We are IntechOpen,
the world's leading publisher of
Open Access books
Built by scientists, for scientists

4,800
Open access books available

122,000
International authors and editors

135M
Downloads

Our authors are among the

154
Countries delivered to

TOP 1%
most cited scientists

12.2%
Contributors from top 500 universities

WEB OF SCIENCE™

Selection of our books indexed in the Book Citation Index
in Web of Science™ Core Collection (BKCI)

Interested in publishing with us?
Contact book.department@intechopen.com

Numbers displayed above are based on latest data collected.
For more information visit www.intechopen.com

Isotopic Uranium and Plutonium Denaturing as an Effective Method for Nuclear Fuel Proliferation Protection in Open and Closed Fuel Cycles

Kryuchkov E.F., Tsvetkov P.V.[1], Shmelev A.N., Apse V.A., Kulikov G.G.,
Masterov S.V., Kulikov E.G. and Glebov V.B.
National Research Nuclear University "MEPhI",
[1]Texas A&M University
Russia
[1]USA

1. Introduction

The paper addresses to the problems related with protection of the existing and advanced nuclear fuel types against unauthorized proliferation via introduction of some admixtures into their composition. So, the task may be defined as follows: these admixtures must create the barriers difficult to overcome for the use of nuclear fuels in nuclear explosive devices (NED) but, at the same time, these admixtures must preserve (or even enhance) energy potential of nuclear fuels for further peaceful use at civilian nuclear power plants (NPP). Such an approach to proliferation protection of nuclear fuels is often called as an isotopic denaturing. The term "isotopic denaturing" is used to designate any artificial changes in isotopic composition of chemical element which can give some new desirable properties to this element.

2. Protection of nuclear materials in fuel cycle against unauthorized applications

As is known, main raw materials for NPP are natural uranium and thorium. Natural uranium contains 0.71% ^{235}U, and self-sustainable chain fission reaction (CFR) may be maintained by thermal neutrons provided rather large amounts of uranium, graphite or heavy water are available. As for natural thorium, there are no fissile isotopes, like ^{235}U, in its composition at all. So, manufacturing of a small-size NED with uranium charge requires application of isotopic enrichment in order to produce highly enriched (weapon-grade) uranium (HEU, 90-95% ^{235}U). Some artificial fissile materials (plutonium or ^{233}U) can be used as a nuclear charge but they may be produced only under neutron irradiation of natural uranium or thorium in nuclear reactors.

At present, nuclear reactors apply uranium fuels of various enrichments. So, isotopic composition of uranium-based fuel for civilian NPP takes an intermediate position between

natural uranium and HEU. Conversion of civilian uranium fuel into HEU, or weapon-grade uranium, requires application of isotopic re-enrichment operations.

Plutonium, suitable element for NED, is produced by nuclear reactors in the process of their operation. As is known, weapon-grade plutonium (WGPu) consists mainly of ^{239}Pu with small content of ^{240}Pu (4-7%) and negligible amounts of heavier plutonium isotopes. WGPu may be produced in the special reactors with short irradiation cycle. Power reactors at NPP are operated with relatively long irradiation cycle and, thus, with high fuel burn-up. So, plutonium produced in power reactors (reactor-grade plutonium, RGPu) contains the larger amounts of heavier plutonium isotopes. Lengthy irradiation of uranium fuel can change significantly isotopic composition of uranium (in particular, isotopes ^{232}U and ^{236}U do appear). As an example, isotopic composition of uranium fuel (initial enrichment - 4.4% ^{235}U) after LWR operation up to fuel burn-up of 4% HM is presented in Table 1. Thus, uranium fuel may be used to produce fissile materials, suitable for NED, in nuclear reactors but it requires application of some special operations.

Composition of fresh fuel		Composition of uranium and plutonium in spent fuel, % (fuel burn-up - 4 % HM)				
4.4 % ^{235}U	Uranium	^{232}U	^{235}U	^{236}U	^{238}U	
		$1.4\cdot10^{-9}$	1.26	0.59	98.15	
	Plutonium (RGPu)	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
		1.7	58.0	22.3	12.3	5.7

Table 1. Isotopic compositions of uranium and plutonium in spent LWR fuel

Artificial fissile uranium isotope ^{233}U, high-efficiency material for NED, may be produced by neutron irradiation of natural thorium. However, undesirable by-products are generated in this process including other uranium isotopes, namely ^{232}U, ^{234}U and ^{236}U. In particular, light uranium isotope ^{232}U can complicate significantly any operations with produced uranium.

Radiochemical reprocessing of spent nuclear fuel (SNF) includes extraction of radioactive fission products (FP) whose intense emission of ionizing radiation creates a protective barrier against unauthorized access to fissile materials. So, SNF reprocessing can be regarded as an operation which can remove (or, at least, weaken under incomplete purification) the radiation barrier. SNF partitioning into separate elements or groups of elements can facilitate diversion of fissile materials. In order to reduce the diversion risk, appropriate protective actions should be undertaken. In addition to the organizing measures related with physical protection of nuclear enterprises and technological control, the following barriers against diversion of fissile materials should be considered:

1. Radiation barrier mainly formed by FP. Such a barrier can be formed in the process of the reactor operation and by a special short-term irradiation of fresh fuel assemblies in the dedicated nuclear facilities. Radiation background of fissile materials is one else component of the radiation barrier.
2. Incomplete SNF purification from radioactive FP can result in intense radiation fields which make very difficult any further operations with extracted fuel.
3. Isotopic dilution of ^{235}U, i.e. the use of relatively low-enriched uranium (LEU).
4. Incomplete separation of uranium from plutonium (or full exclusion of any technological procedures needed for uranium-plutonium separation) in the course of SNF reprocessing. This measure makes it impossible to use these materials directly in NED.

5. Application of automatic remote technology for fuel, fuel rods and fuel assemblies fabrication. This measure can complicate access to fissile materials.

6. Dilution (or denaturing) of fissile materials by their isotopes which can complicate the use of fissile materials in NED. A particular case of isotopic denaturing is a well-known dilution of ^{235}U by ^{238}U, i.e. the use of LEU fuel. Another example: neutron irradiation of mixed uranium-thorium fuel in nuclear reactors can produce fissile isotope ^{233}U isotopically diluted by ^{238}U.

If we consider fissile plutonium isotope ^{239}Pu, then the heavier plutonium isotopes (mainly, ^{240}Pu) play the same role of isotopic diluents. In addition, light plutonium isotope ^{238}Pu is an intense source of spontaneous fission neutrons and intense source of thermal energy from α-decays (half-life of ^{238}Pu is equal to 87.7 years).

In thorium fuel cycle the same role may be played by ^{232}U (half-life – 68.9 years).

In practice, the measures listed above may be used in combinations. For example, open fuel cycle of power LWR is provided with the following protective barriers:

- Isotopic dilution (LEU fuel).
- Intense radiation background caused mainly by FP.
- SNF contains non-separated mixture of uranium and plutonium.

Just this set of protective barriers represents a basis for the USA standard on SNF proliferation protection (Spent Fuel Standard (USA National Academy of Sciences, 2000)).

As ^{232}U and its neutron predecessor ^{231}Pa are nuclides of low abundance, it seems reasonable to consider their properties in detail.

3. Nuclear properties of ^{232}U and ^{231}Pa

Basic nuclear properties of main uranium isotopes are presented in Table 2 (Reilly et al., 1991; OECD Nuclear Energy Agency, 1997). As it may be seen, some nuclear properties of ^{232}U make it a valuable material for proliferation protection of uranium-based nuclear fuel.

	^{232}U	^{234}U	^{235}U	^{238}U
Half-life, years	68.9	2.45×10^5	7.04×10^8	4.47×10^9
Specific yield of α-particles, $1/(g \cdot s)$	8×10^{11}	2.3×10^8	7.9×10^4	1.2×10^4
Mean energy of α-particles, MeV	5.3	4.76	4.4	4.19
Specific yield of spontaneous fission neutrons, $1/(g\ s)$	1.3	5.02×10^{-3}	2.99×10^{-4}	1.36×10^{-2}
Fission cross-section ($E_n = 0.0253$ eV), barns	77.15	0.465	583.2	1.2×10^{-5}

Table 2. Basic nuclear properties of main uranium isotopes

^{232}U is a starting isotope for chain of radioactive decays, and some ^{232}U decay products (^{208}Tl, ^{212}Bi) emit high-energy gamma-radiation (2.6 MeV and 1.8 MeV, respectively) that improves detectability of ^{232}U-containing nuclear materials (Gilfoyle & Parmentola, 2001) and complicates radiation conditions, especially for any unauthorized actions. Nuclear properties of main ^{232}U decay products are presented in Table 3.

