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Radiation characteristics of REMIX fuel during multiple recycling in VVER-1000 reactors

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Abstract

Changes in the isotopic composition of three REMIX fuel types in the process of operation in a VVER-1000 reactor have been calculated. It has been shown that it is possible to recycle REMIX-A and REMIX-A2 fuel more than five times without a major decrease in its nuclear value. The equivalent dose rate from fresh REMIX fuel assemblies has been calculated depending on the recycle number and the fresh fuel decay time after fabrication. The relative contribution of different radionuclides to the equivalent dose rate has been analyzed. The calculation results for the buildup of ^{232}U and even Pu isotopes depending on the fuel recycle number are presented.
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Keywords: REMIX fuel; Fuel cycle; VVER-1000 light-water reactor; Uranium; Plutonium; Uranium recycling; Plutonium recycling; Isotopic composition; Equivalent dose rate; Spent nuclear fuel.

Introduction

Recycling of reprocessed uranium and plutonium in thermal neutron reactors may provide for a more efficient use of nuclear fuel, a reduction in the amount of spent nuclear fuel (SNF) to be disposed, and a decrease in the risk of nuclear proliferation. Use of REMIX fuel is one of the options for the nuclear fuel cycle closing in the operation of VVER reactors [1]. However, recycling of plutonium is known to result in a buildup of its even isotopes in fuel [2], and uranium recycling leads to a build-up of ^{232}U [3]. Both cause the radiation characteristics of fresh FAs to deteriorate. Use of REMIX fuel in VVER reactors suggests recycling of both uranium and plutonium. The paper presents calculation results for the variation in the equivalent dose rate in the proximity of FAs with REMIX fuel during recycling. The equivalent dose rate in the proximity of FAs is an important contributor to the radiological hazard from nuclear fuel. Knowing it is essential in design of shipping casks, and in planning refueling operations in the fresh fuel region at a nuclear power plant.

Three types of REMIX fuel are considered in the paper: A, A2 and B. The REMIX-A fuel [1,4] is a non-separated mixture of reprocessed uranium and plutonium following the addition of 17–20 wt. % of enriched natural uranium (ENU) containing 19.75% of ^{235}U. The REMIX-A2 fuel differs from the REMIX-A fuel in a higher plutonium content [5]. For calculations, the plutonium weight content in the REMIX-A2 fuel is assumed to be 3%. Such an increase in the plutonium content makes it possible to reduce the amount of enriched uranium added after each recycle. The third fuel type, REMIX-B, [6] is fabricated from separated uranium and plutonium products obtained during SNF processing, with all of the extracted uranium being re-enriched and then added to the extracted plutonium. The plutonium content in this fuel is also 3%. This fuel is not recycled.

In this study, the change in the nuclide composition of the considered REMIX fuel types in the process of operation in a VVER-1000 reactor was calculated. The fuel burnup was calculated for an infinite FA grid using precision

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Table 1
Isotopic composition of initial uranium–plutonium regenerate.

Composition	^{238}U	^{235}U	^{236}U	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
Content, kg/THM	973.72	8.58	6.04	0.30	6.42	2.71	1.40	0.83
Total, kg/THM	988.3			11.7				

Table 2
Isotopic composition of plutonium addition to REMIX-A2.

Composition	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
Content, %	1.3	63.3	25.1	5.2	5.1

Monte Carlo codes. A 4-year reactor operation cycle was calculated. The equivalent dose from FAs was estimated by Monte-Carlo method. Neutron and gamma radiation contributions were taken into account. Other radiation types (α- and β-) do not contribute to the dose rate in the proximity of FAs, since electrons and α-particles have small path lengths and are delayed by the fuel cladding. The equivalent dose rates from FAs with the different REMIX fuel types were compared.

Multiple recycling of REMIX-A and REMIX-A2 fuels

The MCU code [7] and the MURE code [8] plus the MCNP code [9] were used to calculate the fuel composition after burn-up. To compare the performance of the codes, the REMIX-A fuel burn-up was calculated using both the MCU and the MURE+MCNP software packages. After the successful data verification, the REMIX-A2 fuel burn-up was calculated using MURE+MCNP, and the REMIX-B burn-up was calculated based on MCU.

