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A Study on High-Intensity Radiation Protection of MOX-Fuel Doped with Protactinium

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The paper addresses the problem of MOX-fuel proliferation protection. The analysed measures are aimed at forming the enhanced protective radiation barrier inside of MOX-fuel during full cycle of its management. These measures include an admixture of (Pa-U)-getter into MOX-fuel composition. It is demonstrated that, under irradiation of MOX-fuel, a long-term radiation barrier is formed in the fuel. In case of the uranium recycle, the barrier is transferred naturally to fresh MOX-fuel. It is shown that, under long-term storage of spent fuel, introduction of (Pa-U) getter makes it possible to give a proliferation self-protection property to MOX-fuel even against short-term unauthorized actions. The preventive measures are proposed to decrease gamma-activity of nuclear materials at fabrication stage of MOX-fuel assemblies.

KEYWORDS: proliferation, proliferation safety, radiation barrier, getter, lethal dose, protactinium, spent fuel standard, LWR, closed fuel cycle, thorium

1. Introduction

Development of nuclear power industry in order to satisfy the growing demands for energy must take into consideration a series of principal conditions including economical competitiveness, radiation and nuclear safety, inherent protection of nuclear materials (NM) and nuclear technologies in respect of their potential usage for creation of nuclear explosive devices (proliferation safety). Proliferation safety of NM and technologies involved into nuclear fuel cycle (NFC) must give a proper response to covert and obvious threats of nuclear weapon proliferation including abuses and errors in management of NM, associated equipment and technologies. Proliferation safety of NM and NFC technologies can be ensured by means of inherent self-protection properties which impede any unauthorized actions of potential adversaries.

An important component of MOX-fuel protection-in-depth system is constituted by the internal radiation barriers which can be created in the fuel under irradiation. The internal radiation barriers give an inherent self-protection property to a MOX-fuel, and they are continuously acting barriers. There are no simple methods to remove these barriers. So, inherent radiation barriers can serve as a tool for nuclear fuel proliferation safety assurance. Just that is why spent fuel in Russia is regarded as a nuclear material of the lowest attractiveness category independent on spent fuel amount. [1] In the USA the Spent Fuel Standard is adopted for characterization of NM self-protection degree below which a risk of unauthorized plutonium diversion is evaluated as significant. [2]

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However, fresh fuel is not self-protected NM, and so, fresh fuel is vulnerable to the unauthorized actions. Proliferation protection of fresh fuel requires the special measures to be undertaken for giving the self-protection property to fresh fuel assemblies.

The radiation barriers arising in nuclear fuel after its irradiation can be characterized by the following main parameters:

- minimal protection level L in terms of rate of equivalent dose (RED, rem/h; 1 Sv/s = 3.6 10^5 rem/h) at given distance from fuel assembly (FA);
- duration T of continuous action of the radiation barrier at the protection level no lower than L;
- time non-uniformity of NM protection during given duration T of the radiation barrier action.

Obviously, if we want to protect MOX-fuel, our interest is to have high protection level and long-term action because, in opposite case, it would be very simple to overcome this barrier - it is necessary to wait some time only, till radioactivity of MOX-fuel decreases to an acceptable level.

It is desirable that proliferation protection of MOX-fuel assemblies would be at the level high enough to withstand any unauthorized actions including short-term actions. In the latter case, proliferation protection must correspond to receiving the lethal dose (~500 rem) for time period of minute scale.

Unfortunately, short-term preliminary irradiation of MOX-fuel does not yield the desirable result. In this case, inherent radiation barrier is mainly defined by gamma-activity of fission products. It leads to the following consequences:

- on the one hand, MOX-fuel is reliably protected during the first 10 years of FA storage (time till receiving the lethal dose at 30 cm-distance from FA is no longer than 7 minutes);
- on the other hand, time dependence of RED is characterized by high non-uniformity with excessive protection in initial period and insufficient protection for a long-term FA storage. For example, if cooling time t_c of irradiated FA is prolonged from 10 years to 100 years, then time till receiving the lethal dose will be longer by an order of magnitude also. Thus, inherent radiation barrier of MOX-FA with long cooling time is not efficient countermeasure against short-term unauthorized actions.