Decay products	^{228}Th	^{224}Ra	^{220}Rn	^{216}Po	^{212}Bi	^{212}Po
Half-life	1.91 years	3.62 days	55.6 sec	0.145 sec	1.01 hours	3×10^{-7} sec
Energy of α-particles, MeV (relative intensity)	5.42 (71.7%) 5.34 (27.6%)	5.69 (94.9%) 5.45 (5.1%)	6.29 (100%)	6.78 (100%)	6.09 (9.7%) 6.05 (25.2%)	8.78 (100%)

Table 3. ^{232}U decay products, emitters of α-particles

Like ^{238}U, isotope ^{231}Pa can play a role of a fertile nuclide which is not split by thermal neutrons but promotes breeding of fissile materials. Dependencies of radiative capture cross-sections on neutron energy are presented in Fig. 1 for isotopes ^{238}U and ^{231}Pa.
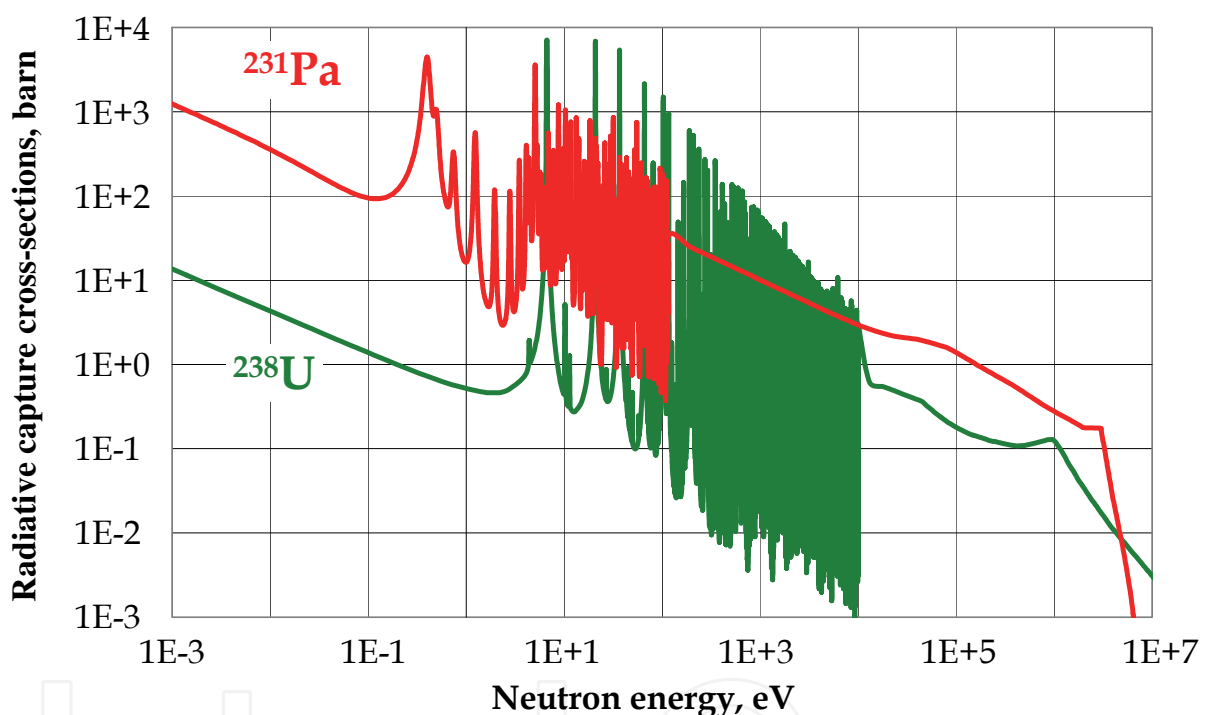
Fig. 1. Dependency of radiative capture cross-sections on neutron energy for ^{238}U and ^{231}Pa

The following two important aspects should be noted here. Firstly, within thermal energy range, ^{231}Pa is a superior neutron absorber as compared with ^{238}U. For example, radiative capture cross sections of thermal neutrons ($E_n = 0.0253$ eV) for these two isotopes are equal to: $\sigma_c(^{231}$Pa$) = 227$ barns, $\sigma_c(^{238}$U$) = 3$ barns. So, the presence of ^{231}Pa in fuel composition can promote effective generation of fissile isotopes ^{232}U and ^{233}U. Secondly, there is a rather large energy distance between capture resonances of ^{238}U and ^{231}Pa. Capture resonances of ^{231}Pa belong to relatively low energies (below 100 eV). This means the presence of ^{231}Pa in fuel composition can depress thermal region in energy spectrum of neutrons (see Fig. 2).

It can be seen that, although neutron energy spectrum in VVER-1000 contains a certain fraction of thermal neutrons, introduction of ^{231}Pa into fuel composition can remove the thermal fraction completely. That is why stainless steel may be used here as a structural material. Indeed, the absence of thermal fraction in neutron spectrum does not result in

additional neutron loss but fuel rods can keep their ability for working up to the higher values of fuel burn-up than with zirconium-based alloys.

Now let us consider nuclear properties of ^{232}U, product of neutron capture by ^{231}Pa and rapid β-decay of ^{232}Pa ($T_{1/2}$ (^{232}Pa) = 1,3 days (Babichev et al.,1991)). Like ^{235}U, isotope ^{232}U is a fissile nuclide. Dependencies of fission cross-sections on neutron energy are presented in Fig. 3 for isotopes ^{235}U and ^{232}U.

It can be seen that, within thermal energy range, fission cross-sections of ^{232}U are substantially lower than those for ^{235}U while radiative capture cross-sections of these isotopes are comparable each other. For example, radiative capture cross sections of thermal neutrons (E_n = 0.0253 eV) for these two isotopes are equal to: σ_c(^{232}U) = 73 barns, σ_c(^{235}U) = 99 barns. So, neutron-multiplying properties of ^{232}U are inferior to those of ^{235}U within thermal energy range.

This conclusion can be confirmed by Fig. 4 which demonstrates energy dependency of (v_{ef} - 1), i.e. the number of excess fission neutrons per one absorbed neutron.

^{235}U looks superior to ^{232}U within thermal energy range but quite another situation takes place in resonance range. So, it may be expected that introduction of ^{231}Pa into uranium-based fuel composition with aim to increase fuel burn-up will be more efficient just in resonance neutron spectrum.
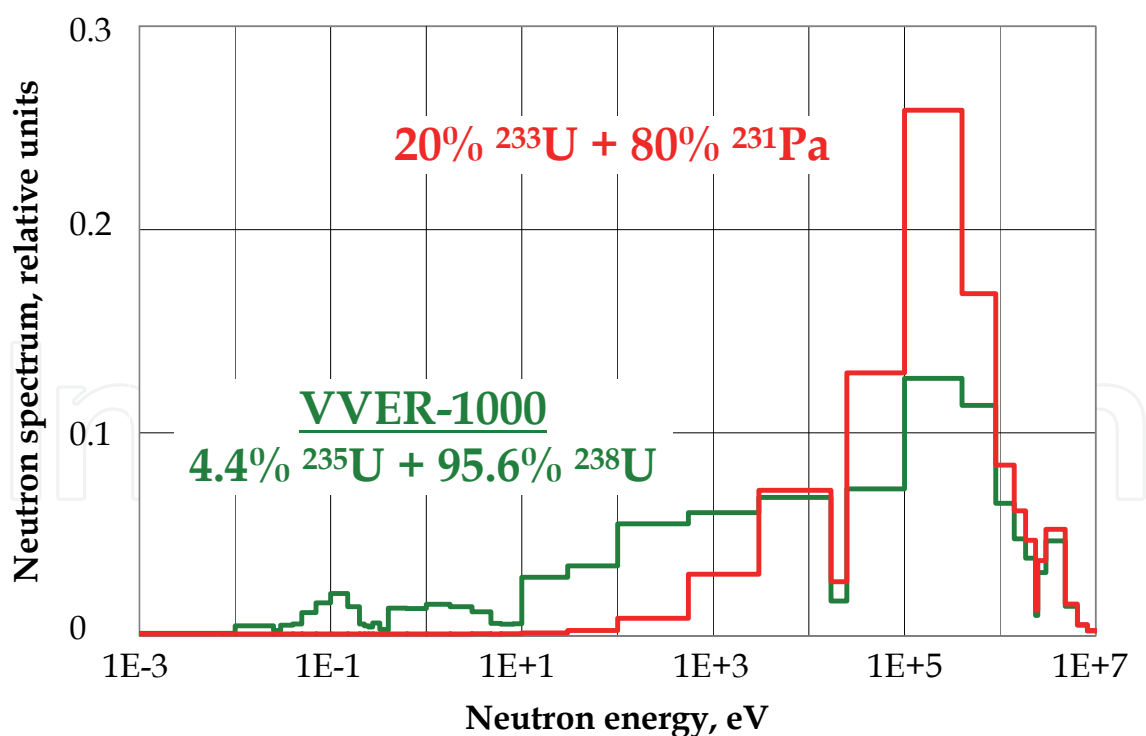

