APPLICATION OF ADS FOR CREATION OF LONG-TERM RADIATION BARRIER IN MOX-FUEL

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ABSTRACT

The problem of nuclear materials protection against uncontrolled proliferation is considered. There are different potential approaches to create the inaccessibility conditions for nuclear materials (NM) with respect to possible unauthorized actions.
Creation of inherent radiation barriers inside of highly attractive NM (plutonium of any grade, highly enriched uranium) could be substantial deterrent, especially under changes in the NM protection conditions.
Within the frames of the physical measures it is proposed the way consisting of two consecutive procedures:

- Admixture of ^{231}Pa to MOX-fuel prior to fabrication of sub-assemblies (S/A);
- Short-term irradiation of fresh fuel assemblies in blanket of accelerator-driven facility.

The problem is to find such an amount of the admixed protactinium isotope and to define such irradiation conditions so that rate of equivalent dose from the irradiated fuel assemblies would not be below the specified protection level.
It is shown the possibility to produce the proliferation resistant MOX-fuel with inherent radiation barrier of long-term continuous action at different protection levels.

1. INTRODUCTION

At present time, one of major aspects in assurance of nuclear weapons non-proliferation is a creation of inaccessibility conditions to nuclear materials (NM) with respect to any unauthorized actions. In the first turn, it concerns NM of high attractiveness degree (plutonium-containing NM, for instance) suitable for manufacturing of nuclear explosive devices.

NM inaccessibilty can be reached by:

- Traditional measures of special NM management including NM physical protection, control and accounting.
- Physical measures imparting a self-protection property to NM; for example, introduction of ionizing radiation sources into NM composition (inherent radiation barrier).

The problem of NM vulnerability under international NM transactions is considered in the «Convention on the Physical Protection of Nuclear Material» [1] which was opened for

signature on March 3, 1980. By February 1997, 57 States had brought the Convention into force. Unfortunately, some developing countries did not join the Convention. Under these circumstances, the inherent radiation barriers are able to be an additional deterrent factor which seem reasonable to apply for non-proliferation assurance in international transfers of highly-attractive NM (fresh MOX-fuel assemblies, for instance) to the suspect world regions.

The purpose of the present work is to study a possibility to create the inherent radiation barrier in MOX-fuel by introducing isotope ^{231}Pa into fuel composition followed by short-term irradiation in an accelerator-driven system (ADS). As a result of this two-step procedure, MOX-fuel will be produced with property of long-term radiation self-protection against unauthorized actions. In the paper, mathematical model is described which was used for evaluating the procedure parameters (amount of admixed ^{231}Pa and exposure time in ADS) and associated parameters of inherent radiation barrier (rate of equivalent dose, duration of the radiation barrier action).

2. INHERENT RADIATION BARRIER

The inherent radiation barrier may be characterized with two main parameters:

- The protection level L. For the radiation barriers the protection level may be evaluated in terms of the rate of equivalent dose (RED, rem/h).
- The duration T for continuous action of radiation barrier at the protection level above L value.

Three protection levels were considered for fresh MOX-fuel:

- LD-protection, i.e. appropriate RED results in the lethal dose for one-minute exposure at 30 cm-distance from MOX-fuel assembly [2];
- SFS-protection, i.e. protection at level of the «Spent Fuel Standard» [3];
- INM-protection, i.e. protection at level of the irradiated NM [4].

3. WAYS TO CREATE THE INHERENT PROTECTION BARRIER

There are many ways to create the inherent radiation barrier in NM, neutron irradiation, for instance. Under neutron irradiation of fissionable NM, fission products (FP) are accumulated inside causing induced gamma-radioactivity. Another way is an admixture of radionuclides to NM composition. These approaches are able to create the inherent radiation barriers in NM with wide range of the protection level.