A model of the TVS-2 M fuel assembly with mirroring boundary conditions was used for the calculations. The initial material for the REMIX fuel was the VVER-1000 spent uranium fuel with a burn-up of 49.2 GWd/THM (the average design fuel burn-up in an FA with an initial uranium enrichment of 4.33% after a 4-year fuel cycle). The FA irradiation time was 1200 effective days, and the decay time to the fabrication of reprocessed fuel was 5 years.

Fabrication of REMIX fuel requires uranium–plutonium regenerate to be extracted from the initial material. Its composition is given in Table 1.

During calculations, uranium with an invariable composition and an enrichment of 19.75% (a commercial product of JSC "SCC") was used as the initial material for the makeup fuel; in addition to highly-enriched uranium, plutonium extracted from the VVER-440 reactor spent fuel (from PA "Mayak") with an average burn-up of 36 GWd/THM was used for the REMIX-A2 makeup. The isotopic composition of the added plutonium is given in Table 2.

The REMIX burn-up was assumed to be 49.2 GWd/THM. For all recycles, the composition of the fresh REMIX fuel is adjusted such that to obtain an effective enrichment of 4.95%. The effective fuel enrichment is calculated with regard for the specific content of ^{235}U, ^{239}Pu and ^{241}Pu in the fuel and the content compensation factors for ^{236}U, ^{240}Pu and ^{242}Pu [10]. The calculation results for the REMIX-A fuel burn-up after five recycles are given in Table 3.

Since the REMIX-A fuel concept suggests the use of a non-separated mixture of uranium and plutonium, the plutonium content in the fuel increases with each recycle. It can be also seen that the fuel composition "deteriorates" with each recycle: there is an increase in the ^{236}U concentration which reduces the reactor reactivity, the content of fissionable Pu isotopes (^{239}Pu and ^{241}Pu) decreases, and the content of non-fissionable Pu isotopes (^{240}Pu and ^{242}Pu) grows. By the fifth recycle, the content of even plutonium isotopes increases from 33% to 40%. There is a growth in the buildup of ^{232}U whose decay products produce hard gamma radiation. After each recycle, there is an increase in the content of americium and curium being intensive neutron sources.

Table 3
U and Pu content (kg/THM) in REMIX-A fuel.

Composition	Recycle 1		Recycle 2		Recycle 3		Recycle 4		Recycle 5	
	Before	After	Before	After	Before	After	Before	After	Before	After
^{232}U	2×10^{-6}	5×10^{-6}	5×10^{-6}	8×10^{-6}	7×10^{-6}	10×10^{-6}	8×10^{-6}	1×10^{-5}	9×10^{-6}	1×10^{-5}
^{234}U	0.6	0.4	0.7	0.4	0.7	0.5	0.8	0.5	0.8	0.5
^{235}U	47.9	14.5	48.5	16.1	49.4	17.2	50.1	18.0	50.7	18.5
^{236}U	4.8	9.9	8.6	13.2	11.5	15.8	13.8	17.9	15.6	19.6
^{238}U	937.6	907.4	929.5	899.7	923.9	894.6	91.8	890.7	916.7	887.7
Σ(U)	990.9	932.2	987.2	929.4	985.4	928.0	984.4	927.1	983.7	926.4
Pu (even)	3.1	5.4	4.8	6.5	5.9	7.2	6.5	7.6	6.9	7.9
Pu (odd)	6.0	9.6	8.0	10.6	8.8	11.0	9.2	11.3	9.4	11.5
Σ(Pu)	9.1	14.8	12.8	16.9	14.6	18.0	15.6	18.7	16.3	19.2
Am+Cm	–	1.4	–	1.8	–	2.0	–	2.1	–	2.2
U + Pu	1000	947.0	1000	946.3	1000	946.0	1000	945.8	1000	945.5
Eff. enrich., %	4.95	1.66	4.95	1.72	4.95	1.75	4.95	1.76	4.95	1.76

Table 4
U and Pu content (kg/THM) in REMIX-A2 fuel.