The present paper is devoted to analysis of special measures which are able to give a self-protection property to MOX-fuel in form of enhanced radiation barrier with prolonged action. The main object of the consideration is a vibro-packed MOX-fuel containing protactinium as a getter: (U, Pu)O_2 + Pa. Such a fuel composition is characterized by self-protection property for full cycle of nuclear fuel management. Achievable levels of proliferation self-protection are studied for such kind of MOX-fuel at its usage in the operation cycle of light-water reactors.

2. Fabrication of inherently protected MOX-fuel

At present, it was demonstrated a feasibility for remote fabrication of the vibro-packed fuel in hot cells. [3] The vibro-packed fuel can be manufactured with effective density of ~ 9 g/cm^3. Simplicity and reliability of the technological process facilitate its automation and remote control. The latter circumstance confirms a feasibility to apply the vibro-packing technology for fabrication of fuel elements containing highly-active fuel meat.

In order to correct an oxygen potential of fuel, a getter is introduced into fuel composition. The getter applied now represents a metal uranium powder (diameter of particles is about 100 microns). The getter is uniformly distributed over volume of fuel. Principal result of the uranium getter introduction is an exclusion of the corrosion processes initiated by some fission products (cesium, iodine, tellurium). As a consequence, the limitation on fuel burn-up related with fuel-cladding interactions is practically removed. [3]

Introduction of protactinium into fuel composition leads to the following process: under neutron irradiation, ^{231}Pa (the only long-lived protactinium isotope) transforms into ^{232}U through the following chain:

$$^{231}\text{Pa}(n, \gamma)^{232}\text{Pa}(\beta^-, T_{1/2} = 1.31\ \text{d})^{232}\text{U}$$

^{232}U takes a particular place amongst isotopes-emitters of gamma-radiation. High-energy gamma-radiation of ^{232}U is caused by its decay products, mainly by isotope ^{208}Tl. As a result, generation of ^{232}U provides a long-term protective radiation barrier. In case of uranium recycle, the radiation barrier is naturally transferred to fresh MOX-fuel. [4]

Chemical properties of protactinium are similar to those of uranium. These elements are characterized by close valences: III, IV and V for protactinium and II, III, IV, V and VI for uranium. [5,6] In some publications it is noted that protactinium can form compounds with halogens, alkaline and alkaline-earth elements. As applied to the considered protactinium function as a getter (like metal uranium), these are the compounds which are able to bind cesium and iodine. As for protactinium compounds with tellurium, we have not found any publications about it.

If protactinium is introduced in fuel composition in form of small metal particles, then, under neutron irradiation, protactinium intensely transforms into ^{232}U, i.e. significant amount of the uranium getter (in form of uranium-protactinium alloy) is generated in fuel. Thus, presence of the uranium-protactinium getter in MOX-fuel can substantially suppress the corrosion processes caused by some fission products.

The circumstance mentioned above constitutes a necessary condition for reaching high fuel burn-up. In parallel, isotope ^{231}Pa, acting, in essence, like a burning absorber, reduces effectively an initial reactivity margin and promotes realization of prolonged MOX-fuel life-time.

The uranium-protactinium getter makes it possible to create long-term inherent radiation barrier in uranium oxide fuel and in MOX-fuel. The radiation barrier can be characterized as follows:

- the barrier acts on total uranium mass;
- removal of the barrier requires fine purification of plutonium from uranium and impurities because residual ^{232}U is able to initiate (α,n)-reactions with light impurities.

3. Campaign of MOX-fuel containing Pa-getter in light-water reactor. Evaluation of U-getter formation.

Here, it is considered a nuclear fuel cycle of light-water reactor (LWR) loaded with MOX-fuel containing weapons-grade plutonium. Pa-getter (5% HM) is introduced into fresh fuel composition. The reactor operates at fixed power level with average specific power generation rate of 110 kW/l.

Isotopic and radiation parameters of spent LWR fuel are calculated using the computer code package SCALE. [7] Rate of equivalent dose (RED) at 30 cm-distance from fuel assembly is regarded as a main parameter of the protective radiation barrier.