Each way has his own advantages and shortcomings. For example, FP gamma-activity rapidly decreases. It means the creation of long-term protection barrier requires multiply excessive FP gamma-activity at initial moment. Direct admixture of radionuclides-sources of gamma-radiation (^{106}Ru, ^{144}Ce, ^{60}Co [2]) allows to create the radiation barriers with any specified protection level and duration of action. However, in this case NM fabrication will require additional radiation protection of the personnel involved. Also, a possible way is an admixture to NM of isotopes - neutron predecessors for gamma-emitters, i.e. the isotopes which can be transformed into intense sources of gamma-radiation through neutron interactions.

Isotope ^{232}U takes a particular place amongst the isotopes-sources of gamma-radiation. High-energy gamma-radiation of this isotope is caused by ^{232}U decay products, mainly by ^{208}Tl. So, ^{232}U activity at first increases, then the equilibrium state is reached between ^{232}U and its decay products (maximum of ^{232}U activity). Further, ^{232}U activity decreases with decay half-time of ^{232}U (69 years). As a result, admixture of ^{232}U ensures the prolonged duration of NM protection action but provides reduced value of the protection level at initial period («vulnerability window»).

Thus, the long-term NM protection barriers without «vulnerability windows», without excessive initial activity and without substantial complication of NM fabrication procedures can be expediently created by applying the proper combination of admixing the especially selected isotopes to NM composition and neutron irradiation of NM.

4. APPLICATION OF ADS FOR CREATING THE RADIATION PROTECTION BARRIER IN MOX-FUEL ASSEMBLIES

The proposed approach [5] to creating the long-term radiation barrier in MOX-fuel includes two consecutive procedures:

- Admixture of radionuclides (^{232}U or its neutron predecessor ^{231}Pa) to MOX-fuel prior to fabrication of fuel rods and fuel assemblies.
- Short-term neutron irradiation of fresh MOX-fuel assemblies (MOX-FA) in ADS blanket.

These two procedures are able to form the long-lived (^{232}U) and short-lived (FP) components of induced MOX-fuel gamma-activity.

Isotope ^{231}Pa is selected as an isotope to be admixed because ^{231}Pa is preferable one as compared with ^{232}U because the former produces no real increase of a gamma-radiation background at step of MOX-fuel fabrication and transforms into ^{232}U under short-term neutron irradiation. It seems reasonable to introduce ^{231}Pa into MOX-fuel composition at large production plants under stringent international control.

Production of isotope ^{231}Pa is a separate issue, outside of the paper's frames. In principle, isotope ^{231}Pa can be produced in fast neutron spectrum through $^{232}Th(n,2n)$ reaction or in thermal neutron spectrum through $^{230}Th(n,\gamma)$ reaction.

Direct admixture of ^{232}U is possible too but ^{232}U content in nuclear fuel is strictly limited (~ 0.01% HM) by the regulatory constraints on radiation background at the operating MOX-fuel fabrication plants.

Selection of ADS for short-term irradiation of MOX-FA was caused by, at first, the circumstance that such a facility distinguishes with the upgraded nuclear safety since here the reactivity-induced accidents are excluded at deterministic level. Secondly, light-water pool-type ADS where MOX-FA to be irradiated are allocated around a solid target is a well-suitable facility for short-term irradiations and for practically continiuous reloading of fuel assemblies.

5. PROBLEM DEFINITION

The problem is to determine amount of ^{231}Pa to be admixed and exposure time Δt_{ir}^{min} of MOX-FA in ADS blanket needed for

$$\dot{D}(t) \geq L, \qquad t \in [0, T] \quad , \tag{1}$$

where $\dot{D}(t)$ – RED; L - the required protection level of MOX-fuel; T - duration of the inherent radiation barrier action.

The evaluations presented in [5] demonstrate that the main contributors to RED are the fuel rods allocated in peripheral layer of the irradiated fuel assembly. Let's assume that these fuel rods contain isotope ^{231}Pa. Then, RED of the irradiated fuel assembly includes two components: gamma-radiation emitted by FP (short-term component) and gamma-radiation emitted by ^{232}U decay products (long-term component). So, RED can be written in the following form:

$$\dot{D}(t) = M_{FP}\, f_{FP}(t) + M_{U2}\, f_{U2}(t) \quad ,$$

where M_{FP}, M_{U2} - mass of FP and ^{232}U in MOX-fuel; f_{FP}, f_{U2} - specific RED per mass unit of FP and ^{232}U, respectively.