Fig. 2. ^{231}Pa effect on energy spectrum of neutrons

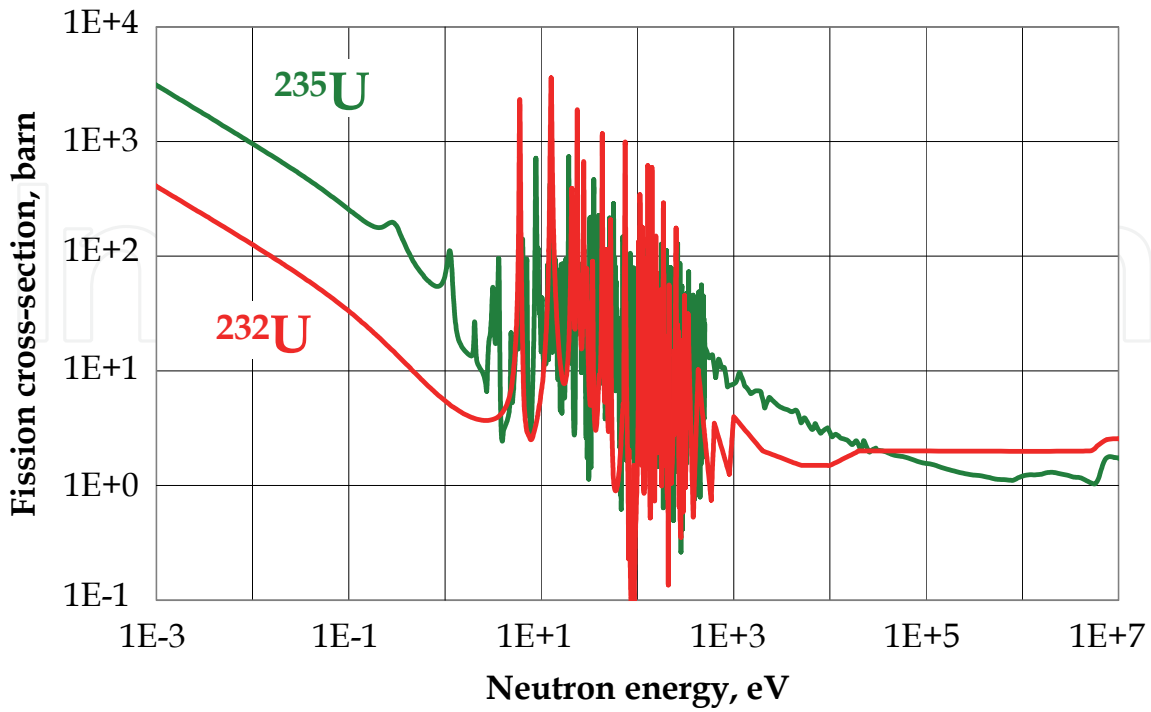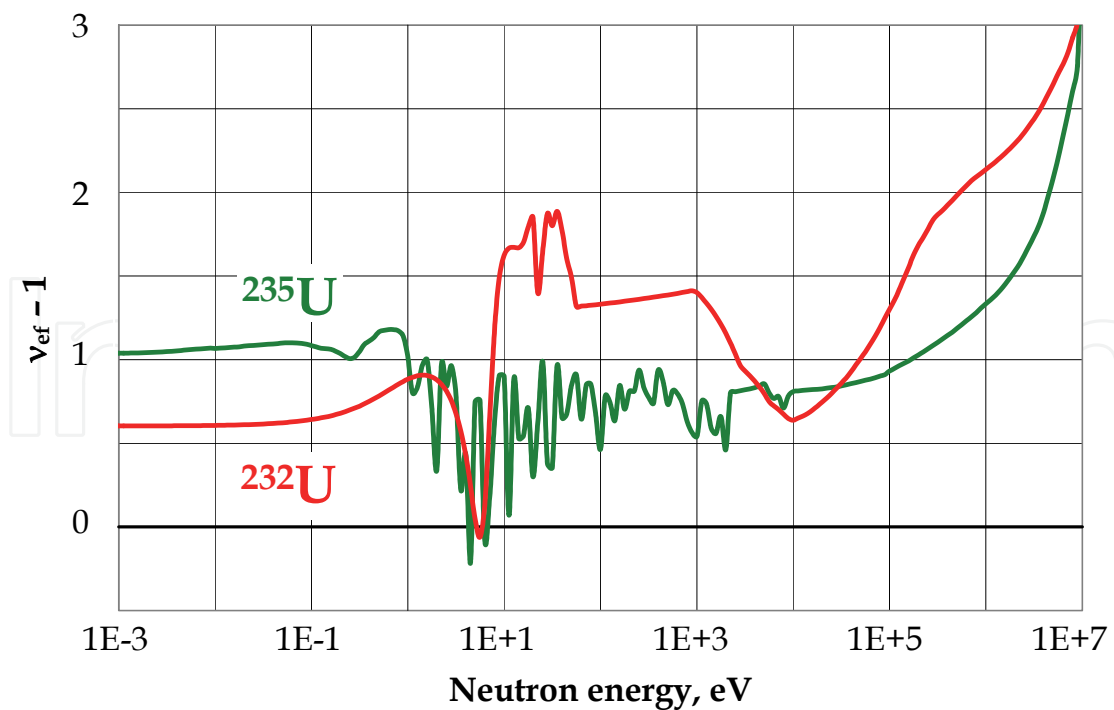Fig. 3. Dependency of fission cross-sections on neutron energy for isotopes ^{235}U and ^{232}U

Fig. 4. Energy dependency of the number of excess fission neutrons per one absorbed neutron for isotopes ^{235}U and ^{232}U

4. Proliferation resistance of nuclear materials in open fuel cycle. The ways for closure of fuel cycle

Presently, there are different points of view on future development of nuclear fuel cycles. Some countries (USA, Canada, Germany and Sweden) are implementing in practice an open nuclear fuel cycle (Fig. 5) that does not foresee a radiochemical SNF reprocessing in the visible future. One of the reasons for this choice is a wish of decreasing a risk of nuclear weapon proliferation. SNF may be only converted into the forms suitable for long-term safe storage. However, such a strategy of nuclear power development has already resulted in large SNF stockpiles, potentially dangerous nuclear materials (NM). So, the preferable option for future development of nuclear power consists in transition to the closed fuel cycles with SNF reprocessing, separation of radioactive FP and recycling of residual fuel.
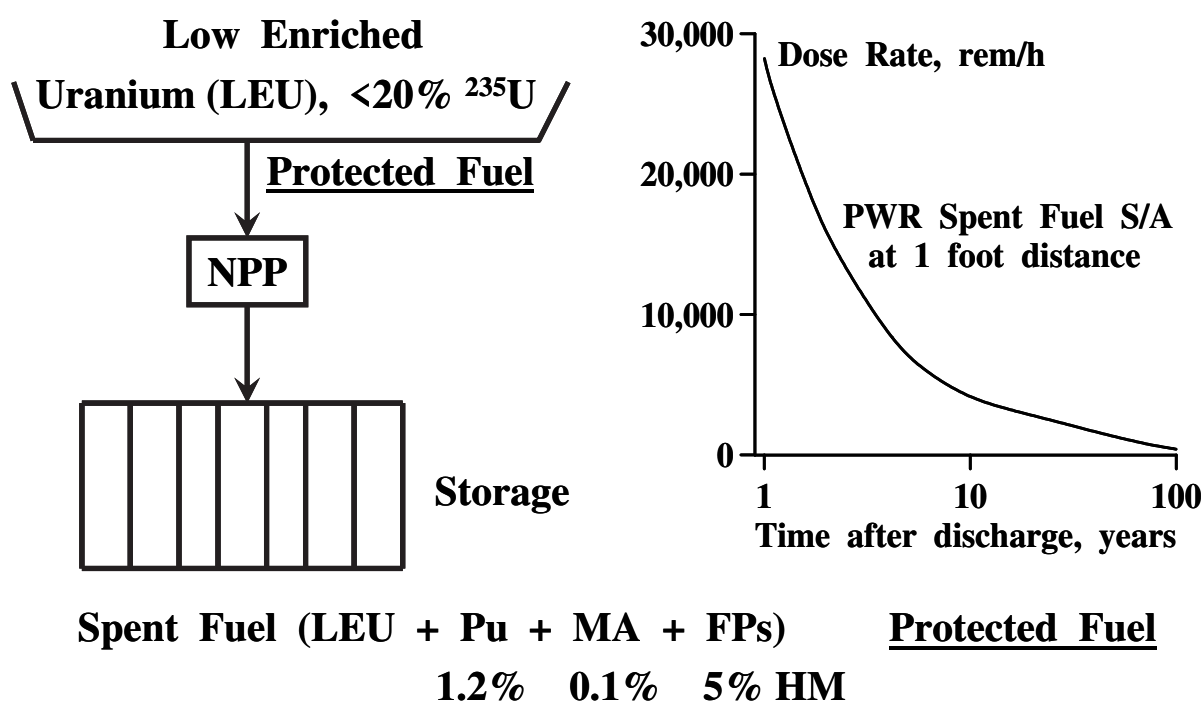
Fig. 5. Open fuel cycle

LEU is a fresh fuel for open nuclear fuel cycle. Plutonium in spent fuel assemblies is protected by intense gamma-radiation of fission products (Fig. 5). That is why unirradiated materials are more vulnerable for unauthorized proliferation.