The evaluations demonstrate that introduction of 5% ^{231}Pa into MOX-fuel makes it possible to prolong fuel life-time by a factor of 2,5 and reach fuel burn-up at level of 10% HM.

Of course, transition to fuel with prolonged life-time can be realized only under conditions of nuclear safety assurance including negative values of the coolant temperature reactivity coefficient. For the considered MOX-Pa-fuel, increase of fuel burn-up to the limits mentioned above is accompanied by conservation of negative values for the coolant temperature reactivity coefficient.

Time dependence of uranium (^{232}U)-getter formation in fuel assembly of VVER-type LWR loaded with MOX-fuel is shown in Fig. 1. It is seen that, during the fuel campaign, amount of U-getter reaches significant fraction (~40%) of initial protactinium loading. Simultaneously, protactinium content becomes three times lower.

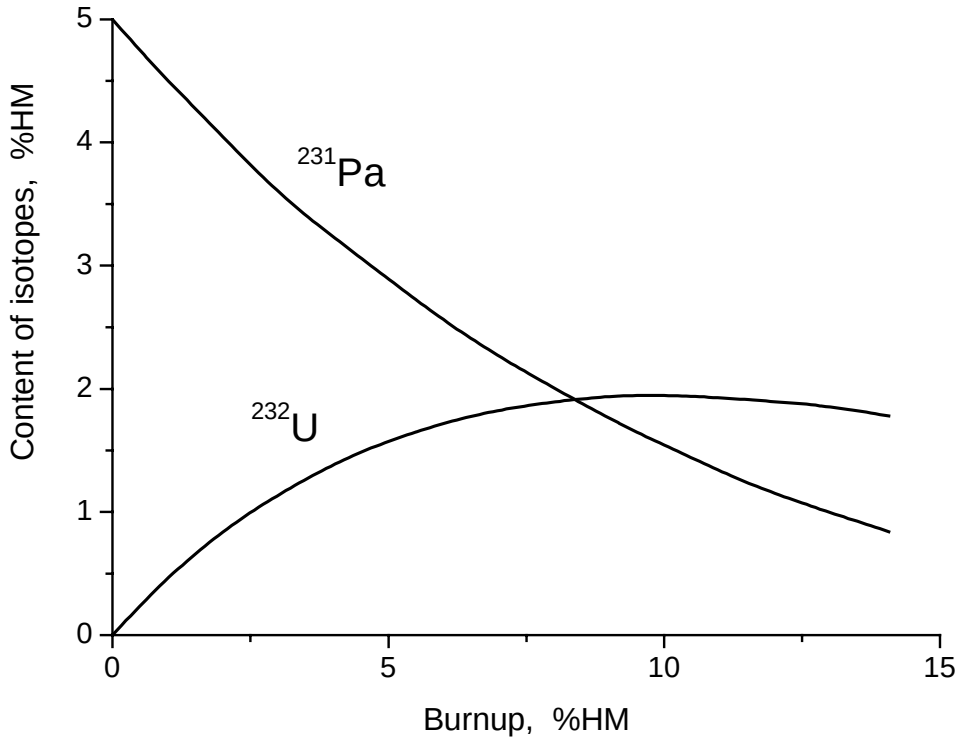

Fig. 1 Accumulation of U-getter in MOX-fuel assembly of LWR.

4. Evaluation of MOX-fuel proliferation protection in closed fuel cycle with prolonged fuel life-time

4.1. Proliferation protection of spent fuel

Many experts in non-proliferation of nuclear materials (NM) consider that spent fuel from power LWR is an unattractive NM in respect of unauthorized actions because of emitted radiation. However, high level of plutonium proliferation protection changes in several decades to insufficient level of proliferation protection in respect of short-term unauthorized actions. Huge amounts of spent fuel have been accumulated for previous decades of nuclear power system operation all over the world. At present, rate of spent fuel production is significantly higher than rate of spent fuel reprocessing. In this connection, an urgent problem arises on proliferation self-protection assurance of spent fuel stored for a long time. Introduction of Pa-getter into fresh MOX-fuel composition affects proliferation protection of spent MOX-fuel. Let's consider the effects induced by Pa-getter.