The time dependencies of both components and total RED are presented in Fig. 1. As it was expected, FP component of RED rapidly reduces with time while just different behavior is observed for ^{232}U component: increase at initial stage, maximal point approximately after 10 years and rather slow decrease (with half-life time of ^{232}U). The time dependence of total RED has two minimal points. The right minimal point is the end of the protection period T while the left minimal point t_m is an internal point whose position is defined by the mass ratio M_{U2}/M_{FP} . So, if condition (1) is satisfied in the points t_m and T, then this condition is correct for all the points belonging to the range [0, T].

6. EQUATION SET FOR THE PROBLEM (1)

RED value depends on exposure time of MOX-fuel in ADS blanket and on fuel cooling time after irradiation. The two-component representation of RED value with respect to these variables can be written in the following form:

$$\dot{D}(t, \Delta t_{ir}) = M_{FP}(\Delta t_{ir}) \{f_{FP}(t, \Delta t_{ir}) + [M_{U2}(\Delta t_{ir})/M_{FP}(\Delta t_{ir})]\, f_{U2}(t, \Delta t_{ir})\}, \tag{2}$$

The time point t_m corresponding to internal minimum of RED dependence can be determined from classical condition:

$$\left.\frac{\partial \dot{D}}{\partial t}\right|_{tm} = 0 \quad ,$$

or, in more detailed form:

$$\left.\frac{\partial f_{FP}}{\partial t}\right|_{tm} = -\frac{(\sigma_c^{231}\cdot\Phi)\cdot\rho^{231}\cdot m_{U2}}{(\Sigma_f\cdot\Phi)\cdot m_{Pu}}\cdot\left.\frac{\partial f_{U2}}{\partial t}\right|_{tm} \quad , \qquad (3)$$

where σ_c^{231} - micro cross-section of neutron capture by isotope ^{231}Pa; ρ^{231} - concentration of ^{231}Pa in fuel; Φ - neutron flux; m_{Pu}, m_{U2} - mass of Pu and ^{232}U, respectively.

Exposure time to reach the protection level L in the left minimal point t_m can be determined from the equality $\dot{D}(t_m) = L$:

$$\Delta t_{ir} = \frac{L}{V\cdot[(\Sigma_f\cdot\Phi)\cdot m_{Pu}\cdot f_{FP}(t_m)+(\sigma_c^{231}\cdot\Phi)\cdot\rho^{231}\cdot m_{U2}\cdot f_{U2}(t_m)]} \quad , \qquad (4)$$

where V - volume of fuel.

The necessary amount of isotope ^{231}Pa to be admixed is defined by RED value at the right minimal point $\dot{D}(T, \Delta t_{ir}) = L$:

$$\rho^{231} = \frac{L - V\cdot\Delta t_{ir}\cdot(\Sigma_f\cdot\Phi)\cdot m_{Pu}\cdot f_{FP}(T)}{V\cdot\Delta t_{ir}\cdot(\sigma_c^{231}\cdot\Phi)\cdot m_{U2}\cdot f_{U2}(T)} \quad . \qquad (5)$$

The unknown values in the equations (3)-(5) are the exposure time Δt_{ir}, initial ^{231}Pa concentration ρ^{231} and neutron flux Φ. Neutron flux $\Phi(r,E,\Omega)$ is a solution of neutron transport equation in elementary cell of ADS blanket. This equation can be written in the following operator form:

$$\hat{P}(\rho^{231})\cdot\Phi = \frac{1}{k_\infty}\cdot\hat{Q}(\rho^{231})\cdot\Phi \quad , \qquad (6)$$

where $\hat{P}$ – operator describing neutron transport, absorption and scattering; $\hat{Q}$ - operator describing generation of secondary neutrons..