Isotopic uranium denaturing may be regarded as an effective method for upgrading self-protection of unirradiated uranium-containing materials. The term "isotopic denaturing" is used conventionally for designation of any artificial changes in natural isotopic composition of a chemical element with aim to give him some new, desirable properties. In practice, uranium may be denatured by the following two ways: direct introduction of intense radioactive isotope ^{232}U into uranium fuel composition or direct introduction of relatively weaker radioactive isotope ^{231}Pa into uranium fuel composition. ^{231}Pa is a neutron predecessor of ^{232}U, main isotope of uranium denaturing. So, only short-term pre-irradiation of fresh fuel assemblies in the research reactors may be sufficient to produce proliferation resistant fuel assemblies, suitable even for export deliveries.

4.1 Isotopic denaturing of uranium as a way for creating an internal source of α-particles

Along with progress in development of high-efficiency enriching technologies, potential threat of LEU diversion and re-enrichment up to the weapon-grade level excites more and more apprehensions. These reasons indicate that, besides reduction of uranium enrichment below 20% ^{235}U, other measures may be also required to upgrade LEU self-protection against its unauthorized re-enrichment. Taking into consideration the growing world-wide scope of LEU utilization, including LEU with enrichment in the vicinity of the upper boundary (~20% ^{235}U), high LEU vulnerability to unauthorized re-enrichment must be recognized. Particular apprehensions are related with 20%-uranium. So, some additional actions should be undertaken to protect LEU against its unauthorized re-enrichment.

The effects of ^{232}U introduction into LEU are caused by the following specific properties of ^{232}U (see Table 2):

- Good neutron-multiplying properties of ^{232}U (Ganesan et al., 2002) and its neutron predecessor ^{231}Pa make it possible to extend time period of continuous reactor operation without refueling up to the values comparable with the reactor life-time. As a result, unauthorized extraction of plutonium from spent fuel assemblies becomes unfeasible.

- It is impossible to remove ^{232}U from denatured uranium without application of sophisticated and expensive isotope separation technologies.

- ^{232}U is a neutron source from spontaneous fission reactions and a source of high-energy α-particles. Alpha-particles emitted by ^{232}U are able to dissociate molecules of uranium hexafluoride and, thus, could make it practically impossible to re-enrich denatured uranium up to the weapon-grade level. Besides, α-particles are able to initiate (α,n)-reactions with impurities of light elements (LE) and, thus, intensify internal neutron generation. Growth of neutron background in the re-enriching process of LEU-uranium containing 0.1÷0.5% ^{232}U can decrease the CFR energy yield by three orders of magnitude. In essence, NED with such a re-enriched uranium is a "dirty" bomb only. Thus, export deliveries of LEU-based fuel assemblies to foreign NPP receive an additional proliferation barrier.

5. Increased burn-up of proliferation protected LWR fuel containing ^{231}Pa

One of specific features in operation of nuclear power reactors consists in a necessity to perform regular refuelings. This necessity is caused by the following effects: depletion of fissile materials, FP accumulation, potential rupture of fuel cladding with intense release of radioactive materials. LWR, the mostly spread type of power reactors, requires refueling every 1-2 years, when fuel burn-up reaches 4-6% HM.

Extension of fuel life-time up to relatively long time periods (several decades, for instance) can reduce drastically the number of refuelings or exclude them at all. Reduction or full exclusion of refueling procedures decreases the demands for fresh fuel and decreases quantity of SNF discharged per unit of produced energy. Those reactors, which are capable to operate for a sufficiently long time without any refueling, may be used as the only energy source in remote regions, at the floating NPP, as energy source for space investigations (research bases on the Moon or Mars, cosmic flights into the outer space). Our studies demonstrated that introduction of ^{231}Pa into LWR fuel composition could extend significantly the fuel life-time and reach ultra-high fuel burn-up.

It should be noted that achievability of ultra-high fuel burn-up was studied here only from the standpoint of neutron-multiplying properties of advanced fuel compositions. The problems of suitable structural materials, evolution of their strength properties and durability for a long fuel life-time are not analyzed here. At present, maximal fuel burn-up (about 30% HM) was achieved in the research fast reactor BOR-60 (Grachev et al., 2003). It may be expected that the higher values of fuel burn-up could be achieved if the following operations would be multiply carried out: partial fuel burn-up (near to the practically achievable value of 30% HM), application of DUPIC-technology for removal of gaseous and volatile FP, re-fabrication of fresh fuel pellets.

5.1 Evolution of neutron-multiplying properties in chains of isotopic transformations

In this section we compared time evolutions of neutron-multiplying properties in two isotopic chains: traditional chain that starts from ^{232}Th (^{232}Th → ^{233}U → ^{234}U → …) and non-traditional chain that starts from ^{231}Pa (^{231}Pa → ^{232}U → ^{233}U → …) (see Fig. 6). Radiative capture cross-sections σ_c and fission cross-sections σ_f were calculated for a typical neutron spectrum of VVER-1000 (β-decays were not taken into account).
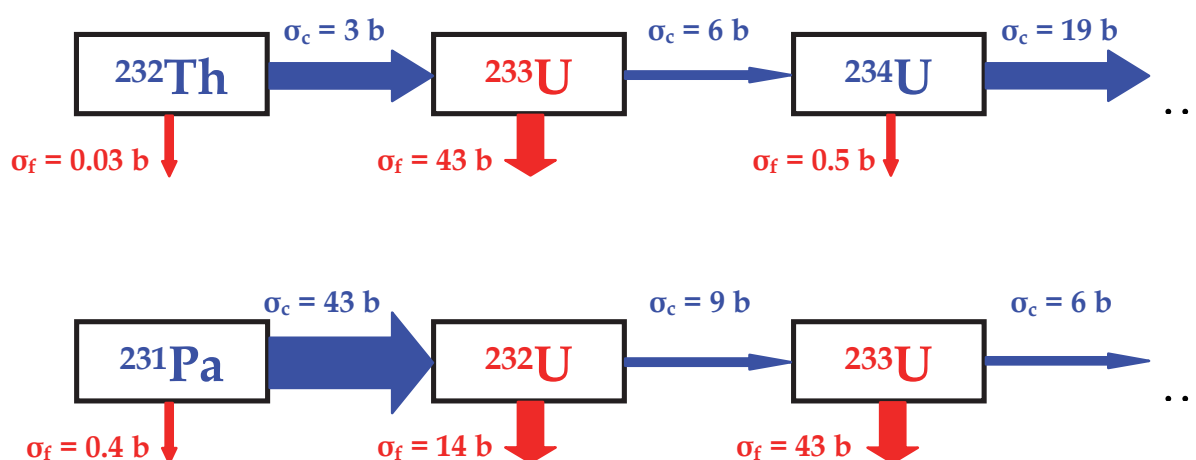

Fig. 6. Chains of isotopic transformations (^{232}Th → ^{233}U → ^{234}U → …) and (^{231}Pa → ^{232}U → ^{233}U → …) (neutron spectrum of VVER-1000)

It can be seen that neutron-multiplying properties in non-traditional chain are gradually improved: the starting isotope ^{231}Pa is a neutron absorber, fission cross-section of the second isotope ^{232}U prevails over its capture cross-section, and the third isotope ^{233}U is a well-fissionable material. So, non-traditional chain represents the combination of two consecutive fissionable isotopes (^{232}U and ^{233}U) while, in traditional chain, the third isotope ^{234}U is a neutron absorber only.

Thus, in non-traditional chain, parasitic neutron absorption by FP and depletion of fissile materials during the reactor operation can be partially compensated by ^{231}Pa feeding. This makes it possible to talk about a possibility for substantial extension of fuel life-time and achievability of ultra-high fuel burn-up. By the way, in traditional LWR fuel, the negative effects caused by FP accumulation and depletion of fissile materials are compensated by ^{238}U(n,γ)^{239}Pu chain significantly weaker than by ^{231}Pa(n,γ)^{232}U(n,γ)^{233}U chain in non-traditional fuel because of lower capture cross-sections: $\sigma_c(^{238}$U) = 0.9 barns, $\sigma_c(^{231}$Pa) = 43 barns.

So, it can be concluded that non-traditional chain (^{231}Pa → ^{232}U → ^{233}U → …) appears to be more attractive from the standpoint of neutron-multiplying properties (as a consequence, from the standpoint of extended fuel life-time or achievability of ultra-high fuel burn-up) in comparison with traditional chain (^{232}Th → ^{233}U → ^{234}U → …) due to the following reasons:

1.   Combination of two consecutive well-fissionable isotopes (^{232}U and ^{233}U).
2.   High rate of their generation from the starting isotope ^{231}Pa, whose neutron capture cross-section is larger substantially than that for the starting nuclide ^{232}Th in traditional chain of isotopic transformations.

