months with the fuel burnt up partially replaced for fresh fuel. This is the reason why an FA is surrounded by other assemblies with different burnups. As a result, the neutron spectrum in the model somewhat differs from the neutron spectrum in the reactor with refueling. And differences in the spectrum inevitably lead to different MA accumulation rates.

To explore the effects of refueling on the neutron spectrum and, accordingly, on the MA accumulation and burnup rates, an additional calculation was undertaken using the FA model simulating the presence of fuel with different burnups in the reactor.

The test model has the FAs divided into four groups of fuel elements (of 78 each); each group is loaded with fuel with burnups corresponding to 0, 1, 2 and 3 years of being in the reactor core. The fuel elements are accommodated in a uniform manner in the FAs, the isotopic composition of the fuel for each group being taken from the calculation results for the four-year burnup of fuel with an enrichment of 4.95% (with no cooling after irradiation). The irradiation time was 365 days, and the FA power was 19.75 MW. The monitored parameter was the MA post-burnup weight. For illustration, the MA weights in fuels with the same burnup from different calculation models were compared.

Table 3. Comparison of MA weights for different fuel irradiation scenarios

Burnup, MW·day/kg	MA weight in one FA, g		$\Delta = m_1 - m_0$, g	Δ/m_0 , %
	m_0 (four-year burnup)	m_1 (with yearly refueling)		
14	98,8	118,0	19,2	16,3
28	226,0	270,8	44,8	16,5
42	417,2	456,4	39,2	8,6
56	658,4*	670,8	12,4	1,8

* The difference from the results in Fig. 2 is explained by the absence of four-year cooling in the cooling pool; a part of ^{241}Pu ($T_{1/2} = 14.3$ g) transforms through β -decay into ^{241}Am which is considered in the MA weight.

The results presented in Table 3 show that the effects of the spectral component resulting from refueling on the MA accumulation rate do not exceed 2% at the fuel life end.

Conclusions

The results of the study allow for the following conclusions.

1. A VVER-type reactor is capable of operating in a mode with complete recycling of minor actinides.
2. Continuous recycling of minor actinides in a VVER reactor is accompanied by their quantity in the cycle becoming constant due to a dynamic equilibrium achieved between the generation and burnup rates.
3. The dynamic level depends directly on the number of the fuel elements in each FA which contain MAs. The larger is the number of fuel elements (and, accordingly, the lower is the MA concentration), the faster the dynamic equilibrium is achieved and the smaller is the weight of the MAs in the cycle. This effect is explained by effects of the heavy nuclei fission cross-section blocking: the higher is the heavy nuclei concentration in the fuel, the greater is the ratio of the mean radiative capture cross-section to fission.
4. MAs form secondary nuclear fuel. High concentration of MAs in fuel elements leads to a high specific power density (and, accordingly, to a higher burnup as compared with standard uranium-oxide fuel), so the number of the fuel elements for recycling needs to be selected with regard for the technological limits for the linear power density. Otherwise, fuel claddings will burn out and a critical heat flux will occur between the fuel and the coolant.
5. Single recycling of minor actinides makes little sense since a major gain in their weight reduction is achieved only when there is a dynamic equilibrium achieved in the fuel cycle. Continuous recycling of minor actinides leads to their remaining weight reduced by 86% as a minimum.

Further evolution

The recycling options discussed in this paper are confined to a scenario when the VVER reactor burns its own MAs. In practice, this means that each power unit with a reactor similar to VVER-1200 shall include special-purpose equipment for handling fresh fuel containing minor actinides (since this fuel is much more active in terms of gamma and neutron radiation than the fuel based on enriched uranium currently in use). It makes sense, therefore, to consider those scenarios where one VVER reactor disposes of minor actinides produced in several reactors (or kept in storage facilities for fissile materials extracted from reprocessed SNF).

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