Increase of inherent protection level in spent nuclear fuel

Alexey Krasnobaev, Edward Kryuchkov* and Vasily Glebov
Moscow Engineering Physics Institute (State University), 31 Kashirskoe shosse, Moscow, 115409, Russian Federation

Abstract

The paper is devoted to upgrading inherent proliferation protection of fissionable nuclear materials (FNM). Some possibilities were investigated to form high radiation barrier inside spent fuel assemblies (SFA) discharged from power reactors of VVER-1000 type and research reactors of IRT type. The radiation barrier is estimated in the terms of rate of equivalent dose (RED) at 30-cm distance from SFA. The values of RED were calculated with application of the computer code package SCALE 4.3.

The paper considers the criteria adopted for estimation of FNM proliferation resistance. The paper presents numerical results on a component-wise analysis of the radiation barrier in SFA from reactors of VVER-1000 and IRT type and on capability of various radionuclides to prolong action of the radiation barrier. Isotopic admixtures were selected and amounts of these admixtures were evaluated for significant prolongation of the radiation barrier action at the levels of the radiation standards used for estimation of FNM proliferation resistance. The paper considers vulnerability of the radiation barriers in respect to thermal processing of spent fuel.

KEYWORDS: spent nuclear fuel, radiation barrier, rate of equivalent dose, standard of proliferation resistance, isotopic admixtures, vulnerability of radiation barrier.

1. Introduction

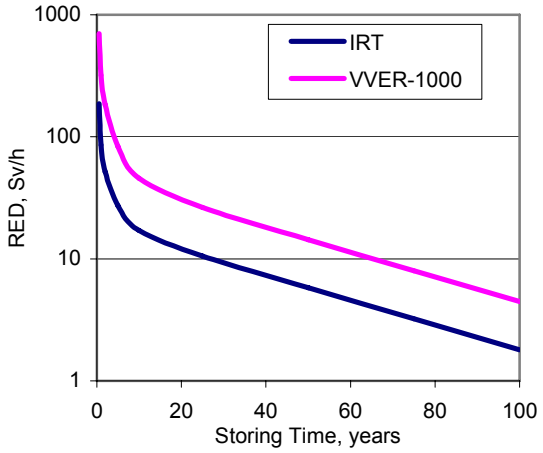
Currently, a possibility for uncontrolled FNM proliferation is regarded as one of main obstacles against wide deployment of nuclear power plants all over the world. Threat of FNM diversion may be reduced by using both exterior security measures and some interior barriers, including the radiation barriers created by emitters of high-energy gamma-rays.

The paper considers some possibilities to upgrade inherent proliferation resistance of FNM, which are contained in SFA from reactors of VVER-1000 and IRT type. In particular, some possibilities were investigated on forming the radiation barriers in SFA from power and research reactors under conditions of their lengthy storing.

Spent nuclear fuel contains large amounts of fission products, which provide intense but rapidly decaying radiation barrier (see Fig. 1).

* Corresponding author, Tel. +7(495) 323-9242, Fax +7(495) 324-7026, E-mail: EFKryuchkov@MEPHI.RU

Fig. 1: RED at 30-cm distance from SFA of research reactor of IRT type and power reactor of VVER-1000 type.



Time dependence of the radiation barrier can be described by the following parameters [1]:

- level — minimal RED value at specified distance from SFA; this is a characteristic that defines proliferation resistance of FNM in spent fuel (the level also could be expressed in the terms of time till receiving the lethal dose at specified distance from the radiation source);
- duration — time period, when RED exceeds specified minimal level.

The following proliferation resistance criteria developed early by the US Academy of Sciences were used in the paper for comparative analysis of the radiation barriers:

1. Proliferation protection standard for FNM, which are contained in SFA: RED value must be above 1 Sv/h at 1-m distance from SFA (Spent Fuel Standard-First, or SFS_F, [4]).