Composition	Recycle 1		Recycle 2		Recycle 3		Recycle 4		Recycle 5	
	Before	After	Before	After	Before	After	Before	After	Before	After
^{232}U	2.4×10^{-6}	6.6×10^{-6}	5.8×10^{-6}	9.8×10^{-6}	8.7×10^{-6}	1.2×10^{-5}	1.1×10^{-5}	1.4×10^{-5}	1.3×10^{-5}	1.6×10^{-5}
^{235}U	40.0	16.5	44.8	18.9	46.7	19.9	47.9	20.5	48.8	21.0
^{236}U	4.9	8.7	7.7	11.6	10.3	14.2	12.6	16.3	14.5	18.1
^{238}U	925.1	895.2	917.5	888.2	912.9	883.9	909.5	880.6	906.7	878.0
Σ(U)	970.0	920.4	970.0	918.7	970.0	917.9	970.0	917.4	970.0	917.0
Pu (even)	9.6	10.6	11.9	11.6	12.7	12.1	13.1	12.5	13.3	12.7
Pu (odd)	20.4	15.3	18.2	15.1	17.4	15.0	17.0	14.9	16.7	14.9
Σ(Pu)	30.0	25.9	30.0	26.7	30.0	27.1	30.0	27.4	30.0	27.6
Am+Cm	–	2.7	–	3.2	–	3.3	–	3.4	–	3.4
U + Pu	1000.0	948.9	1000.0	948.6	1000.0	948.3	1000.0	948.2	1000.0	948.0
Eff. enrich., %	4.95	2.04	4.95	2.11	4.95	2.10	4.95	2.08	4.95	2.06

Despite a certain deterioration of the composition, it can be seen that enrichment of spent fuel is still much more effective than enrichment of natural uranium, which demonstrates that further recycling is possible. With the burn-up increase to 60 GWd/THM, the spent fuel composition after the fifth recycle will become unusable for further recycling.

To calculate the REMIX-A2 fuel composition, a mixture of reprocessed uranium and plutonium was used as the fresh fuel for the next recycle. Notably, the amount of the added plutonium was defined from the condition that the fresh fuel was to contain 3% of plutonium. The required effective fuel enrichment was achieved by adjusting the mass fraction of highly-enriched uranium in the makeup. The calculation results for the REMIX-A2 fuel composition after five recycles are given in Table 4.

Effective enrichment after the fifth recycle is slightly higher than in the event of REMIX-A, so, accordingly, this fuel may be further recycled as well. Based on a comparison against the estimated REMIX-A fuel composition, the following may be pointed out: by the fifth recycle, there is a slightly greater increase (from 32% to 44%) in the fraction of even plutonium isotopes, the content of ^{235}U increases, and the buildup of americium and curium is greater.

^{232}U and even plutonium isotopes build up in the process of recycling for both REMIX fuel types. Figs. 1 and 2 present the content of these isotopes in fresh fuel depending on the recycle number. An increase in the fraction of these isotopes leads to a higher fresh fuel dose rate. To determine more accurately the dose rate increase caused by the uranium and plutonium recycling, the equivalent dose rate from fresh FAs was calculated.

The neutron and gamma dose rates were calculated using the MCNP code, while the neutron spectra were determined using the MCU5 code, and the EASY-2010 code was used to calculate the gamma spectra [11]. For the neutron source calculation, neutrons from spontaneous fission of actinides and neutrons from (α, n)-reactions with the oxygen contained in the fuel were taken into account. To determine the spectral distribution and the gamma radiation source intensities, both the radionuclides initially contained in the fuel and the daughter decay products were taken into account.

Table 5
Ionizing radiation dose rate at a distance of 30 cm from the FA surface.

FA composition	UO_2	REMIX-A	REMIX-A2	REMIX-B
Fresh fuel, μSv/h	5	65	196	214
Fresh fuel after 6-month decay, μSv/h	23	143	314	368
Burnt-up fuel with no decay, Sv/h	6.95×10^4	6.95×10^4	686	–
Burnt-up fuel after 6-month decay, Sv/h	586	584	583	–
Burnt-up fuel after 5-year decay, Sv/h	98	98	99	138

The so calculated neutron and gamma-quantum source was distributed uniformly throughout the nuclear fuel in the FAs, following which the equivalent dose rate around the FAs was calculated.

A model of the TVS-2 M fuel assembly was selected for the calculations. The radiation dose rate was calculated at a distance of 30 cm from the assembly boundary (Fig. 3).

For the first recycle, the dose rate from the burnt-up fuel was calculated for a burn-up of 49.2 GWd/THM. The calculation results are presented in Table 5.