The equations set (2)-(6) defines the main conditions to form the inherent radiation barrier. These equations are non-linear and mutually linked. Solution of the set can be derived by consecutive iterations taking, for example, solution of the unlinked set (4)-(6) as an initial approximation:

$$\rho_0^{231} = 0$$

$$\hat{P}\cdot\Phi_0 = \frac{1}{k_\infty}\cdot\hat{Q}\cdot\Phi_0$$

$$\Delta t_{ir}^{(1)} = \frac{L}{V\cdot(\Sigma_f\cdot\Phi)\cdot m_{Pu}\cdot f_{FP}(t_m)} \quad .$$

7. AN EXAMPLE FOR DETERMINATION OF ^{231}Pa CONTENT AND EXPOSURE TIME OF MOX-FA IN ADS TO REACH 50-YEAR ACTION OF THE INHERENT RADIATION BARRIER

An example for solving the equations set (2)-(6) for T = 50 years and for three protection levels mentioned above is presented in Fig. 2. It is considered MOX-fuel of VVER-1000 reactor. It can be seen that, for every protection level, RED value is equal to L in two time moments while RED value exceeds the required protection level in all other points of time period T.

Position of the right minimal point T defines ^{231}Pa content in MOX-fuel. If the protection level L increases, position of the left minimal point shifts towards the shorter time values because it is necessary to increase ^{231}Pa content in fuel.

The results presented in Fig. 3 demonstrate the time to receive the lethal dose being in direct proximity (30 cm) to the fuel assembly protected at SFS-level is not longer than 5-6 minutes. The results obtained in solving the equations set (2)-(6) are presented in Table I.

Table I. Results of solving the equations set (2)-(6) for different protection levels and for 50-year action of the inherent radiation barrier

Protection level	LD	SFS	INM
Δt_{ir}^{min}, days	25	6.5	2
^{231}Pa content in MOX-fuel, %	25	14	6.5
t_m, days	630	800	1200

It may be concluded from the data shown in Table I the time required for irradiation of MOX-FA in ADS blanket is rather short. Even for LD-protection level the exposure time Δt_{ir} is not longer than one month, for SFS-protection level - one week. Significant content of ^{231}Pa in MOX-fuel is required to ensure LD-protection level and 50-year period of the inherent radiation barrier action. This content can be reduced by increasing Δt_{ir} and decreasing T.

Dependence of ^{231}Pa content on exposure time of MOX-fuel in ADS blanket is demonstrated in Fig. 4. It can be seen the required ^{231}Pa content can be substantially reduced by decreasing the exposure time.

After irradiation in ADS blanket, the protected MOX-FA must be cooled for some time to reduce residual heat generation and intensity of gamma-radiation down to the level acceptable for transportation in standard spent nuclear fuel (SNF) casks. As known [6], the standard casks used for SNF transportation from power reactors of VVER-1000 type are characterized with acceptable value of specific residual heat generation about 23 W/kg after a half-year cooling time. Evidently, intensity of gamma-radiation emitted by MOX-FA irradiated in ADS blanket must not exceed intensity of gamma-radiation emitted by SNF after a half-year cooling time. The performed estimations have demonstrated the necessary cooling time of MOX-FA irradiated in ADS blanket did not cause any substantial delay ($t_{cool} < 1$ month) for their transportation to nuclear power plants in standard SNF casks. So, the upgraded radioactivity of the protected MOX-fuel will not exert negative influence on the environment during MOX-FA transportation in standard SNF casks.