2. Enhanced proliferation protection standard, which was recently adopted by the US Academy of Sciences (Spent Fuel Standard-Current, or SFS_C, [5]). According to the standard, proliferation resistance of FNM must be above the average proliferation resistance of plutonium, which is contained now in spent nuclear fuel (SNF) discharged from power reactors. According to numerical evaluations [5], RED at 30-cm distance from the object containing FNM must be above 20 Sv/h.

The values of RED at 30-cm distance from SFA are regarded as key parameters of inherent radiation barrier. Also, it may be convenient to express proliferation protection in the terms of time till receiving the specified gamma-radiation dose:

$$T_{Dose} = \frac{Dose}{RED} \cdot 60 , \qquad (1)$$

where Dose — specified dose of gamma-radiation, Sv; T_{Dose} – time till receiving the dose, minutes; RED – rate of equivalent dose at 30-cm distance from SFA, Sv/h.

Impact of gamma-radiation on human organism is estimated with application of two boundary values of exposure dose:

a) Dose SD = 1 Sv. Under exposure dose above SD level (for time period from several seconds to three days), acute radiation disease is developing in the human organism [2]. Then, according to formula (1), T_{SD}(IRT-3M) ≈ 8 minutes, T_{SD}(VVER-1000) ≈ 20 minutes for SFS_F standard of proliferation protection.

b) Dose LD = 4,5 Sv. Exposure dose above LD level leads to the lethal outcome in 505 of all cases [1, 3]. So, according to formula (1), T_{LD}(IRT-3M) = T_{LD}(VVER-1000) ≈ 13,5 minutes for

SFS_C standard of proliferation protection.

So, it may be concluded that both standards of FNM proliferation protection (SFS_F and SFS_C) correspond to short-term receiving gamma-radiation doses at SD and LD levels.

Using these standards of FNM proliferation protection, duration of the radiation barrier action can be evaluated. The durations of the radiation barrier action in SFA from power light-water reactor and research reactor are presented in Table 1 for SFS_F and SFS_C standards. General conclusion from Table 1 consists in insufficient FNM proliferation protection for lengthy (> 25 years) SFA storing time. In the first turn, it concerns SFA from research reactor, where insufficient proliferation protection takes place even for the less stringent SFS_F criteria.

Table 1: Duration (in years) of the radiation barrier action for RED ≥ SFS_F and RED ≥ SFS_C in SFA from power reactor and research reactor

	SFS_C	SFS_F
VVER-1000	36	117
IRT-3M	4	20

So, long-term SFA storing leads to sharp increase of FNM vulnerability in respect to any unauthorized actions. This threat can be diminished either by urgent SNF reprocessing or by using fuel with upgraded inherent proliferation resistance (for example, fuel with upgraded radiation barrier). Presently, SNF reprocessing strategies are often related with large international nuclear technology centers [6], construction of which requires significant time expenses. Therefore, the paper considers a variant of SNF storing with upgraded properties on inherent proliferation resistance.

In this connection, the problem was defined to investigate some possibilities for upgrading the radiation barrier in SFA through introduction of isotopic admixtures into fresh fuel of power and research reactors. Fuel assemblies of power VVER-1000 reactor and fuel assembly IRT-3M of research reactor were used as objects of investigation. The computer code package Для основных расчетов использовался программный комплекс SCALE 4.3 [7] is applied for radiation characterization of SNF from light-water systems (determination of RED at specified distance from SFA, content of gamma-emitting radionuclides in SNF at different time moments and some other parameters).

2. Selection of isotopes for upgrading the radiation barrier in SFA of power reactor

2.1. Component-wise analysis of the radiation barrier in SFA of light-water reactor of VVER-1000 type

The first stage of the analysis consisted in determination of those radionuclides contained in SFA, which:

a) give main contribution into RED during 25-year storing time and longer;

b) are capable, in principle, to prolong action of the radiation barrier under lengthy storing time.

