Comparative Assessment of Thermal and Radiation Characteristics of Fresh and Spent REMIX Fuel of Different Designs

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Abstract. The existing structure of the Russian (and world) nuclear power system is based on reactors with the thermal spectrum of neutrons. It is sufficiently provided with resources and fuel fabrication facilities and focused on the operation in the open nuclear fuel cycle. However, the development of nuclear energy industry on such a basis leads to exhaustion of natural uranium resources and accumulation of spent nuclear fuel which in its term leads to economic burden for utilities. In this situation the development of technologies of spent nuclear fuel management and radioactive waste treatment is the key factor for sustainable development of the entire energy system. The aim of this work is to estimate thermal and radiation characteristics of fresh and spent REMIX fuel of different designs. Authors selected technological scenarios and developed physical models to analyse options of application of REMIX fuel for closing nuclear fuel cycle in a nuclear power system based on LWRs. Thermal and radiation characteristics were estimated in terms of recycling of regenerated nuclear materials after SNF reprocessing: activity, heat release, intensity and spectrum of neutron and gamma radiation, dose rates. Selection of the limits (content of even uranium isotopes, spent fuel heat release) of applicability of the REMIX technology in the context of current transport and technological opportunities is carried out.

1. Introduction
Nowadays the structure of the Russian nuclear energy system is based on light water reactors (LWR) and aimed at work in an open fuel cycle (OFC). This structure is well provided with resources and industrial technologies which proved their reliability. But there are several reasons to change the direction and develop technologies of closing fuel cycle within nuclear energy system based on LWRs:

- limited reserves of “cheap” (<80$/kg) natural uranium for fuel fabrication;
- a large amount of accumulated spent nuclear fuel (SNF) at nuclear power plants and storages (~22000 t) and annual increase of its amount (~850 t);
- long-term storage postpones the solution of the problem and leads to financial liabilities and additional expenses for SNF management;
- closing of fuel cycle is one of the main goals of the Rosatom but key role in this scenario is given to reactors with fast neutron spectrum and there is an uncertainty with the commissioning of nuclear power plants with such type of reactors;
- SNF reprocessing technologies industrially developed in Russia;
There are different options of reprocessed nuclear materials involvement in the LWR fuel cycle.

1) **Regenerated uranium dioxide fuel (REPU).** This fuel is produced from uranium extracted during the reprocessing of SNF. REPU fuel can be re-enriched or mixed with the fraction of enriched or depleted uranium. It allows us to save up to 15-20 % of natural uranium compared to OFC. It also allows to perform multiple recycling of uranium in LWRs. On the other hand we need to control uranium isotopic composition because of the accumulation of the U-232 [1] which affects the radiation situation on the fuel fabrication facility, U-234 [2] which affects the quality of the re-enrichment process, U-236 which affects the neutron characteristics of the fuel. We also need to solve problem of the extracted plutonium storage.

2) **Dioxide MOX fuel.** This fuel is produced from plutonium extracted during reprocessing of SNF and depleted uranium. This option allows to save up to 10-15 % of natural uranium but degradation of plutonium isotopic composition during irradiation in thermal spectrum does not allow recycling it multiple times. Other disadvantages are that reprocessed uranium should be stored somewhere and high amount of Pu in fuel complicates fresh fuel fabrication and management.

3) **Dioxide REMIX fuel.** This fuel is fabricated from a mixture of regenerated uranium and plutonium extracted during the reprocessing of SNF. It can be re-enriched and mixed with fractions of enriched or depleted uranium as well as REPU. This variant allows to save up to 20-30 % of natural uranium, to recycle nuclear materials multiple times, to use reprocessed plutonium instead of storing it. On the other side we have to control uranium isotopic composition and the technology of fabrication of this type of fuel is still not well developed.

The main objective of our efforts in this work is the analysis of the radiation and thermal characteristics of fresh and spent REMIX fuel compared to the regular uranium dioxide fuel. Calculations are performed for four different designs of REMIX fuel in order to choose the best one in terms of radiation and heat burden in fuel cycle.

2. **Technological scenarios of the REMIX fuel application**

2.1. **REMIX-A**

The classic approach to the fabrication of REMIX fuel [3, 4] considered in a variety of works is to mix the reprocessed uranium and uranium-plutonium fractions with natural uranium enriched up to 20% by U-235. This concept assumes 100% loading of the REMIX fuel in the core. Reprocessed materials are obtained from the previous irradiation cycle in LWR. Schematic diagram of the recycle process for REMIX-A is presented in figure 1.

![Figure 1. Principal scheme of the recycling for option REMIX-A.](image-url)
The calculated characteristics of the material flows for all considered options of REMIX fuel application in LWR correspond to the standard parameters for a one-and-a-half-year fuel company with an average burnout of 47 MW\textsuperscript{day}/kgHM.