CONCLUSIONS

The proposed approach to creation of the long-term inherent radiation barrier of MOX-fuel may be characterized with the following features:

- Using such an approach, it is possible to produce the proliferation resistant MOX-fuel with inherent radiation barrier of long-term continuous action at different protection levels: from standard irradiated NM level to the level of receiving the lethal dose for one-minute period at 30-cm distance from the protected MOX-fuel assembly.
- Duration of the radiation barrier's continuous action may be varied within rather wide range. Duration of the barrier action is mainly defined by amount of admixed ^{231}Pa and time of its irradiation in ADS blanket.
- For the analysed range of the protection levels and times of the radiation barrier action, time of MOX-FA exposure in ADS blanket is no longer than one month. So, we can say about really short-term irradiation. The latter circumstance leads to two important consequences. Firstly, energy potential of MOX-fuel undergoes no any significant changes during neutron irradiation in ADS blanket. Secondly, it is possible to reach high throughput of ADS in production of MOX-FA protected against uncontrolled proliferation.

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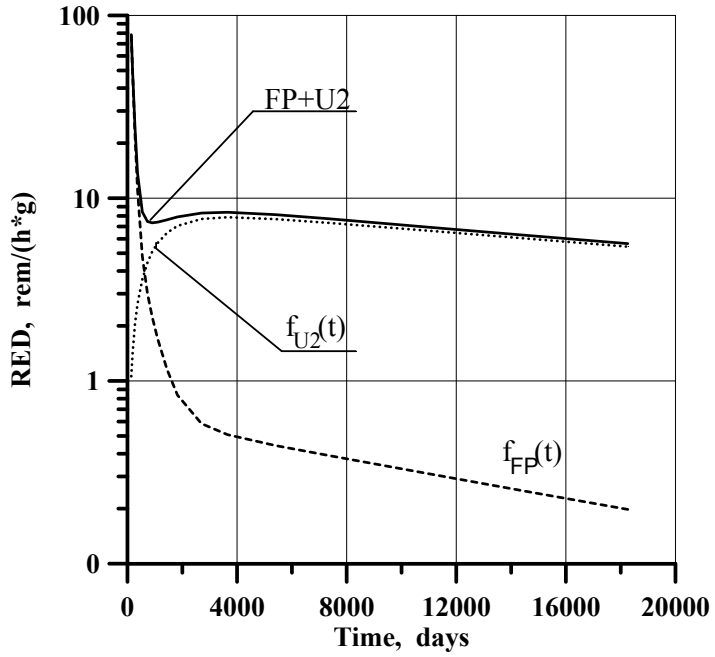

Figure 1. Time dependence of RED from FP and 232U accumulated in MOX-fuel and total RED for $M_{U2}/M_{FP} = 1$.

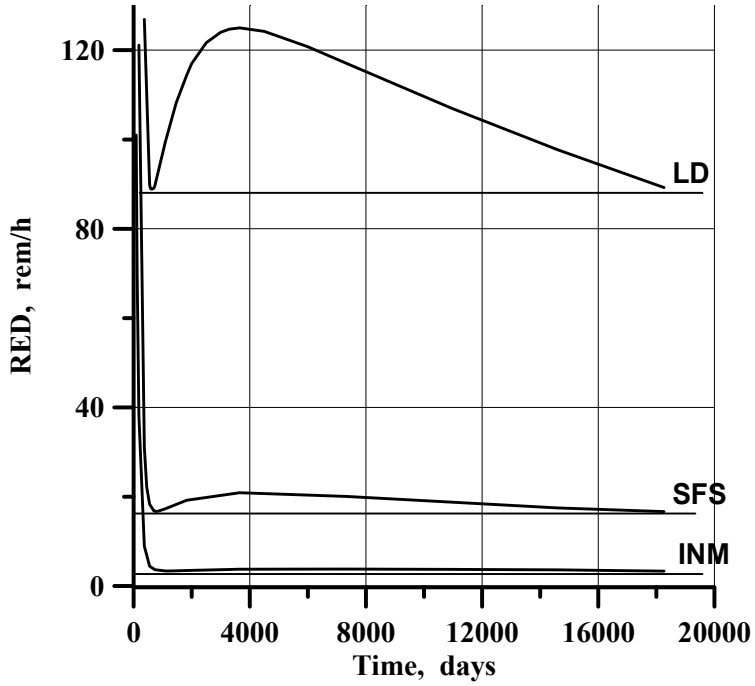

Figure 2. RED at 30 cm distance from MOX-fuel rod of VVER-1000 irradiated in ADS.