Using design parameters of VVER-1000 core and fuel assembly [7], geometrical models were prepared for micro- and macro-cells needed for input data file of the computer code SCALE 4.3. Then, the computations were carried out to determine SNF isotopic composition upon its discharge from the reactor core (cooling time - 10 days). Following from the requirement to improve the radiation barrier for long-term (longer than 25 years) storing time, some middle-lived and short-lived gamma-emitting isotopes were selected from those contained in SNF. Some isotopes from handbook [9] and scientific publication [3] were added to the list. Final list of gamma-emitting isotopes consisted of ^{231}Pa; ^{232}U; $^{236, 238, 239, 240, 241}Pu$; $^{241, 242m, 243}Am$; $^{243, 244, 245, 246}Cm$; ^{249}Bk; $^{249, 250, 251, 252}Cf$; ^{90}Sr; ^{94}Nb; ^{106}Ru; ^{125}Sb; $^{134, 137}Cs$; ^{144}Ce; ^{147}Pm; ^{151}Sm; $^{152, 154,}$

^{155}Eu. These isotopes were regarded as candidates for further analysis of the radiation barrier.

The contributions of these isotopes into total RED from SFA were determined with application of the computer code SCALE 4.3. As it was anticipated, isotope ^{137}Cs is a main contributor to total RED under long storing time of SFA.

Amongst another isotopes, those isotopes, which are able to prolong action of the radiation barrier, represented a particular interest. So, a group of actinide isotopes (except of ^{94}Nb) was selected, which could increase or keep constant in time the value of RED (see Fig. 2 and Fig. 3, respectively). It can be seen that, for typical concentrations of these isotopes in SNF, their contributions are relatively small. But, at current stage of investigation, the absolute values of contributions were not taken into consideration. This is a matter of further stages.

In addition to ^{137}Cs, some other fission products (^{90}Sr, ^{106}Ru, ^{144}Ce, ^{152}Eu, ^{154}Eu and ^{155}Eu) were analyzed. Despite substantially shorter lifetimes of these isotopes in comparison with isotopes of actinide group, they are full-value candidates for prolongation of the radiation barrier action. Particular attention should be given to stable isotope ^{59}Co, which, under neutron irradiation, converts into highly active isotope ^{60}Co.

Fig. 2: Contributions of ^{231}Pa, ^{232}U, ^{241}Am and ^{249}Cf into RED as functions of SFA storing time

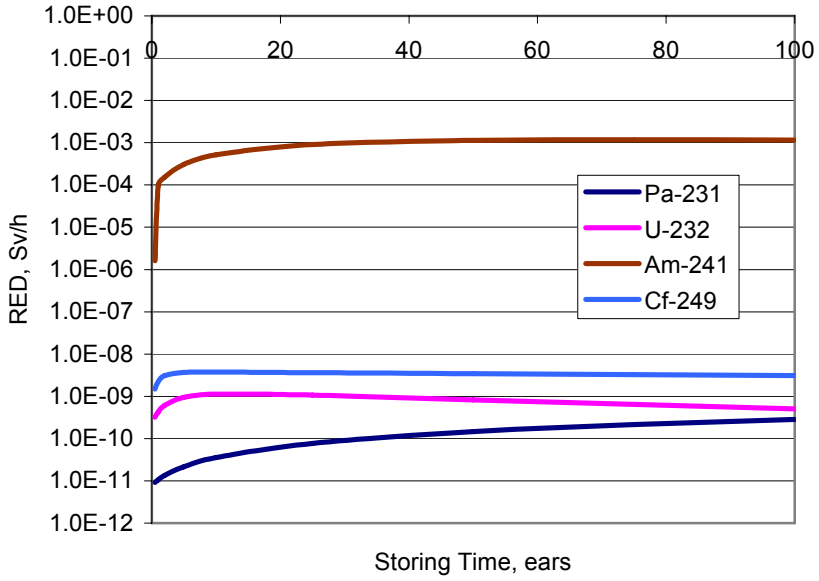
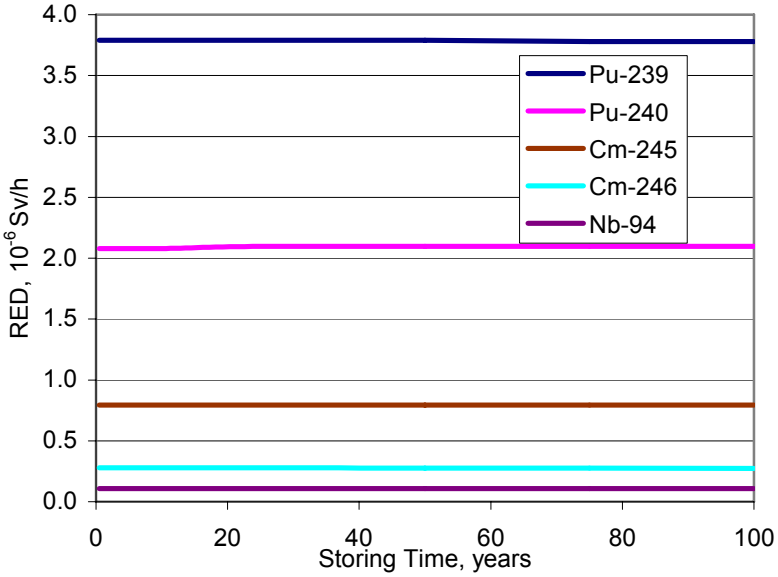