In the event of REMIX-A and REMIX-A2 fuel, the dose rate was calculated for the fresh fuel from each of the 5 recycles. The fuel decay time was assumed to be 180 days. The obtained data is presented in Fig. 4.

Gamma-quanta, neutrons from spontaneous fission of plutonium isotopes and neutrons from (α, n)-reactions are the major contributors to the dose rate. Fig. 5 shows the equivalent dose rate contributions from each uranium and plutonium isotope individually for the fresh REMIX-A fuel from the first recycle. As shown in the figure, the major contributors to the gamma radiation are ^{232}U and ^{241}Pu. The isotopes as such do not contribute greatly to the gamma radiation dose rate. The major contributors to the dose rate are ^{208}Tl and ^{241}Am, their decay products. For the most part, it is even plutonium isotopes that undergo spontaneous fission with the neutron yield. Besides, the fission of plutonium produces α-particles which interact with oxygen with neutron generation.

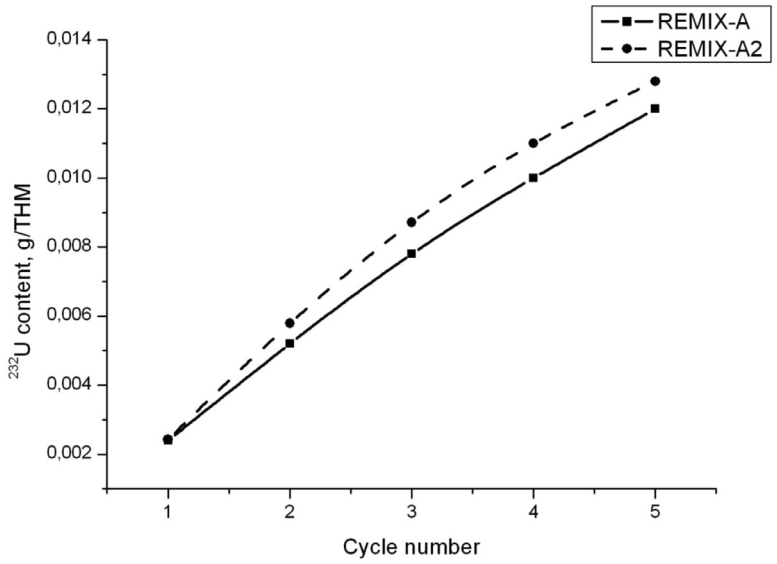

Fig. 1. Content of ^{232}U in fresh REMIX-A and REMIX-A2 fuel depending on the recycle number.

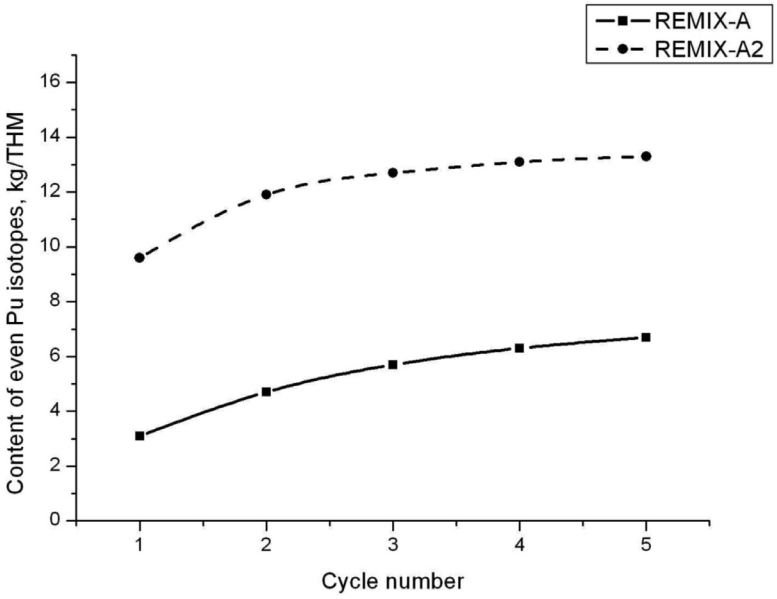

Fig. 2. Content of even plutonium isotopes in fresh REMIX-A and REMIX-A2 fuel depending on the recycle number.

