RADIATION PROTECTION POTENTIAL OF MOX-FUEL DOPED WITH 231Pa AND Cs RADIOISOTOPES
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The paper addresses the problem of MOX-fuel self-protection during full cycle of MOX-fuel management. Under conditions of the closed LWR cycle the proliferation-resistance levels were evaluated for fresh and spent MOX-fuel with 231Pa and Cs feed. As it follows from the paper results, combination of these two admixtures being doped into MOX-fuel is able to enhance the inherent radiation barrier and to weaken shortcomings of both proliferation deterrents.

INTRODUCTION
Creation of inaccessibility conditions for any unauthorised action of MOX-fuel is one of the most important aspects for wide implementation of the closed uranium–plutonium fuel cycle into nuclear power system. So, the measures, which are able to give a self-protection property to MOX-fuel, represent an important factor in the development of protection-in-depth system for MOX-fuel.

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. That is why in Russia spent fuel is regarded as a nuclear material (NM) of the lowest category of attractiveness independent of the amount of fuel spent(1). In the USA, the Spent Fuel Standard is adopted for characterisation of NM self-protection degree, below which a risk of unauthorised plutonium diversion is evaluated as being significant(2).

However, fresh fuel is not self-protected NM, and so, fresh fuel is vulnerable to unauthorised actions. The present paper is devoted to analysis of special measures which are able to give a self-protection property to MOX-fuel in the form of enhanced radiation barrier with prolonged action.

FABRICATION OF INHERENTLY PROTECTED MOX-FUEL
The object of consideration is a vibro-packed MOX-fuel containing the metal protactinium as a getter(3). Such a fuel composition is characterised by self-protection property for the full cycle of nuclear fuel management. Achievable levels of proliferation self-protection are studied for this type of MOX-fuel in its usage in the operation cycle of light-water reactors.

At present, the feasibility of the vibro-packed fuel in hot cells for remote fabrication was demonstrated(3). The vibro-packed fuel can be manufactured with an effective density of ~9 g cm⁻³. Simplicity and reliability of the technological process facilitate its automation and remote control. The latter circumstance confirms the feasibility of applying the vibro-packing technology for the fabrication of fuel elements containing highly active fuel meat.

In order to correct the oxygen potential of a fuel, a getter is introduced into the fuel composition. The getter applied here is a metal uranium powder (diameter of particles is ~100 μm). The getter is uniformly distributed over the volume of fuel. The principal effect of introducing the uranium getter is an exclusion of the corrosion processes initiated by some fission products (cesium, iodine and tellurium). As a consequence, the limitation on fuel burn-up, related to fuel-cladding interactions, is practically removed(3).

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

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

232U takes a special place among isotope-emitters of gamma radiation. High-energy gamma radiation of 232U is caused by its decay products, mainly by the isotope 208Tl. As a result, generation of 232U provides a long-term protective radiation barrier. During uranium recycle, the radiation barrier is naturally transferred to fresh MOX-fuel.

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

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

CAMPAIGN OF MOX-FUEL CONTAINING Pa-GETTER IN LIGHT-WATER REACTOR—EVALUATION OF U-GETTER FORMATION

Here, we consider the nuclear fuel cycle (NFC) of a light-water reactor (LWR) loaded with MOX-fuel containing weapon-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$^{-1}$.

The isotopic and radiation parameters of spent LWR fuel are calculated using the computer code package SCALE$^{(4)}$. The rate of equivalent dose (RED) at a distance of 30 cm from the fuel assembly (FA) 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 lifetime by a factor of 2.5 and reach fuel burn-up at level of 10% HM. During this fuel campaign, the amount of U-getter reaches a significant fraction (~40%) of the initial protactinium loading. Simultaneously, the protactinium content becomes three times lower.

EVALUATION OF MOX-FUEL PROLIFERATION PROTECTION IN A CLOSED FUEL CYCLE WITH PROLONGED FUEL LIFETIME

Proliferation protection of spent fuel

At present, the rate of spent fuel production is significantly higher than the rate of spent fuel reprocessing. In this connection, an urgent problem arises on proliferation self-protection assurance of spent fuel that has been stored for a long time. Figure 1 shows how proliferation protection of MOX-fuel containing a Pa-getter varies after discharge from the reactor in comparison with proliferation protection of MOX-fuel without a Pa-getter. Under long-term storage conditions ($T_{cool} = 100$ y), the time taken to receive the lethal dose in the vicinity of an FA containing the getter is no longer than 1.3 min; this shortens by a factor of ~20 for FA of the same burn-up, and by a factor of ~55 for FA of the 4% HM burn-up. Thus, during 100 y of spent MOX-fuel storing, MOX-fuel is self-protected even against short-term (the minute scale) unauthorised actions. Moreover, the incline of the curves shows that in the next centennial period ($100 < t_{cool} < 200$) FAs with Pa-getter will gainfully differ from others as they keep their self-protection property.

Evaluation of MOX-fuel proliferation protection after radiochemical reprocessing

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

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

1. After cooling (5 y), the spent fuel is reprocessed in the radiochemical way with the extraction of fission products (FPs) and minor actinides. Removal of FPs and minor actinides in the first option leads to ~2-fold decrease of MOX-fuel proliferation protection. However, proliferation protection of MOX-FAs remains at a level that is high enough to withstand short-term unauthorised action. A specific feature of the first option is the fact that MOX-FAs, 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 the period of FA fabrication, gamma-activity of MOX-fuel is rather high and handling 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 together with FP and minor actinides, in spent fuel reprocessing leads to multiple reduction of fresh fuel activity. In the first month of the radiochemical reprocessing of fuel with thorium extraction the activity of MOX-fuel does not exceed 2-3% of its maximal value.