Fig. 3: Contributions of ^{239}Pu, ^{240}Pu, ^{245}Cm, ^{246}Cm and ^{94}Nb into RED as functions of SFA storing time



So, analysis of isotopic composition and radiation properties of SNF from power reactor of VVER-1000 type allowed us to select seventeen isotopes from long-lived actinides, fission products and light elements. These isotopes represent a potential interest for upgrading proliferation resistance of FNM contained in SFA under its long-term storing and require further studying their effects on parameters of the radiation barrier.

2.2. Potential capability of radionuclides on prolongation of the radiation barrier action in SFA from power reactor of VVER-1000 type

In order to evaluate potential of these radionuclides, some variations were introduced into fresh fuel composition, and appropriate changes of RED from SFA of power VVER-1000 reactor were calculated for long-term storing. The variations of fuel composition included relatively small admixture (0,2 % HM) of the selected isotopes. The values of RED from SFA were calculated for all the isotopic compositions for 100-year storing time.

Application of the variation procedure demonstrated that only about 50% (below 50% for SFS_F criterion) of these isotopes can prolong action of the radiation barriers, at least, on several years. The isotopes and the effects of their admixing into fresh fuel composition on prolongation of the radiation barrier action are presented in Table 2.

The effects of admixing these isotopes on prolongation of the radiation barrier action are shown in Fig. 4 in the terms of time till receiving the lethal dose. It can be concluded that isotopes ^{232}U, ^{137}Cs, ^{152}Eu and ^{59}Co can be used for prolongation of the radiation barrier action at high levels of proliferation resistance, i.e. at short time till receiving the lethal dose дозы ($T_{LD} \leq$ 13 minutes). Another isotopes do not contribute significantly because of small gamma-radiation intensity and/or short half-life. The latter two isotopes can be used only for short-term (no longer than 10 years) upgrading of proliferation resistance.