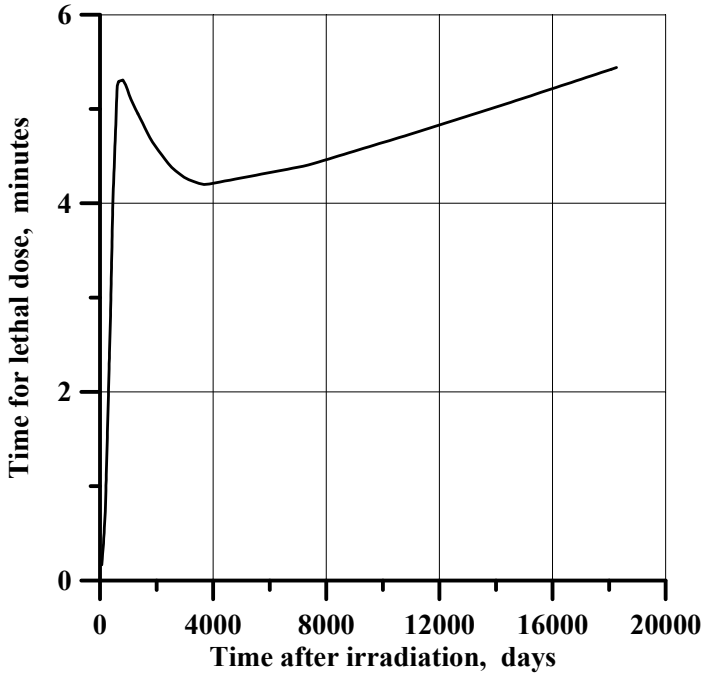

Figure 3. Time for receiving the lethal dose at 30 cm distance from MOX-FA

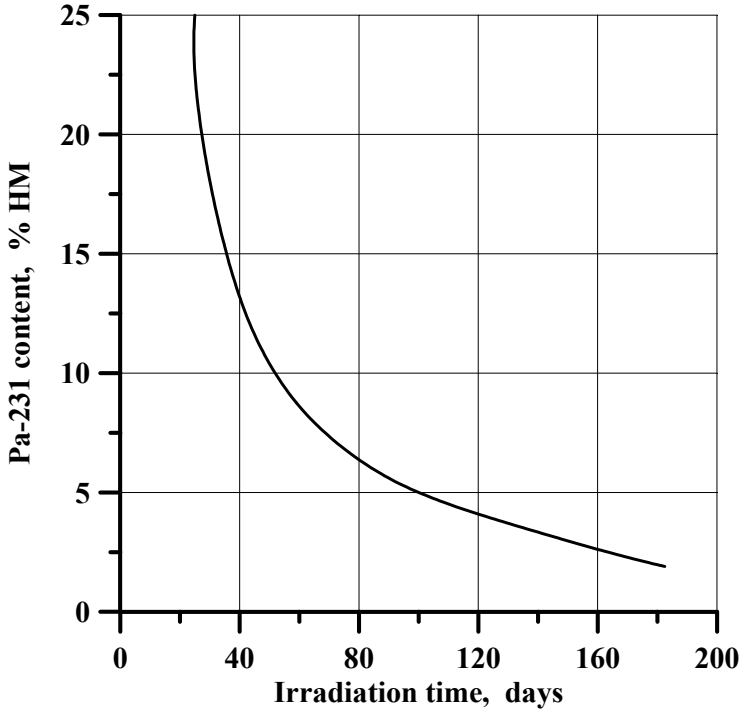

Figure 4. Required ^{231}Pa content in MOX-fuel to provide LD-protection level during 50-year period after irradiation in ADS blanket.