From proliferation protection standpoint for spent fuel to be stored, it is desirable to prolong fuel life-time in the reactor by such a way that, at the end of irradiation cycle, content of isotope ^{232}U (the

dominant contributor to long-term component of the protective radiation barrier) reaches a maximal value. It is demonstrated in Fig. 1 that prolongation of U-Pu-Pa-fuel life-time till burn-up about 10% HM allows to accumulate maximal (under adopted conditions) content of ^{232}U – about 2% HM. Proliferation protection of fuel assemblies can be expressed in terms of time till receiving the lethal dose at unauthorized actions. Figure 2 shows how proliferation protection of MOX-fuel containing Pa-getter varies after discharge from the reactor in comparison with proliferation protection of MOX-fuel without Pa-getter. Under long-term storage conditions (T_{cool} = 100 years), time till receiving the lethal dose in vicinity of fuel assembly containing the getter is no longer than 1,3 minutes and shortens by a factor of ~ 20 in comparison with fuel assembly of the same burnup and by a factor of ~ 55 in comparison with fuel assembly of the 4% HM burnup. Thus, during 100 years of spent MOX-fuel storing, MOX-fuel is self-protected even against short-term (the minute scale) unauthorized actions. More over, incline of the curves shows that in next centennial period ($100 < t_{cool} < 200$) fuel assemblies with Pa-getter will gainfully differ from others due to keeping the self-protection property.

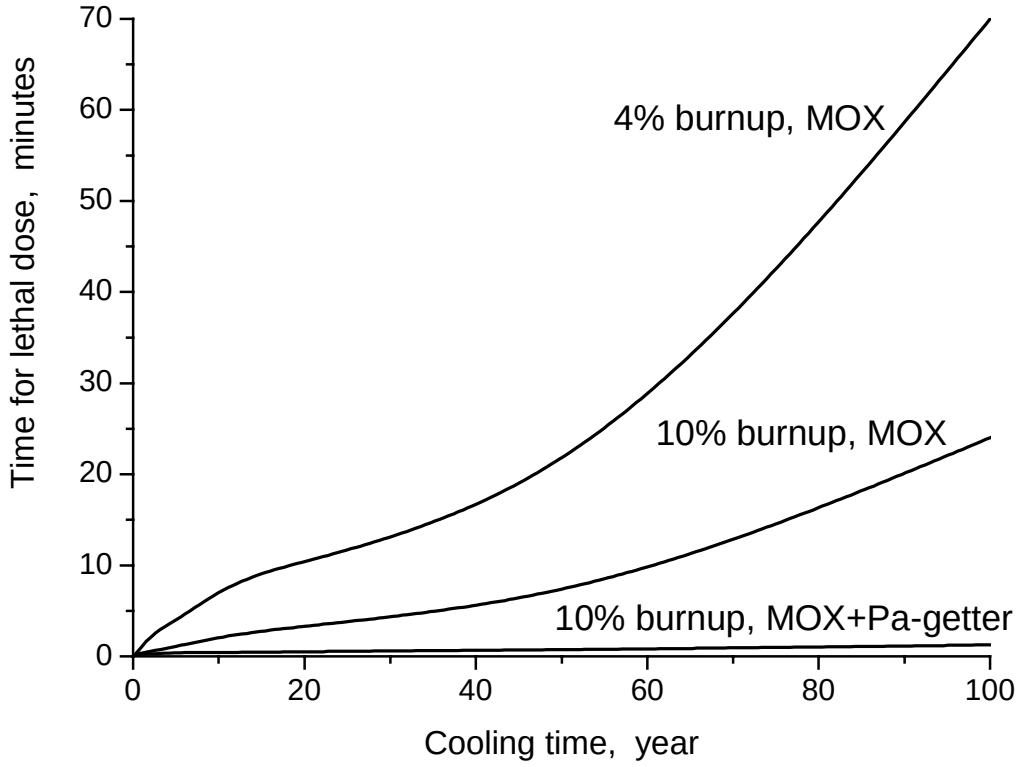

Fig. 2 Change in proliferation protection of spent fuel assembly caused by introduction of Pa-getter.