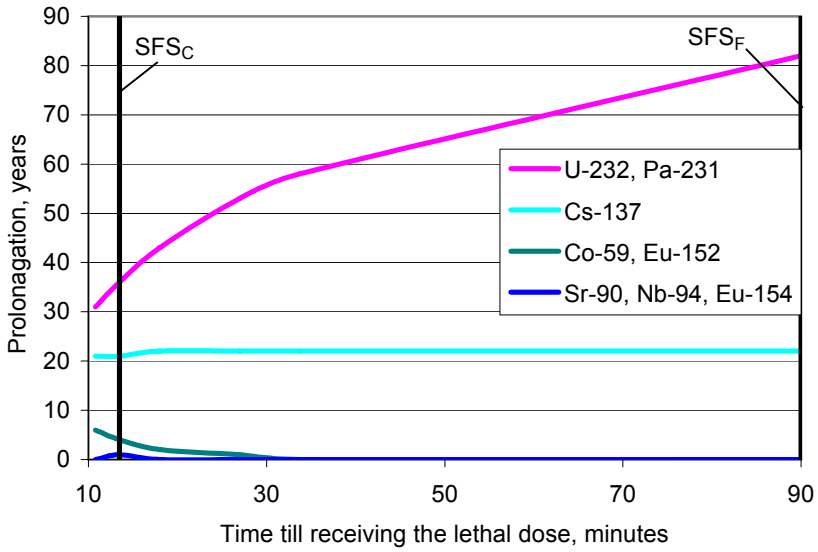
Only two isotopes (^{232}U and ^{137}Cs) are able to prolong action of relatively low radiation barriers ($T_{LD} > 15$ minutes). Isotopes ^{152}Eu and ^{60}Co are excluded from analysis because of short half-lives, which are lower by factors of 8,8 and 22,2, respectively, than the specified duration of

the radiation barrier action (117 years).

Table 2: Duration (years) of the radiation barrier action for two SFS criteria from SFA of power reactor with/without isotopic admixtures.

	SFS_C	SFS_F
Without admixtures	36	117
With ^{231}Pa	72	199
With ^{232}U	72	199
With ^{90}Sr	37	118
With ^{137}Cs	57	139
With ^{94}Nb	37	119
With ^{59}Co	39	117
With ^{152}Eu	40	117
With ^{154}Eu	37	117

Fig. 4: Prolongation of the radiation barrier action as a function of time till receiving the lethal dose T_{LD}



3. Selection of isotopes for improvement of the radiation barrier parameters in SFA from research reactor

The following two problems were resolved for analysis of a possibility to prolong action of the radiation barrier in SFA IRT-3M:

Problem 1: Evaluate effects of small isotopic admixtures into fresh fuel composition on RED from SFA IRT-3M under its long-term storing.

For resolving the problem, fresh fuel compositions were varied by admixing 0,2% HM of the following isotopes: ^{232}U, ^{90}Sr, ^{137}Cs, ^{94}Nb, ^{59}Co, ^{152}Eu and ^{154}Eu. Time dependencies of RED at 30-cm distance from SFA were calculated for all the fuel compositions and for 100-year storing time.

However, the calculations demonstrated rather small relative extension of the radiation barrier

action at SFS_F level (about 10-20% only). It may be explained by insufficient effect of small (0,2% HM) admixtures on duration of the radiation barrier action.

Therefore, the following problem 2 was set up: determine minimal concentrations of admixed isotopes, which would be able to provide action of the radiation barrier for specified time period and at the level higher than specified one.

Two levels of the radiation barrier (SFS_F and SFS_C) were considered in numerical solutions of the problem 2. Let level of the radiation barrier correspond to the lower SFS_F criterion and duration of its action be 100 years. Then, only two isotopes - ^{137}Cs (5,2% HM) and ^{232}U (15% HM) - are applicable for such an extension of the radiation barrier action. Despite a significant percentage of ^{232}U content, real quantity of ^{232}U in one fuel assembly IRT-3M is rather small (about 20 g). Large percentage of ^{137}Cs is explained by the fact that duration of the radiation barrier action (100 years) is significantly longer than half-life of ^{137}Cs. Use of ^{137}Cs as a fuel admixture is reasonable, if duration of the radiation barrier action is comparable with half-life of ^{137}Cs.

If the higher SFS_C criterion is used as a level of the radiation barrier, then extension of the radiation barrier action up to 40 years requires increasing contents of ^{232}U and ^{137}Cs up to 6,1% and 7,9%, respectively. Further extension seems unreasonable because of large consumption of these isotopes for maintaining the higher radiation barrier.