It is noteworthy that ^{231}Pa may be regarded, to a certain extent, as a burnable neutron poison: for fuel life-time ^{231}Pa is burnt up to 80% and converted into well-fissionable isotopes, neutron capture cross-section of ^{231}Pa is substantially larger than that of fertile isotope ^{232}Th.

As is known, the existing LWRs are characterized by thermal neutron spectrum. In advanced LWR designs, for example, in LWR with supercritical coolant parameters (SCLWR), different regions of the reactor core are characterized by different neutron spectra depending on coolant density. Thermal spectrum prevails within the core region containing dense coolant ($\gamma = 0.72$ g/cm^3) while resonance neutron spectrum dominates within the core region containing coolant of the lower density ($\gamma = 0.1$ g/cm^3) (Kulikov, 2007).

Reasonability of ^{231}Pa introduction into fuel composition for the cases of thermal and resonance neutron spectra is analyzed in the next section.

5.2 Reasonability of ^{231}Pa involvement in the case of thermal neutron spectrum

Numerical analyses of fuel depletion process were carried out with application of the computer code SCALE-4.3 (Oak Ridge National Laboratory, 1995) and evaluated nuclear data file ENDF/B-V for elementary cells of VVER-1000. The only exception consisted in the use of martensite steel MA956 (elemental composition: 74,5% Fe, 20% Cr, 4,5% Al, 0,5% Ti and 0,5% Y$_2$O$_3$) instead of zircaloy as a fuel cladding material. Substitution of martensite steel for zirconium-based cladding is caused by the higher values of fuel burn-up.

Traditional (^{232}Th-^{233}U) and non-traditional (^{231}Pa-^{232}Th-^{233}U) fuel compositions were compared for the case of thermal neutron spectrum (coolant density – 0.72 g/cm^3). Infinite neutron multiplication factor K$_\infty$ is shown in Fig. 7 as a function of fuel burn-up.

It can be seen that substitution of ^{231}Pa for ^{232}Th decreases K$_\infty$ at the beginning of cycle, i.e. decreases an initial reactivity margin to be compensated. This effect is caused by different capture cross-sections of these isotopes - ^{231}Pa is a significantly stronger neutron absorber than ^{232}Th. In parallel, thanks to the larger capture cross-section of ^{231}Pa, intense breeding of two consecutive well-fissionable isotopes (^{232}U and ^{233}U) takes place. So, gradual introduction of ^{231}Pa into fuel composition results in the smoother relaxation of neutron multiplication factor in the process of fuel burn-up.

Acceptable fraction of ^{231}Pa in non-traditional fuel composition is limited by the value of neutron multiplication factor (above unity) at the beginning of cycle. So, the effects caused by introduction of ^{231}Pa may take place only in those fuel compositions where fraction of main fissile isotope is sufficiently large. For example, fraction of main fissile isotope ^{233}U may be increased up to the level corresponding to the situation when neutron multiplication factor at the beginning of cycle is equal to about 1.10 at full replacement of ^{232}Th by ^{231}Pa. The calculations showed that this condition may be satisfied at maximal ^{233}U fraction about 30%. Evolution of neutron multiplication factor in the process of fuel burn-up is presented in Fig. 8 for traditional and non-traditional fuel compositions.
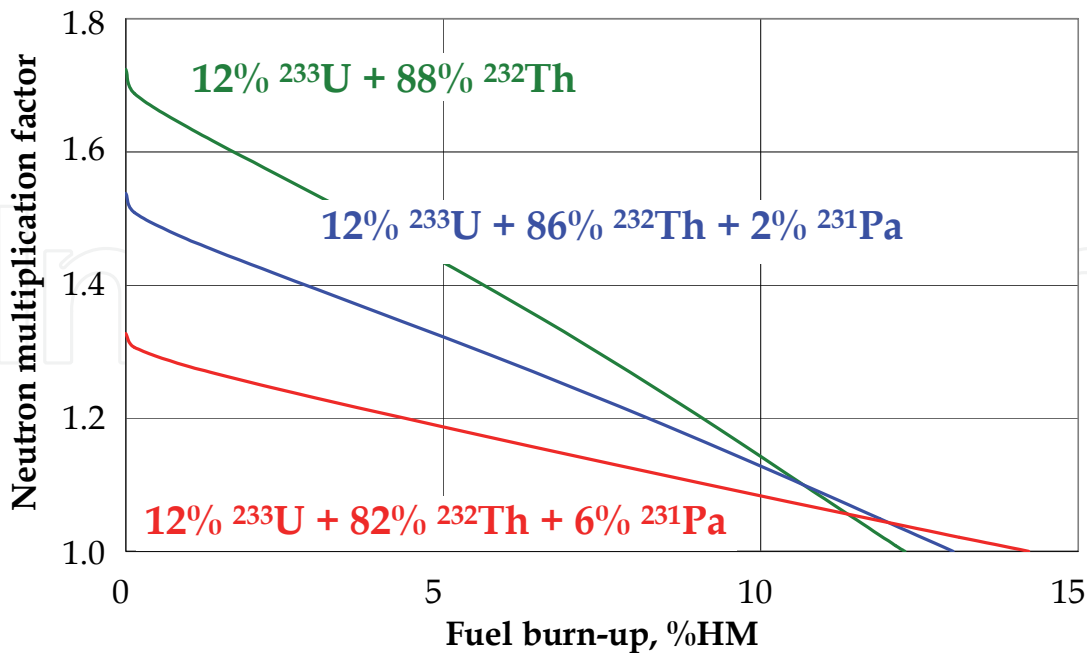
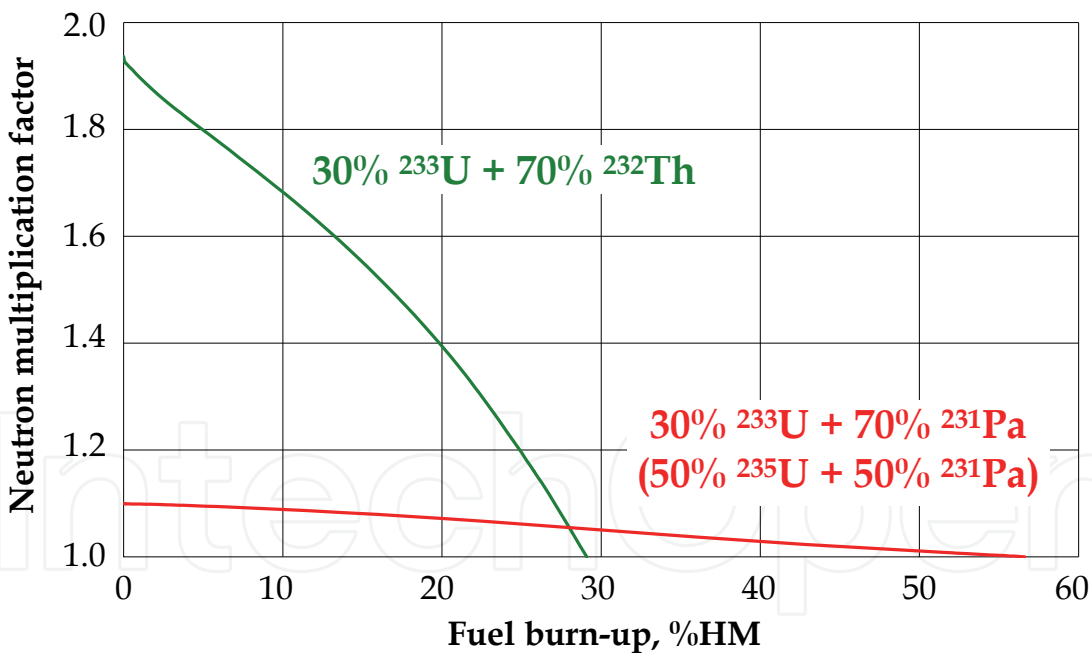Fig. 7. ^{231}Pa effects on fuel burn-up in thermal neutron spectrum

Fig. 8. Achievability of ultra-high fuel burn-up by introduction of ^{231}Pa (thermal neutron spectrum)

As is seen from Fig. 8, traditional thorium-based fuel (30% ^{233}U + 70% ^{232}Th) provides rather high reactivity margin (K_∞ (BOC) ≈ 1,9) with achievable value of fuel burn-up about 29% HM. Introduction of ^{231}Pa into fuel composition decreases initial reactivity margin but, at the same time, increases fuel burn-up. If ^{232}Th is completely replaced by ^{231}Pa, i.e. (30% ^{233}U + 70% ^{231}Pa) fuel composition is analyzed, then neutron multiplication factor remains

practically unchanged in the vicinity of unity for a full duration of fuel life-time. This means that the negative effects from neutron absorption by FP and depletion of fissile isotope are almost completely compensated by breeding of secondary fissile isotopes from [231]Pa. In this case, about 80%-part of [231]Pa is converted into secondary fissile isotopes which can provide ultra-high fuel burn-up (near to 57% HM).

If fuel loading in such a reactor is similar to the fuel loading of VVER-1000 (about 66 tons), then achievable value of fuel life-time is near to 40 years for the reactor power of 3000 MWt.