4.2 Evaluation of MOX-fuel proliferation protection after radiochemical reprocessing

Importance of Pa-getter consists not only in formation of long-term protective radiation barrier of spent fuel but also in natural transfer of the barrier to fresh MOX-fuel under conditions of uranium recycle. In this case, the vibro-packing technology applied for fuel rods fabrication from granular fuel allows to arrange an automatic and remotely controlled process.

There are two possible options for closed nuclear fuel cycle of LWR:

(1) After cooling (5 years) spent fuel is reprocessed by radiochemical way with extraction of fission products and minor actinides.
(2) At radiochemical reprocessing of spent fuel, in addition to fission products and minor actinides, thorium fraction is extracted from the fuel also.

Removal of fission products and minor actinides in the first option leads to approximately two-fold decrease of MOX-fuel proliferation protection. However, proliferation protection of MOX-fuel assemblies remains at the level high enough to withstand short-term unauthorized actions (see Fig. 3). Specific feature of the first option is the fact that MOX-fuel assemblies, just after fabrication, have a high level of proliferation protection, and they may be transported to nuclear power plants.

It should be noted that, in period of fuel assemblies fabrication, gamma-activity of MOX-fuel is rather high, and handling with this material requires application of reliable radiation shielding for the personnel involved. In this connection, it is desirable to have a «window» of reduced activity for the fabrication period. Removal of thorium fraction from the reprocessed fuel serves to this purpose. Figure 3 shows that removal of ^{228}Th, together with fission products and minor actinides, in spent fuel reprocessing leads to multiple reduction of fresh fuel activity.

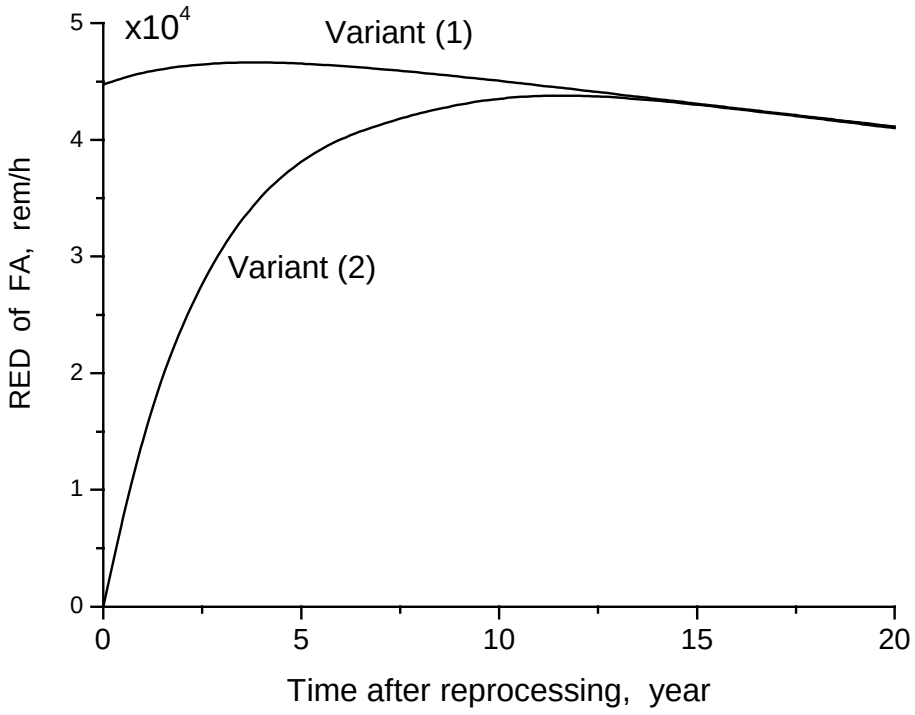

Fig. 3 Time dependence of fuel assembly (FA) gamma-activity after radiochemical reprocessing.