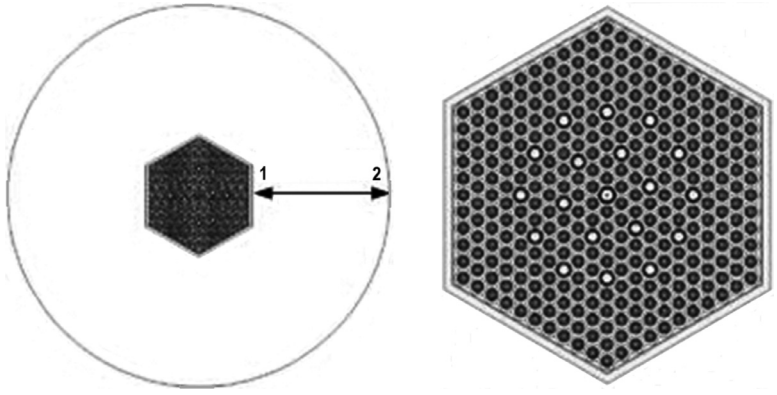

Fig. 3. A VVER-1000 FA. Calculation points 1 (on the surface) and 2 (30 cm from the surface).

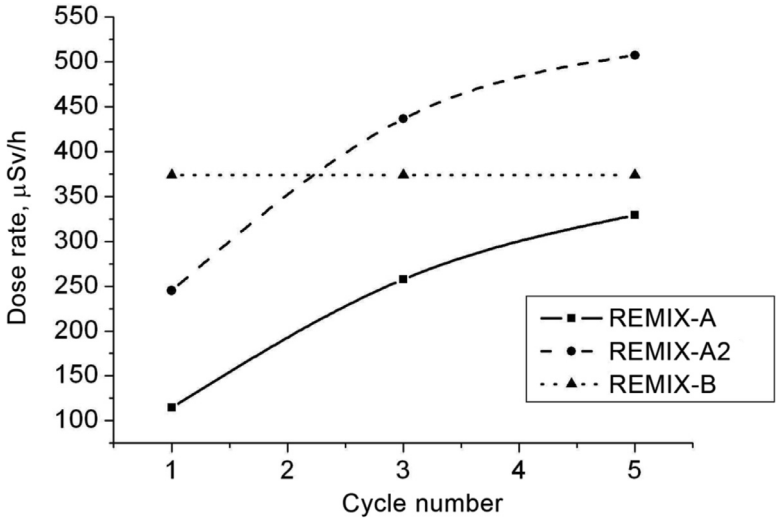

Fig. 4. Equivalent dose rate from an FA with fresh REMIX fuel for five recycles at a distance of 30 cm from its surface.

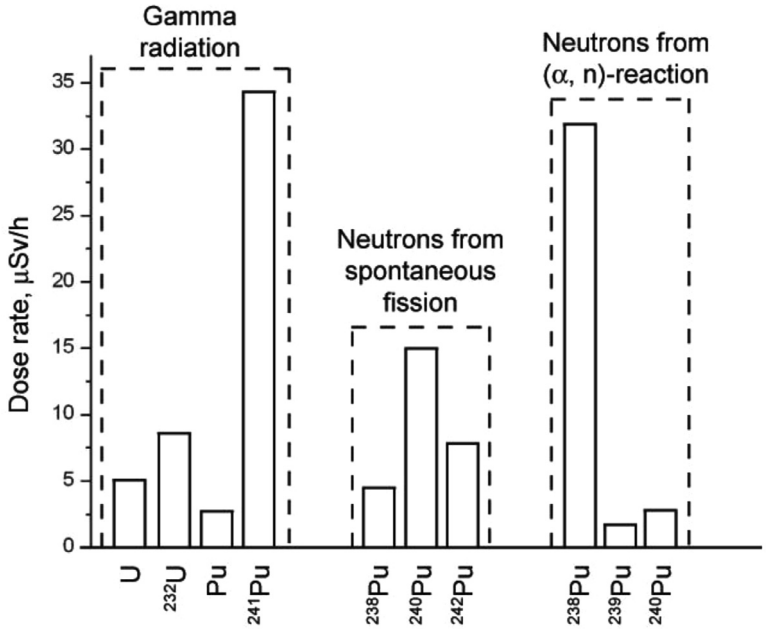

Fig. 5. Contribution of uranium and plutonium isotopes to the equivalent dose rate.