It is interesting to note that [235]U as well as [233]U may be used to achieve ultra-high fuel burn-up. Moreover, [235]U option looks very attractive because of two reasons: firstly, [235]U resources are more available than resources of [233]U, and, secondly, achievement of the same fuel burn-up will require lower quantity of [231]Pa, artificial isotope to be produced in the dedicated nuclear power facilities.

5.3 Reasonability of [231]Pa involvement in the case of resonance neutron spectrum

Traditional ([232]Th-[233]U) and non-traditional ([231]Pa-[232]Th-[233]U) fuel compositions were compared for the case of resonance neutron spectrum (coolant density – 0.1 g/cm³). Infinite neutron multiplication factor K_∞ is shown in Fig. 9 as a function of fuel burn-up.
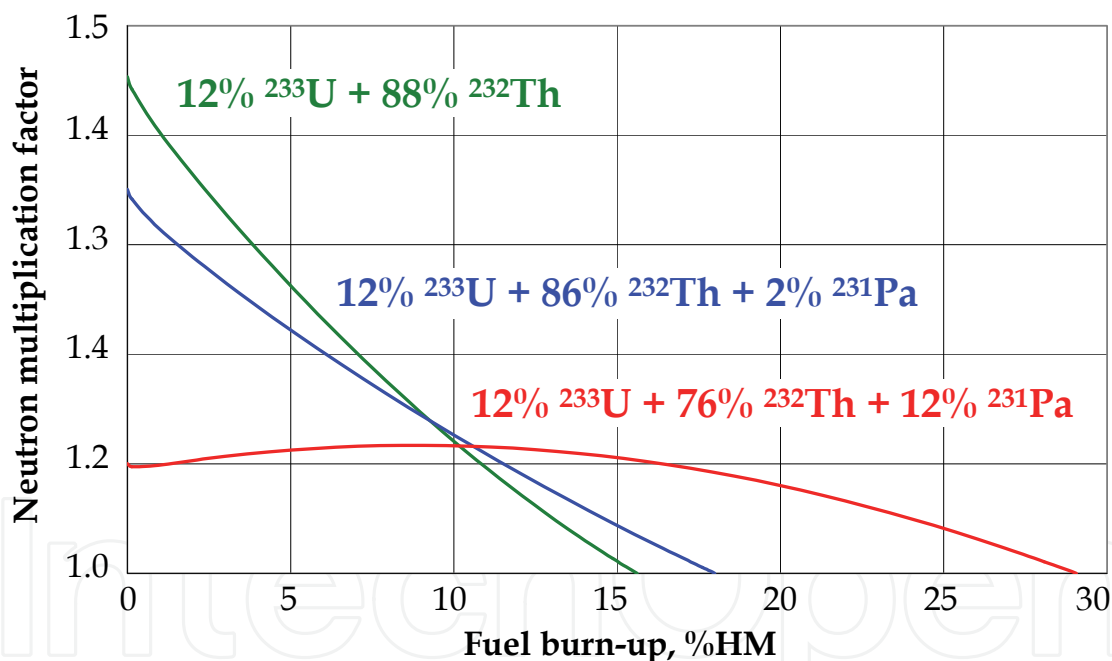

Fig. 9. [231]Pa effects on fuel burn-up in resonance neutron spectrum

Comparison of the curves presented in Figs. 7, 9 allows us to conclude that introduction of [231]Pa into fuel composition is more preferable from the standpoint of higher fuel burn-up in the case of resonance neutron spectrum. This conclusion can be explained by better neutron-multiplying properties of [232]U just in resonance neutron spectrum as compared with thermal neutron spectrum (see Fig. 4).

As it follows from Fig. 9, introduction of only 12% [231]Pa increased fuel burn-up twice. Neutron multiplication factor at the beginning of cycle increased too, i.e. neutron-multiplying properties of fuel composition became better.

Like previous analysis, fraction of main fissile isotope ^{233}U may be increased up to the level corresponding to the situation when neutron multiplication factor at the beginning of cycle is equal to about 1.10 at full replacement of ^{232}Th by ^{231}Pa. In addition, potential use of ^{235}U instead of ^{233}U was analyzed to evaluate a possibility for achieving ultra-high fuel burn-up.

So, numerical studies confirmed reasonability for introduction of ^{231}Pa into fuel composition because this introduction results in reduction of initial reactivity margin and in substantial growth of fuel burn-up. Maximal positive effect from introduction of ^{231}Pa may be observed in resonance neutron spectrum. Besides, introduction of ^{231}Pa makes it possible to reach ultra-high fuel burn-up regardless of what main fissile isotope is used, ^{233}U or ^{235}U. In particular, (20% ^{233}U + 80% ^{231}Pa) fuel composition can reach fuel burn-up of 76% HM in resonance neutron spectrum (see Fig. 10).
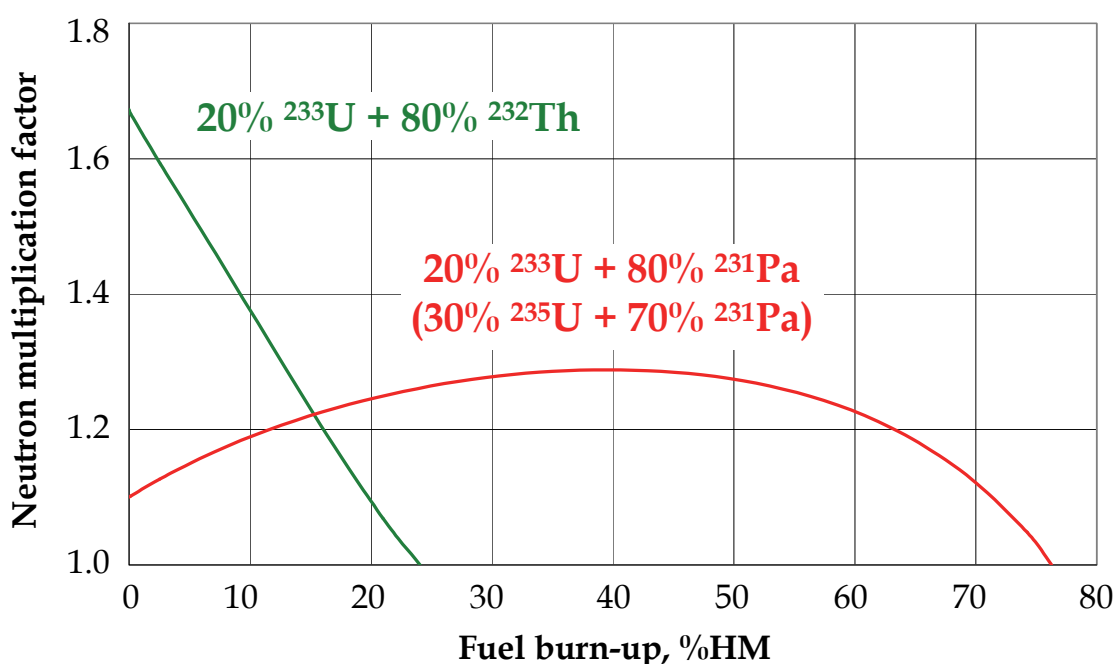

Fig. 10. Achievability of ultra-high fuel burn-up by introduction of ^{231}Pa (resonance neutron spectrum)

5.4 Effects of ^{231}Pa on safety of the reactor operation

On the one hand, introduction of ^{231}Pa into fuel composition can provide small value of initial reactivity margin and high value of fuel burn-up. On the other hand, if relatively large ^{231}Pa fraction is introduced into fuel composition, reactivity feedback on coolant temperature becomes positive, and safety of the reactor operation worsens.

Numerical studies demonstrated that, if maintenance of favorable reactivity feedback on coolant temperature during fuel life-time is a mandatory requirement, then, in thermal neutron spectrum, ^{231}Pa fraction in fuel composition is limited by a quite certain value while, in resonance neutron spectrum, introduction of ^{231}Pa is impossible at all. However, this conclusion is correct only for large-sized reactors, where neutron leakage is negligible.