Percentages of isotopic admixtures in fresh fuel for extension of the radiation barrier action at the level of SFS_C criterion from 25 years to 40 years are presented in Table 3. It can be seen that long duration of the radiation barrier action is achievable by proper isotope admixing. However, even 25-year duration requires significant percentage of ^{152}Eu and extremely large percentage of ^{59}Co. It is explained by rather short half-lives of ^{152}Eu and ^{60}Co in comparison with necessary time intervals. Nevertheless, percentages of ^{232}U and ^{137}Cs, whose half-lives are comparable with or longer than necessary duration of the radiation barrier action, remain below 5% HM.

Table 3: Content of isotopic admixtures for extension of the radiation barrier action at the level of SFS_C criterion

Duration, years	Isotopes	Fraction of isotope, % HM
25	^{232}U	4,5
	^{59}Co	34,8
	^{137}Cs	4,8
	^{152}Eu	10,4
30	^{232}U	5,0
	^{137}Cs	5,7
	^{152}Eu	16,5
40	^{232}U	6,1
	^{137}Cs	7,9

4. Impact of SNF reprocessing technologies on vulnerability of the radiation barrier

Presently, SNF reprocessing includes a series of sequential technological procedures, which are carried out after SNF storing in the cooling pools at nuclear power plants. Main contemporary technologies of SNF reprocessing are based on procedures of PUREX process.

Aqueous PUREX-type technologies are very sophisticated and, as a rule, these technologies result in production of high-purity FNM, including plutonium separated from uranium and fission products, and generation of large amounts of highly-active liquid radioactive wastes (RAW).

In parallel with aqueous technologies, alternative non-aqueous (dry) technologies of SNF

reprocessing are under intense development now. These technologies are regarded as relatively less sophisticated ones as compared with PREX-process, and application of dry technologies result in decreasing RAW generation rate. From standpoint of nuclear non-proliferation, advantage of dry technologies consists in the fact that, as a rule, they do not produce high-purity FNM. Dry technologies of SNF reprocessing essentially are high-temperature processes, and hot temperature conditions lead to release of volatile fission products (VFP). Of dry technologies, the first of all, the following two technologies should be noted as the most promising ones, namely pyro-electrochemical technology developed by the Russian research center RIAR (Dimitrovgrad) and DUPIC-technology, joint product of South Korea, Canada and the USA cooperation.

Pyro-electrochemical technology [13] includes the following processes: thermal treatment in vacuum, SNF dissolution in molten salts, electrolysis, collection and removal of fission products.

After reprocessing, fuel contains (in per cents of initial quantity) uranium - 99,5%, plutonium – 98,8% and minor actinides – 70%.

DUPIC-technology [14] has been developed for re-fabrication of SFA from light-water PWR-type reactors into fresh fuel bundles for heavy-water CANDU-type reactors. DUPIC-technology includes the following main stages:

1. SFA dismantling.
2. De-cladding of fuel rods by oxidation in air at 400-450^{o}C.
3. Transformation of fuel pellets into fine powder by alternation of oxidation and reduction reactions in OREOX-process. As a result, main VFP are removed from SNF.
4. Fabrication of fresh fuel pellets, fuel rods and fuel bundles for CANDU reactors.

So, both dry technologies include thermal treatment of SNF. As a consequence, main VFP are removed from fuel (Ru and Tc – 100% removal, Kr, I, Xe and Cs – 98% removal [15]).

Data presented in Table 4 demonstrate that VFP removal results in sharp decrease of RED. In the first turn, this effect is caused by removal of ^{137}Cs, which is a main contributor to RED. Also, VFP release from SNF leads to a multi-fold shortening of the radiation barrier action at the levels of SFS_F and SFS_C criteria (see Table 5). So, SFA rapidly transform into the objects, which are quite vulnerable to any unauthorized actions.