Fig. 6 shows the contributions of ^{232}U, ^{241}Pu and even Pu isotopes to the equivalent dose rate for the fresh REMIX-A fuel depending on the recycle number. As can be seen, the contribution of the gamma radiation from the ^{232}U and ^{241}Pu decay products increases with each recycle, but this increase is not great as compared to the dose rate contribution from even plutonium isotopes.

REMIX-B fuel

Let us consider the REMIX-B fuel. This fuel concept suggests the complete separation of uranium and plutonium during SNF reprocessing, and further enrichment of reprocessed uranium. The composition of the so produced fresh fuel is given in Table 6. To estimate the potential of this fuel, its burn-up was calculated for different values using the MCU5 code. The calculation results are also presented in Table 6. The isotopic composition of reprocessed uranium in the REMIX-B fuel shows that it cannot be further used as the fuel in the closed nuclear fuel cycle (CNFC) of thermal neutron reactors without additional removal of ^{232}U and ^{236}U.

The equivalent doses were calculated for the REMIX-B fuel at a distance of 30 cm from the FA surface. The data obtained is presented in Table 5 and in Fig. 4. When reprocessed uranium is enriched, the content of ^{232}U in it increases and, accordingly, the contribution of its decay products to the dose rate increases as well when compared to that for the REMIX-A fuel. Due to the plutonium content in the fuel increasing to 3%, the contribution of plutonium isotopes to the dose rate also increases (Fig. 7).

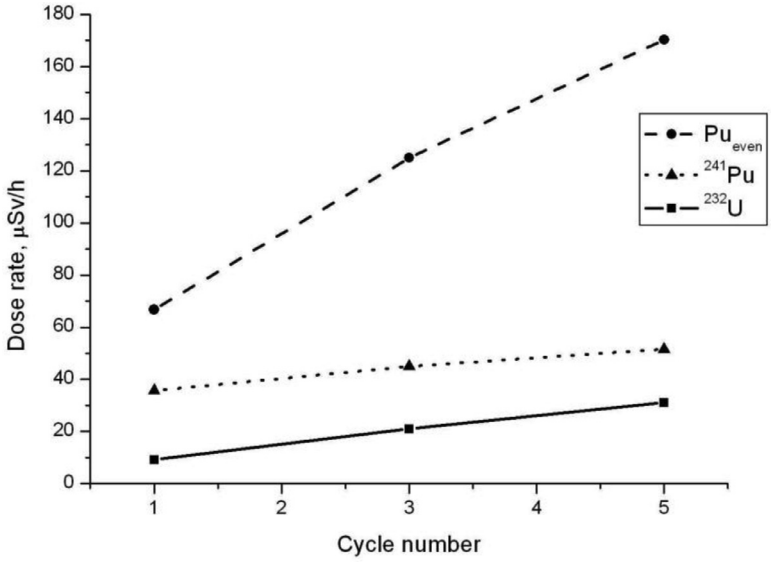

Fig. 6. Contribution of different isotopes to the equivalent dose rate from an FA with fresh REMIX-A fuel for 5 recycles at a distance of 30 cm from the FA surface.

Table 6
U and Pu content (kg/THM) in REMIX-B fuel.

Composition	Uranium–plutonium fuel, kg/t of initial fuel				
	Fresh	Burn-up, GWd/THM			
		49.2	60	65	70
^{232}U	1.58×10^{-5}	1.62×10^{-5}	1.83×10^{-5}	1.94×10^{-5}	2.05×10^{-5}
^{235}U	43.7	19.4	15.4	13.7	12.1
^{236}U	22.0	25.8	26.1	26.2	26.2
^{238}U	904.2	926.8	930.4	932.0	933.3
Σ(U)	970.0	972.0	972.0	971.9	971.8
Pu (even)	10.5	12.2	12.8	13.0	13.3
Pu (odd)	19.5	15.8	15.3	15.1	14.9
Σ(Pu)	30.0	28.0	28.0	28.1	28.2
U + Pu	1000.0	1000.0	1000.0	1000.0	1000.0
Eff. enrich., %	4.65	1.66	1.18	0.99	0.80