So, only thermal neutron spectra should be considered to provide favorable reactivity feedback on coolant temperature. The results presented in Fig. 11 demonstrate a possibility for increasing fuel burn-up in thermal neutron spectrum by introduction of ^{231}Pa into fuel composition.
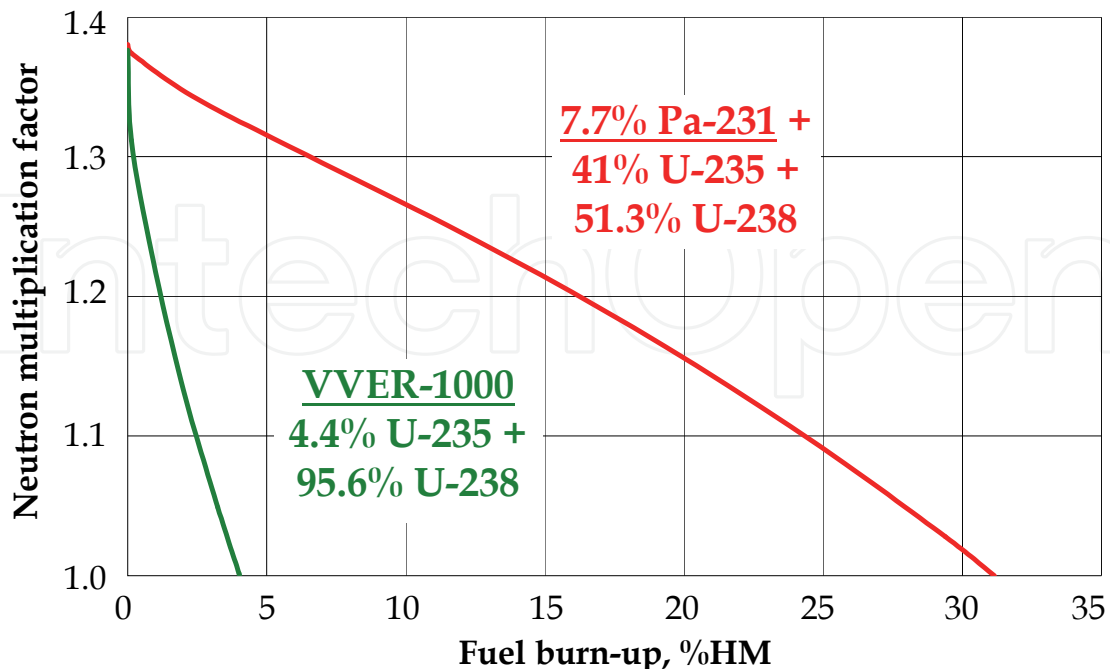
Fig. 11. Achievability of ultra-high fuel burn-up by introduction of ^{231}Pa with conservation of favorable feedback on coolant temperature (thermal neutron spectrum)

As is known, fuel burn-up in VVER-1000 can reach a value about 4% HM. Introduction of ^{231}Pa and higher contents of ^{235}U can increase fuel burn-up by a factor of 8 with the same initial reactivity margin, i.e. more powerful system of reactivity compensation is not required.

Requirement of favorable reactivity feedback on coolant temperature completely excludes any introduction of ^{231}Pa into fuel composition in the case of large-sized reactors with resonance neutron spectra. But , introduction of ^{231}Pa into fuel composition of small-sized reactors does not worsen safety of the reactor operation because of relatively large neutron leakage. This indicates that the mostly attractive area for ^{231}Pa applications is a small nuclear power including small-sized NPP for remote regions, for the floating NPP, for space stations on the Moon or Mars and for cosmic flights into the outer space.

The following conclusions can be made in respect of potential ^{231}Pa applications:

- Application of ^{231}Pa as a burnable neutron poison can reduce initial reactivity margin and increase fuel burn-up.
- Introduction of ^{231}Pa into fuel composition makes it possible to reach ultra-high fuel burn-up (above 30% HM) both in thermal and resonance neutron spectra.
- The actual problem of ^{231}Pa production in significant amounts should be resolved.

6. Proliferation protection of nuclear materials in closed uranium-plutonium fuel cycle

NPP operation in open fuel cycle results in accumulation of huge SNF stockpiles that represents a long-term hazard to the humankind. Ultimate SNF disposal is a difficult technical problem requiring large number of practically "eternal" deep underground repositories. That is why many various options for closure of nuclear fuel cycle (NFC) are

currently under research and development including extraction of residual uranium, plutonium and minor actinides from SNF.

As known, closed uranium-plutonium NFC includes reprocessing and recycling of nuclear fuel and evokes a lot of contradictory opinions with respect to potential risk of plutonium proliferation. This connected with two points:

- Although plutonium extracted from SNF of power reactors (for example, LWR of PWR, BWR or VVER type) is not the best material for nuclear weapons, nevertheless it can be used in NED of moderate energy yield (Mark, 1993).
- Recycled plutonium will be disposed at the facilities of closed NFC, and this will increase the probability of it using for illegal aims (diversion, theft).

Under these conditions, the absence of any internationally coordinated plan concerning the utilization or ultimate SNF disposal enforced the leading nuclear countries to undertake the steps directed to strengthening the nonproliferation regime (IAEA safeguards, Euratom's embargo on the export of SNF reprocessing technology). But several countries, in the first turn the USA, refused from deployment of breeder reactors which are intended for operation in closed NFC, and focused at once-through NFC. On the other hand, the social demand of solving excess fissile materials (plutonium, the first of all) problem which have both civil and military origins, stimulated carrying out the research on plutonium utilization in MOX-fuel. At the same time, the studies of advanced NFC protected against uncontrolled proliferation of fissile materials have been initiated.

6.1 Radiation protection of MOX-fuel. GNEP initiative

Specialists from ORNL (USA) investigated the ways for introduction of γ-radiation sources into fresh fuel (Selle et al., 1979). Sixty-four γ-active radionuclides were selected and studied as candidates for admixing into fresh fuel (see Fig. 12).

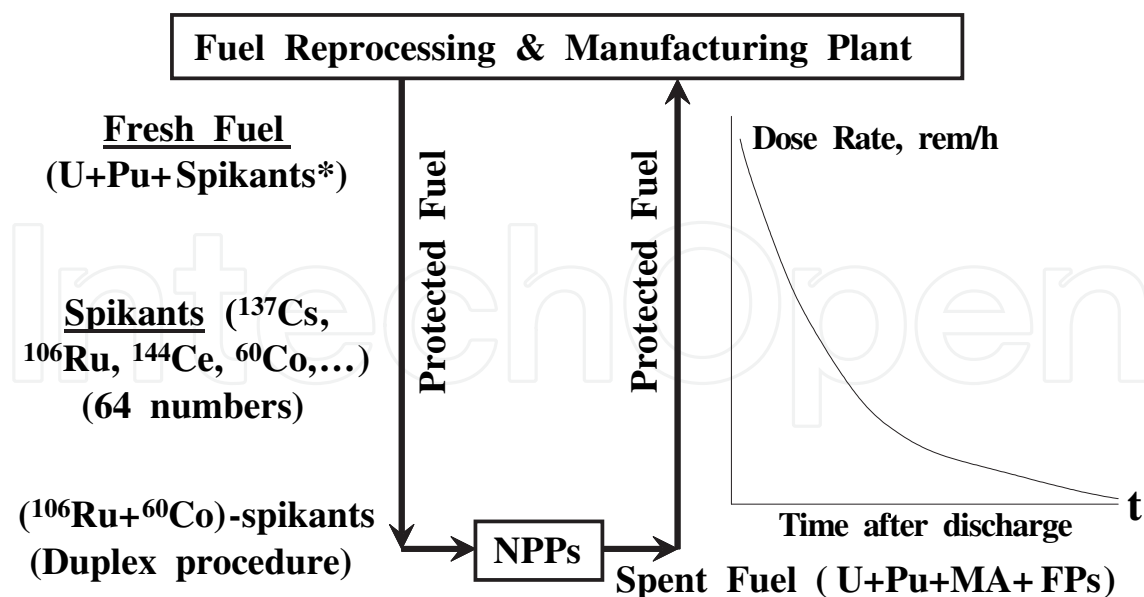Fig. 12. Closed (U-Pu)-fule cycle protected (ORNL, USA)*

Radionuclides ^{137}Cs ($T_{1/2} = 30$ years) and ^{60}Co ($T_{1/2} = 5.27$ years) appeared the most preferable candidates. But cesium is a volatile element, and it can be easily removed from fuel by heating up. Intensity of γ-radiation emitted by ^{60}Co rapidly relaxes.

Specialists from LANL (USA) proposed the advanced version of the international NFC that enhances proliferation resistance of plutonium (Cunningham et al., 1997). This proposal constituted a basis for the US President's initiative on the Global Nuclear Energy Partnership (GNEP) that was supported by many countries (including Russia) with well-developed nuclear technologies (see Fig. 13).