Due to radioactive decay of ^{232}U, the «window» of reduced activity becomes gradually «close». Rate of activity growth is defined by ^{232}U content. The activity growth for 2% HM content of ^{232}U in fuel is demonstrated in Fig. 4. During the first month after radiochemical reprocessing of fuel with thorium extraction, activity of MOX-fuel does not exceed (2÷3)% of maximal value. However, despite of sufficiently long time interval for handling with relatively low active MOX-fuel, it seems reasonable to unite the stages of spent fuel reprocessing and fresh MOX-fuel fabrication at one site or at two near located production sites.

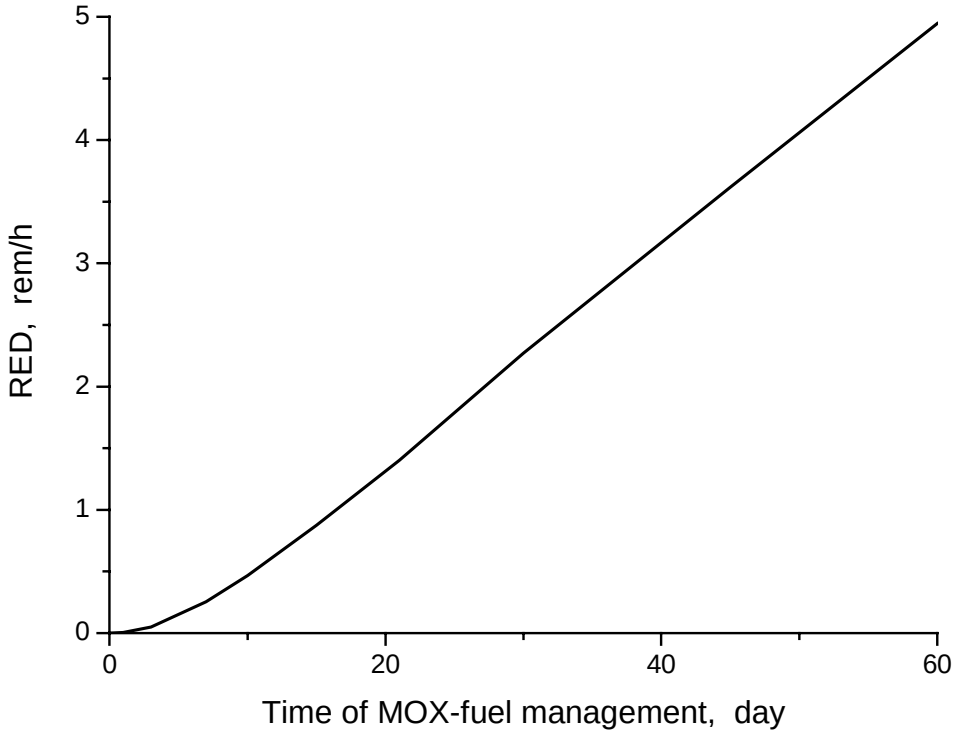

Fig. 4 Growth of gamma-activity after separation of fission products, minor actinides and thorium (per 1 kg of MOX-fuel). The RED values are evaluated at 30-cm distance from MOX-fuel.

In the second option of spent fuel reprocessing, it is important to know how long time does it take to restore the protective radiation barrier of fresh MOX-fuel at the level high enough to counteract successfully against short-term unauthorized actions, i.e. when MOX-fuel assemblies could be sent to a customer. Proliferation protection of MOX-fuel assembly as a function of storage time before its transportation to a nuclear power plant is presented in Fig. 5 for fuel with extracted thorium fraction. Two-months storage of MOX-fuel assembly ensures restoration of the protective radiation barrier at the level corresponding to short time ($\leq$ 10 minutes) till receiving the lethal dose.