Table 4: Reduction of the radiation barrier by VFP removal from SFA of power and research reactors

Storing time, years	RED(-VFP)/RED* for SFA IRT-3M		RED(-VFP)/RED* for SFA VVER-1000	
	Without admixture	5% ^{232}U	Without admixture	0,2% ^{232}U
0,5	0,83	0,83	0,71	0,71
1	0,59	0,67	0,45	0,48
2	0,32	0,53	0,26	0,32
5	0,17	0,59	0,10	0,27
10	0,15	0,67	0,07	0,34
25	0,14	0,71	0,06	0,43
50	0,13	0,77	0,06	0,53
75	0,12	0,83	0,06	0,59
100	0,12	0,83	0,05	0,67

* RED(-VFP)/RED — ratio of RED without VFP contribution to total RED of SFA.

Table 5: Duration (years) of the radiation barrier action with/without VFP in SNF

	SFA IRT-3M		SFA VVVER-1000	
	With VFP	Without VFP	With VFP	Without VFP
SFS_F	20	2,5	117	9
SFS_C	4	1	36	3

So, it may be concluded that wide use of dry technologies for SNF reprocessing, on the one hand, limits a possibility to produce high-purity FNM. Dry technologies can reprocess highly radioactive SNF. On the other hand, dry technologies are relatively simple instruments for radical reduction of the radiation barriers in SFA from power and research reactors.

One of directions towards decreasing vulnerability of the radiation barriers in long-stored SFA in respect to thermal treatment (important step of any dry technology) consists in modification of fresh fuel composition by special isotope admixing. In this connection, isotope ^{232}U should be distinguished because dry SNF reprocessing can't remove it. As it can be seen from Table 5, the radiation barrier formed by ^{232}U admixing into fresh fuel composition becomes less vulnerable to high-temperature SNF reprocessing.

5. Conclusions

Long-term SFA storing leads to sharp increase of FNM vulnerability in respect to any unauthorized actions. In this connection, a possibility was evaluated to enhance proliferation protection of SFA under their long-term storing. Using SFA from power VVER-1000 type reactor and research IRT-type reactor as typical examples, a possibility was analyzed for improving parameters of the radiation barriers generated by introduction of isotopic admixtures into fresh fuel compositions.

The results obtained in numerical studies allowed us to make the following conclusions about prolongation of the radiation barrier action in SFA from power VVER-1000 type reactors:

- Isotopes ^{232}U, ^{137}Cs, ^{152}Eu and ^{59}Co can be used for prolongation of the radiation barrier action at the level of the most stringent SFS_C criterion. However, only two first isotopes are able to provide significant (20 years and longer) prolongation.
- Isotopes ^{232}U and ^{137}Cs can be used for prolongation of the radiation barrier action at the level of the less stringent SFS_F criterion.

Minimal concentrations were determined for isotopic admixtures to be introduced into fresh fuel composition of IRT-3M fuel assembly for specified level and duration of the radiation barrier action. Only ^{232}U and ^{137}Cs can prolong the radiation barrier action up to 100 years at the level of SFS_F criterion. Creation of the higher radiation barrier, at the level of SFS_C criterion for 100-year cooling time of SFA IRT-3M seems quite difficult because of small mass and size (in comparison with SFA from power reactor). However, SFS_C criterion may be supported for 40 years by ^{232}U and ^{137}Cs admixing up to 6-8% HM.

So, it was revealed that ^{232}U and ^{137}Cs are the most promising isotopes for enhancing proliferation resistance of SFA from power and research reactors.. However, it should be noted that the radiation barrier produced by ^{137}Cs admixing is very vulnerable in respect to thermal treatment because of cesium volatility. That is why ^{232}U should be regarded as a top-priority isotope for FNM proliferation protection. The main arguments in favor of ^{232}U are related with impossibility of ^{232}U extraction from uranium fuel and time behavior of high-energy gamma-radiation emitted by ^{232}U and its decay products.

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