The presented approach does not allow to achieve a complete recycling of regenerated materials in a closed system since the necessary fraction of enriched natural uranium is \(~18\%\) wt. Some quantity of the REPU should be sent to the storage facility.

2.2. REMIX-C

The technology process of the REMIX fuel fabrication can be divided into two chains according to the obtained flows of regenerated materials during the recycling. The mixture of uranium and plutonium is sent directly to the fabrication of the REMIX fuel while the fraction of REPU is sent to the additional re-enrichment. Schematic diagram of the recycle process for REMIX-C is presented in figure 2.

![Figure 2. Principal scheme of the recycling for option REMIX-C.](image)

This approach makes it possible to fully use the potential of the regenerated uranium and plutonium. The fraction of REPU is mixed with the flow of natural uranium during re-enrichment. This allows to control the amount of the even isotopes of the uranium in the fuel. This technological approach allows to improve saving of natural uranium and amount of separation work compared to the REMIX-A. The main difference between these scenarios is reduction of U-236 quantity during re-enrichment because partially it goes to the waste. REMIX-C option is also characterized by a reduced accumulation of U-232, Pu-236 and Pu-238 after several cycles.

2.3. REMIX-Het

There is an option of dividing material flows during reprocessing to produce REMIX and uranium dioxide fuel separately. These two types of fuel in the end are placed in a heterogeneous fuel assembly [5]. Such an approach allows to separate technological processes of fabrication of hazardous REMIX fuel and relatively safe fabrication of REPU fuel. Figure 5 presents a schematic diagram of fuel rod placement in the fuel assembly.

The content of plutonium and the number and arrangement of REMIX fuel rods in the FA is selected to ensure the same average burnup in the uranium and REMIX fuel rods. This approach makes it possible to obtain a balanced field of energy release during the entire fuel company. The spatial separation of enriched uranium and plutonium enhances the fission properties of FA due to less U-235 shielding as well as better fission of plutonium in the thermal spectrum. The content of plutonium in REMIX fuel rods is \(~5\%\) wt which is almost 2 times less than the similar content of plutonium in MOX FA with the same burn-up parameters. The total content of plutonium in heterogeneous REMIX FA corresponds to its content in REMIX-A and REMIX-C. It should be noted that the number of uranium-plutonium fuel rods in the FA increases from cycle to cycle. The balance of nuclear materials in fresh and spent fuel should be constant.
2.4. **REMIX-B**

Option REMIX-B [5] corresponds to the approach of option REMIX-C but with a higher amount of plutonium (4-6%) and excluding involvement of natural uranium fraction. Fuel multiplying properties are provided by additional enrichment of the regenerated uranium fraction and increased content of plutonium in. This approach is interesting due to minimizing the fabrication of expensive and difficult in management REMIX fuel. Schematic diagram of the recycle process for REMIX-B is presented in figure 5.

It is worth noticing that all previous options considered for scenario “one-to-one” which means that SNF from one reactor is used to produce fresh fuel for one reactor. In case of REMIX-B we assume that there is a system of four reactors. One of them works on REMIX-B. Three others work on regular uranium dioxide fuel. SNF from all four reactors is used to produce REMIX-B fuel to provide enough amount of plutonium.
It should be noted that selected variants do not describe the full range of options for the use of uranium-plutonium oxide fuel in LWRs. Consideration is limited to the existing most reasonable technological approaches that can be implemented in the short term period.

3. Calculation models for estimation of thermal and radiation characteristics

Calculations are performed using SCALE code system [6] which is designed for nuclear and radiation safety analysis. SCALE code system is a set of tools that allow to carry out calculations of criticality, fuel burnout, materials activation, characteristics of radiation sources and radiation protection. In this work we used two computational sequences under control of TRITON and MAVRIC modules. TRITON sequence manages the preparation of the required cross sections and the calculation of neutron transport in two-dimensional configurations by NEWT module. ORIGEN module in combination with NEWT module provides the calculation of the isotope composition, the intensity of radiation sources and decay heat. At this stage calculations are carried out using 2-D FA models in conditions corresponding to the operation mode in a reactor with constant power. Calculation models are provided in figure 6.

MAVRIC sequence organizes the calculation of particle transport by the Monte-Carlo method and estimation of gamma and neutron fluxes for further determination of dose rates at points at a given distance from the external surface of the 3-D full scale FA. It is worth noticing that in this work we do not consider any protection for FA.

Figure 5. Principal scheme of the recycling for option REMIX-B.