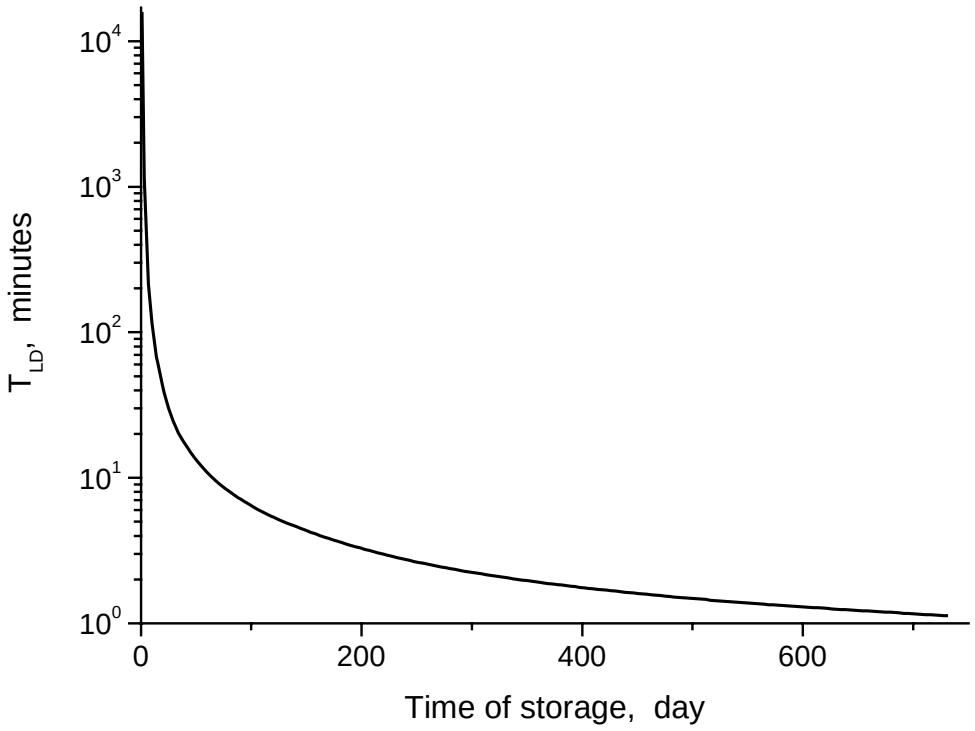

Fig. 5 Time dependence of MOX-fuel assembly proliferation protection during its storage before transportation to nuclear power plant. T_{LD} – time till receiving the lethal dose in vicinity (30 cm) of fuel assembly.

5. Conclusion

Analysis of the ways for ensuring the proliferation self-protection of MOX-fuel by introduction of Pa-getter and comparison of this way with other options for MOX-fuel proliferation protection against unauthorized actions allow us to make the following conclusions:

1. An expediency for application of protactinium as a getter material is defined by the following properties of protactinium:

- Neutron irradiation converts protactinium into isotope ^{232}U, long-lived source of high-energy gamma-radiation.
- If protactinium is introduced into MOX-fuel in form of small metal particles, then, under neutron irradiation, uranium getter is produced in the fuel in form of U-Pa alloy.
- Being, in essence, a burnable absorber, isotope ^{231}Pa is able to decrease effectively an initial reactivity margin and, thus, to promote realization of prolonged MOX-fuel life-times.

2. The performed evaluations show that accumulation of uranium getter constitutes significant fraction (~40%) of initial protactinium loading. Formation of U-Pa getter containing significant uranium fraction is an important condition for reaching high fuel burn-up. Naturally, the higher fuel burn-up, the higher proliferation protection of spent and reprocessed fuel.

3. Under conditions of the increasing terrorist activity all over the world, the proliferation protection standards used now for spent fuel stored for a long time need to be corrected. Proliferation protection of spent fuel can be significantly enhanced by introducing protactinium getter into fresh fuel composition. Additional effect of Pa-getter is a possibility to increase fuel burn-up to 10% HM and above.
4. Removal of thorium together with fission products in spent fuel radiochemical reprocessing allows to decrease by many times radioactivity of fuel and fabricate fresh fuel under more comfortable radiation conditions.
5. At present, the vibro-packed fuel fabrication technology is mastered in practice. Application of this technology really resolves the problem of handling with MOX-fuel with inherent protective radiation barrier.
6. Within the frames of the considered direction, application of combined U-Pa getter and assessment of its effectiveness require a separate investigation.

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