Owing to radioactive decay of $^{232}\text{U}$, the “window” of reduced activity (in the second option) becomes gradually “close”. In this connection it is important to know how much time it takes to restore the protective radiation barrier of fresh MOX-fuel at a level that is high enough to successfully counteract the short-term unauthorised action, i.e. when the MOX-FA could be sent to a customer. Storage of MOX-FA for two months ensures restoration of the protective radiation barrier at the level corresponding to the short time (≤10 min), upto receiving the lethal dose.

APPLIED USE OF MOX-FUEL

Among gamma-active radionuclides, we considered mixtures of those nuclides which are FP or products of their transformations under neutron irradiation. Isotope $^{137}\text{Cs}$ can be regarded, in the first turn, as the mostly long-lived gamma-active FP. Let us analyse a possible way to arrange the proliferation protection of nuclear fuel using $^{137}\text{Cs}$, and compare the proliferation protection with that obtained by the addition of $^{231}\text{Pa}$.

Isotope $^{137}\text{Cs}$ is characterised by the following specific features: relatively long half-life ($T_{1/2} = 30\text{ y}$) and, at the same time, low-neutron-capture cross section ($\sigma_c \approx 0.02$ barn in LWR spectrum). So, theoretical value of equilibrium $^{137}\text{Cs}$ content is >10 times higher than that spent nuclear fuel (SNF) of typical LWR (0.23% HM) with fuel burn-up of 4% HM. The proliferation protection of $^{137}\text{Cs}$-containing FA with $^{137}\text{Cs}$ contents close to equilibrium value significantly exceeds the proliferation protection criterion of the lethal dose receiving for a minute. Such a feature leads to the fact that, under multiple fuel recycle, only partial extraction of $^{137}\text{Cs}$ from SNF would be sufficient.

However, it should be noted that caesium oxide is a less stable material than uranium and plutonium dioxides. In an area of columnar grains, caesium can exist in the form of separate atoms. The boiling temperature of caesium (678°C) is significantly lower than the working temperatures in power LWR. So, caesium can exist in a vapour phase and migrate in oxide fuel, thus causing the enhanced fuel-cladding interactions. Therefore, in the present paper, caesium content twice as much as that in traditional LWR-SNF is adopted as an upper limit. Let us assess, under such a limitation, the proliferation protection of the reprocessed fuel in the most vulnerable NFC stages (fabrication and transportation to nuclear power plant) for MOX-fuel management regime described above.

Caesium content in fuel after the second campaign corresponds to the limiting value. So, for the next irradiation cycles, excess caesium (~36%) should be removed. The proliferation protection of fresh FA containing fuel for the second campaign corresponds to RED $= 8650$ rem per h at a distance of 30 cm from the FA, i.e. the time up to receiving the lethal dose is ~3 min in the vicinity of the FA. Multiple repetition of irradiation cycles with the removal of excess caesium leads to equilibrium isotopic composition of the caesium fraction. In contrast to another radioactive FP (Ce, Ru, etc.), the share of $^{137}\text{Cs}$ in the isotopic composition is kept at a significant level (~30%) under multiple recycling.

Despite the low share of $^{134}\text{Cs}$ in caesium isotopic compositions (0.57%) and its short half-life ($T_{1/2} = 2.06$ y), $^{134}\text{Cs}$ gives a substantial contribution to the proliferation protection level (~40%). In contrast to $^{137}\text{Cs}$ and $^{134}\text{Cs}$, the isotopes $^{133}\text{Cs}$ and $^{135}\text{Cs}$ give no practical contribution to the RED.
value because $^{133}\text{Cs}$ is a stable isotope while $^{135}\text{Cs}$ has a very long half-life ($T_{1/2} = 2.3 \times 10^6$ y).

CONCLUSION

Analysis of the ways of ensuring the proliferation self-protection of MOX-fuel by the introduction of Pa-getter and Cs and comparison of these options for MOX-fuel proliferation protection against unauthorised action allow us to come to the following conclusions:

1. An expediency for applying protactinium as a getter material is defined by the following properties of protactinium:
   - Neutron irradiation converts protactinium into isotope $^{232}\text{U}$, long-lived source of high-energy gamma radiation.
   - If protactinium is introduced into MOX-fuel in the form of small metal particles, then, under neutron irradiation, U-getter is produced in the fuel in form of U–Pa alloy.
   - Being, in essence, a burnable absorber, isotope $^{231}\text{Pa}$ is able to decrease effectively an initial reactivity margin and, thus, to promote realisation of prolonged MOX-fuel lifetimes.

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

3. Under conditions of the increasing terrorist activity all over the world, the proliferation protection standards now used 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 the radioactivity of fuel to decrease many times and allows the fabrication of 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.

7. Application of caesium from fission products (mainly, owing to the availability of isotope $^{137}\text{Cs}$) can enable us to keep the inherent radiation barrier at the level of time till the lethal dose receiving within the minute range. However, high volatility and easy separation from fuel by calcination make caesium less attractive as a proliferation deterrent. The proliferation protection barrier based on $^{231}\text{Pa}$ addition into the fuel is free of this disadvantage, but requires a special technology for $^{233}\text{Pa}$ production. Apparently, combination of these two additions to MOX-fuel is able to enhance the inherent radiation barrier and to weaken shortcomings of both proliferation deterrents.

REFERENCES

1. The main guidelines on nuclear materials control and accounting (NP-030-01) approved by the RF GOSNADZOR’s Decree No. 7 of July 9 (2001).