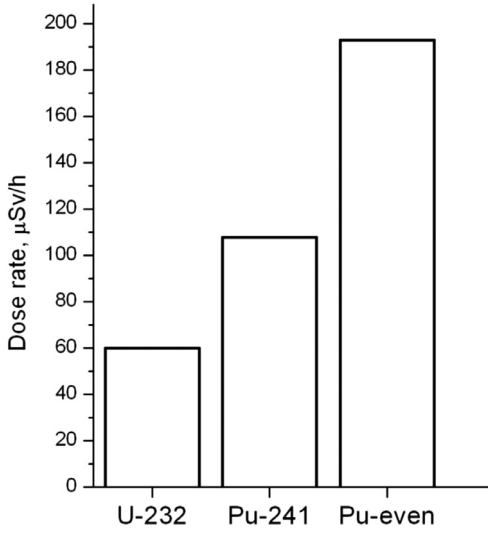

Fig. 7. Contribution of uranium and plutonium isotopes to the equivalent dose rate for REMIX-B fuel.

Since the content of the ^{232}U and ^{241}Pu decay products (^{208}Tl and ^{241}Am respectively) depends heavily on the fuel post-fabrication decay time, the variation in the equivalent dose rate from FAs with fresh REMIX fuel, depending on the decay time, was analyzed in an additional study. The study was conducted on fresh REMIX-B fuel (the fuel composition is given in Table 6). To that effect, the EASY-2010 code was used to calculate the gamma spectra from fresh fuel for different decay times, and then the equivalent dose rate from FAs with this fuel was calculated using the MCNP code. The results of the study are presented in Fig. 8.

As can be seen from the figure, the equivalent dose rate from fresh REMIX fuel increases greatly with its decay time, so minimizing the dose rate from FAs with fresh REMIX fuel requires the time between the fabrication of the fuel and its loading into the reactor to be cut to the maximum.

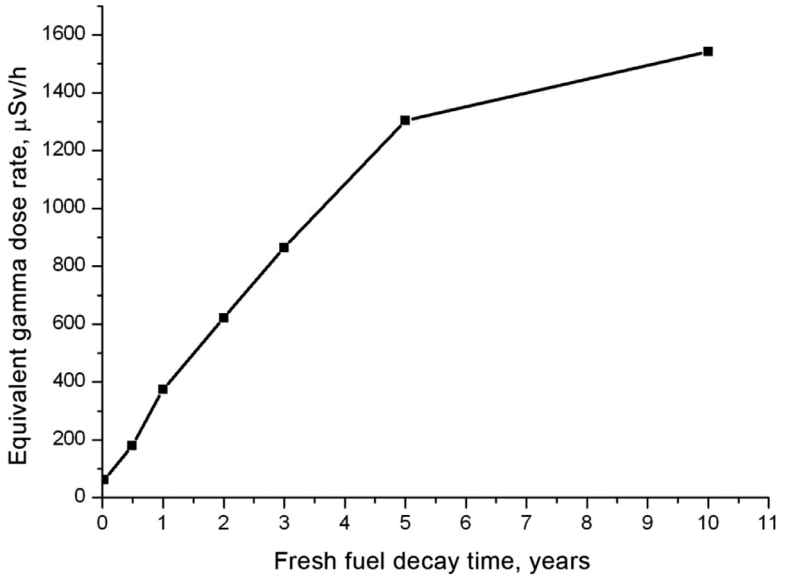

Fig. 8. Equivalent gamma dose rate from an FA with fresh REMIX-B fuel versus the fresh fuel decay time after fabrication.

Conclusion

The changes in the isotopic composition of three fresh REMIX fuel types in the process of recycling in VVER-1000 reactors were calculated. It has been shown that it is possible to recycle the REMIX-A and REMIX-A2 fuels more than five times without a major decrease in the fuel nuclear value.

Uranium and plutonium recycling leads to an increase in the equivalent dose rate from fresh fuel. The major contributors to the dose rate are even plutonium isotopes the content of which in the fuel increases with each recycle. Other key contributors to the dose rate are ^{232}U and ^{241}Pu decay products. The contribution from ^{232}U is the greatest in the event of REMIX-B fuel due to an increase in its content in the process of uranium enrichment. Switching to REMIX fuel is most likely to require extra personnel protection in the FA fabrication and transportation. To minimize the dose rate contribution from ^{232}U and ^{241}Pu decay products in fresh REMIX fuel, the time of the fuel delivery to reactors shall be cut to the maximum.

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