Figure 6. Calculation models of:

a – REMIX-A, -C, -B; b – REMIX-Het (red – UO₂, green – REMIX).
Irradiation is carried out for the following case:
- time of storage for fresh fuel before irradiation – 2 years;
- time of storage for spent fuel – 5 years;
- maximum amount of U-232 – $5 \times 10^{-7}$ wt;
- maximum amount of U-234 – 20 mg/1 g of U-235.

A set of characteristics estimated for the initial assessment of the radiation situation and heat release in the fuel cycle:
- activity;
- heat release;
- intensity and spectrum of gamma and neutron radiation;
- effective dose from FA.

4. Results

4.1. Fresh REMIX fuel

Analysis of the results of estimation of the radiation characteristics of REMIX fuel before first irradiation cycle showed that its activity is several orders of magnitude higher than the activity of regular uranium dioxide fuel (figure 7). In turn, among the different designs of FA with REMIX, REMIX-B stands out, exceeding other options about 5 times due to the high content of plutonium while the activity of fresh REMIX-A, -C and -Het is practically the same. In general, REMIX fuel activity slightly decreases with increase of storage time before irradiation.

Figure 7. Activity of the fresh fuel before first irradiation cycle.

Figure 8. Heat release of the fresh fuel before first irradiation cycle.
A similar situation is observed for heat release (figure 8), intensity of gamma (figure 9) and neutron radiation (figure 10). However, unlike activity heat release and intensity of gamma radiation increases with increase of storage time. The intensity of neutron radiation slightly decreases.

![Figure 9. Intensity of gamma radiation of the fresh fuel before first irradiation cycle.](image)

![Figure 10. Intensity of neutron radiation of the fresh fuel before first irradiation cycle.](image)

The estimation of the dose rates for fresh fuel showed that the REMIX-A and-C designs in this indicator are an order of magnitude higher than the regular UO$_2$. The dose rate from a REMIX-Het about 15% lower than the dose rate from REMIX-A and – C. REMIX-B exceeds them about 70%. Figure 11 shows the dose rate of unprotected FA versus the distance from it after 2 years of storage.

![Figure 11. Dose rates of the fresh fuel before first irradiation cycle.](image)
Described above results are obtained for fuel compositions before first irradiation cycle. The ratio of radiation and thermal characteristics of different fuel designs is similar on every next cycle. Analysis of the activity of different fuel designs during six cycles of irradiation showed that it increases dramatically on the second cycle compared to the first (~35%) for every REMIX fuel. Further difference is not so high and decreases from cycle to cycle due to reduction of the Pu-241 and Pu-238 accumulation rate (figure 12).

The change in heat release even sharper between first and second cycles (~55%) but the downward trend is the same as for an activity (figure 13). Similar situation is for intensity of the gamma and neutron radiation and the dose rates (figures 14, 15, 16).
It should be noted that the number of cycles for REMIX-Het are limited because of the limitation of the maximum amount of even isotopes of the uranium in the reprocessed materials. For REMIX-B there is also a limitation because of the heat release due to necessity of providing its value from one FA after irradiation and storage in pool on the level of 2 kW \[7\] (table 1) because of the technical characteristics of the transport cask. This level of heat release should be provided not later than 10 years after irradiation to ensure stable operation of the nuclear energy system and to meet economic requirements.

**Table 1.** Heat release from SNF in one FA after storage in reactor pool, W

| Cycle | REMIX-A       | REMIX-C       | REMIX-B       | REMIX-Het      |
|-------|---------------|---------------|---------------|----------------|
| 1     | 1.6761E+03    | 1.6634E+03    | 2.0097E+03    | 1.5827E+03     |
|       | (5 years of storage) | (5 years of storage) | (7 years of storage) | (5 years of storage) |
| 2     | 1.9782E+03    | 1.9502E+03    | 2.2963E+03    | 1.8236E+03     |
|       | (5 years of storage) | (5 years of storage) | (10 years of storage) | (5 years of storage) |
| 3     | 1.9660E+03    | 1.9196E+03    | -             | 1.9903E+03     |
|       | (6 years of storage) | (6 years of storage) |               | (5 years of storage) |
| 4     | 2.0014E+03    | 2.0041E+03    | -             | -              |
|       | (6,5 years of storage) | (6 years of storage) |               |               |
| 5     | 2.0062E+03    | 2.0038E+03    | -             | -              |
|       | (7 years of storage) | (6,5 years of storage) |               |               |
| 6     | 1.9830E+03    | 1.9804E+03    | -             | -              |
|       | (8 years of storage) | (7 years of storage) |               |               |
4.2. Spent REMIX fuel
Analysis of the results showed that the activity of spent REMIX fuel increases compared to regular UO₂ fuel during storage after irradiation. This is due to the fact that at the beginning of storage interval activity is mainly determined by fission products which amounts depends on burnout and the same for all fuels. In the end of storage interval activity (figure 17) is determined by the content of Pu-241 and Pu-238 which accumulates the most in REMIX-B. Similar situation is observed for heat release (figure 18).

![Figure 17. Activity of the spent fuel after first irradiation cycle.](image)

![Figure 18. Heat release of the spent fuel after first irradiation cycle.](image)

The intensity of gamma radiation is approximately the same during all storage time (figure 19). However, it is necessary to pay attention to the spectrum of the gamma radiation (figure 20). The total intensity is determined by low-energy photons. Comparative analysis of the spectra for the considered options showed that the intensity of high-energy photons (>5MeV) for spent REMIX fuels is about an order of magnitude higher than for UO₂ fuel. One of reasons for that is accumulation of U-232 in REMIX fuel and its further decay into Tl-208. In case of estimation dose rates from transport cask with SNF these photons will play a major role which will affect the difference between results for REMIX and UO₂ fuels. But in our case dose rates are practically the same (figure 21) because we do not consider radiation protection from FA.
Figure 19. Intensity of gamma radiation of the spent fuel after first irradiation cycle.

Figure 20. Spectrum of gamma radiation of the spent fuel after first irradiation cycle.

Figure 21. Dose rates of the spent fuel after 5 years of storage after first irradiation cycle.

The intensity of neutron radiation of spent REMIX fuel is several times higher than the intensity of neutron radiation of UO$_2$ fuel (figure 22) which is associated with a high content of plutonium in REMIX. Since it is the amount of plutonium that determines the intensity of the radiation, REMIX-B exceeds the other types of REMIX fuels by about two times.
Figure 22. Intensity of neutron radiation of the spent fuel after first irradiation cycle

The ratio of radiation and thermal characteristics of different fuel designs is similar on every next cycle. The activity of spent REMIX fuel increases from cycle to cycle compared to UO$_2$ fuel (figure 23) due to accumulation of plutonium.

Figure 23. Activity of the spent fuel during six cycles.

Changes in heat release and intensity of neutron radiation have a similar character but differences between cycles are higher for them (figure 24, 25).

Figure 24. Heat release of the spent fuel during six cycles.
The intensity of gamma radiation practically does not change from cycle to cycle (figure 26). It is seen from figure above that intensity of neutron radiation around seven orders in magnitude lower than intensity of gamma radiation. Due to that dose rates are determined mainly by photons and does not change from cycle to cycle as well.

5. Conclusions
The main achievements of this work are the following:
1) Selection of technological scenarios and development of physical models for performing calculation of material flows in nuclear fuel cycle and analyzing options of application of REMIX fuel for closing nuclear fuel cycle in nuclear power system based on LWRs.
2) Estimation of thermal and radiation characteristics of REMIX fuel compared to uranium dioxide fuel.
3) Selection of the limits (content of even uranium isotopes, spent fuel heat release) of applicability of the REMIX technology in the context of current transport and technological opportunities.
4) It was shown for the fresh fuel that:
   ✓ the activity of REMIX fuels by several orders of magnitude higher than the activity of UO₂ fuel;
   ✓ among the various options of REMIX fuels, REMIX-B activity exceeds the activity of other options about 5 times due to the high content of plutonium;
   ✓ similar situation is observed for heat release, intensity of gamma and neutron radiation;
✓ the values of dose rates for fresh REMIX-A and-C are an order of magnitude higher than the corresponding values for UO$_2$;
✓ dose rate for REMIX-Het is around 15% lower than for REMIX-A , and REMIX-B dose rate exceeds the REMIX-A by about 70%.
✓ there is a sharp increase of values of heat and radiation characteristics on the second cycle compared to the first one;
✓ further increase on the next cycles is not so significant, and its value is decreasing from cycle to cycle due to reduction of the Pu-238 and Pu-241 accumulation rate;
5) It was shown for the spent fuel that:
✓ activity and heat release of spent REMIX fuel increases relatively to the UO$_2$ spent fuel with increase of storage time;
✓ values of total intensity of gamma radiation, which are determined by low-energy photons, are approximately the same for all fuel compositions;
✓ but amount of high-energy photons (> 5 MeV) in all REMIX fuels is about one order of magnitude higher than in UO$_2$;
✓ values of total intensity of neutron radiation for spent REMIX fuels are several times higher than for UO$_2$;
✓ estimation of changes of the heat and radiation characteristics of spent REMIX fuels during 6 cycles showed that all of them with the exception of the intensity of gamma radiation increase from cycle to cycle compared to UO$_2$ mainly due to the accumulation of Pu-238 and Pu-241, U-232.

Comparative assessment showed that the most successful REMIX designs in terms of radiation characteristics are REMIX-C and REMIX-het. However, accumulation of even isotopes of uranium limits number of cycles which makes REMIX-Het option less preferable.

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