According to the proposal, spent fuel assemblies discharged from power reactors of a country-user must be transported to the Nuclear Club countries for full-scale reprocessing. Extracted plutonium and minor actinides must be incinerated in the reactors placed on the territory of the International nuclear technology centers. Plutonium is not recycled in power reactors of a country-user. The Nuclear Club countries provide fresh LEU fuel deliveries into a country-user.
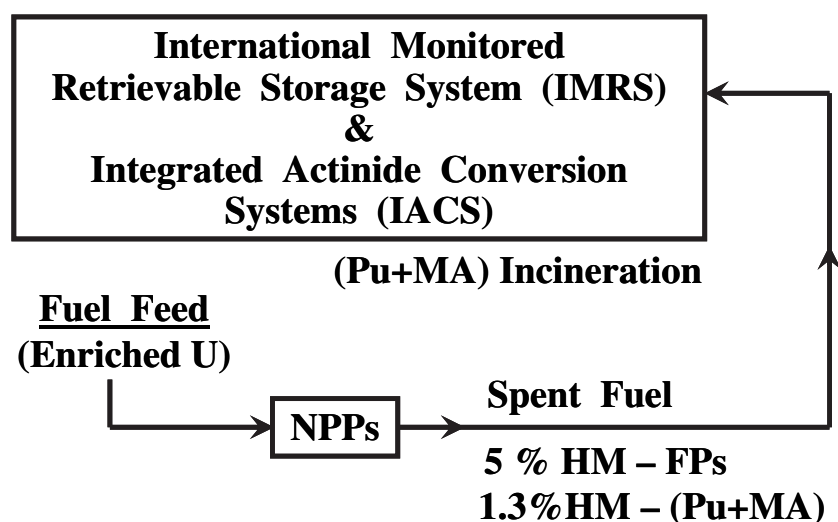

Fig. 13. Open fuel cycle protected (LANL, USA)

Upon exhaustion of rich and cheap uranium resources, nuclear power has to use artificial kinds of fresh fuel (plutonium, ^{233}U or their mixtures). The GNEP initiative does not consider this opportunity. It is proposed to use such power reactors which are able to work without refueling for 15-20 years. After this time interval they must be returned to the Nuclear Club countries for SNF discharging and reprocessing and for insertion of fresh fuel. The concentrated incineration of plutonium and minor actinides in the International nuclear technology centers can lead to unacceptably large local release of thermal energy with unpredictable negative environmental and climatic effects. As for reactors with long-life cores, these are small and medium-sized power reactors. Besides, during transportation and mounting, they can be very attractive sources of plutonium in amounts large enough for manufacturing of several dozens of nuclear bombs.

6.2 Enhancement of LWR MOX-fuel cycle proliferation resistance by plutonium denaturing

Some nuclear properties of ^{238}Pu make this isotope a valuable material for proliferation protection of uranium-plutonium fuel. Firstly, ^{238}Pu is an intense source of thermal energy ($T_{1/2} = 87$ years, specific heat generation - 570 W/kg). So, introduction of ^{238}Pu into plutonium creates almost insuperable barrier to manufacturing of even primitive implosion-type NED. Plutonium heating up by isotope ^{238}Pu can provoke undesirable phase transitions

and thermal pyrolysis of conventional explosives applied for compression of central plutonium charge. Secondly, ^{238}Pu is an intense source of spontaneous fission neutrons, even more intense than ^{240}Pu. As a consequence, probability of premature CFR initiation in NED sharply increases while energy yield of nuclear explosion drastically drops down to the levels comparable with energy yield of conventional explosives. Thus, LWR MOX-fuel cycle with ternary fuel compositions (Np-U-Pu) is characterized by enhanced proliferation resistance.

Like uranium, plutonium can be isotopically denatured by two ways: either direct introduction of intensely radioactive isotope ^{238}Pu into MOX-fuel composition or introduction of relatively low intense radioactive isotope ^{237}Np into MOX-fuel composition. ^{237}Np is the nearest neutron predecessor of main denaturing isotope ^{238}Pu. So, only short-term pre-irradiation of fresh MOX-fuel assemblies would be sufficient to produce proliferation resistant fuel assemblies, suitable even for export deliveries to any countries.

6.2.1 The effect of ^{237}Np and ^{238}Pu introduction on Pu protection in LWR fuel

It is proposed that the equilibrium isotope vectors are obtained for MOX-fuel circulating between LWR, spent fuel reprocessing as fuel manufacturing facilities. The fuel feed includes isotopes ^{237}Np, ^{238}Pu and ^{239}Pu is produced in Hybrid Thermonuclear Installation (HTI) blankets.

Using the code GETERA (Belousov et al., 1992) for cell calculations of fuel burn-up, Pu isotopic compositions of MOX-fueled PWR were determined for moments of the beginning and end of cycle. ^{238}Pu fraction in plutonium was adopted to be an index of Pu protection against uncontrolled proliferation. It means that the impact of higher plutonium isotopes on neutronics of chain reaction in imploded plutonium charge of NED was not taken into account.

The fuel being loaded in PWR may be considered as material consisting of two parts: the first part includes equilibrium composition of ^{238}U and plutonium isotopes produced by ^{238}U while the second part ("feed part of fuel") includes equilibrium composition of ^{237}Np, ^{238}Pu and other plutonium isotopes produced entirely by the feed. Equilibrium contents of ^{238}Pu in plutonium of PWR fuel depending on ^{238}Pu contents in plutonium of feed (with different ^{237}Np fractions in "feed part of fuel") for equilibrium multi-cycle operation regime are presented in Fig. 14.

The plot region situated under the bisectrix B is a region where plutonium protection in feed is higher than plutonium protection in fuel. Respectively, the plot region situated above the bisectrix B is a region where plutonium protection in fuel is higher than that in feed. The curves of this figure characterize the correlation between plutonium protection levels in feed and fuel when the "feed part of fuel" contains ^{237}Np in addition to plutonium. Basing on these data, it is possible to select the appropriate equilibrium regime of NFC.

Proper selection of the feed compositions, i.e. fractions of ^{238}Pu and ^{237}Np, makes it possible to attain the same level of fuel plutonium protection for various combinations of ^{238}Pu and ^{237}Np content in feed. For example, 32%-level of fuel plutonium protection can be attained in case of feed containing (0% ^{237}Np, 52% ^{238}Pu) or (20% ^{237}Np, 43% ^{238}Pu) or (40% ^{237}Np, 32% ^{238}Pu). The latter option corresponds to equal level of plutonium protection both in fuel and in feed. The line "S" that connects the right ends of the curves shown in Fig. 14 may be regarded as an "ultimate option" of the (Np-U-Pu) NFC considered here. The points of this line correspond to particular option of the (Np-U-Pu) NFC where ^{238}U is absent in fuel composition, and its fertile functions passed to ^{238}Pu and ^{237}Np. So, this NFC may be called as a (Np-Pu) NFC. In this NFC the highest fuel Pu protection level (65% ^{238}Pu) can be

reached with feed Pu protection of 90% ^{238}Pu. As known, the IAEA safeguards are not applied to plutonium containing 80% ^{238}Pu or more (Rolland-Piegue, 1995; Willrich & Taylor, 1974; Massey & Schneider, 1982).
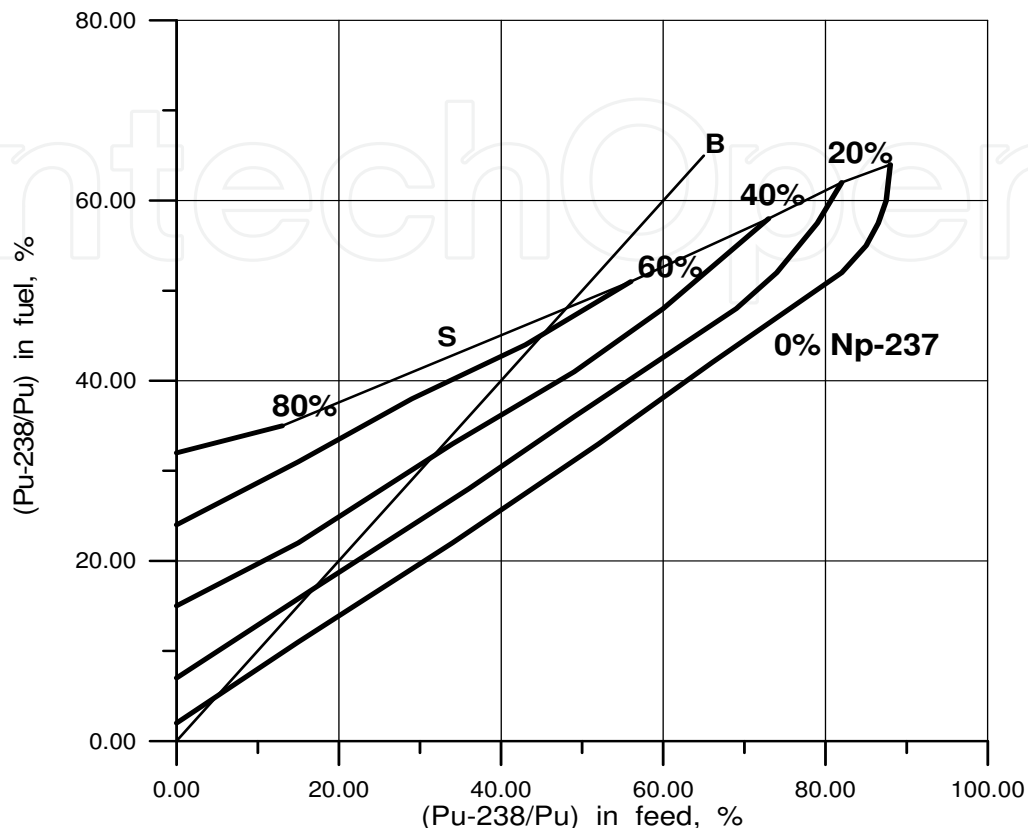

Fig. 14. Proliferation resistance of plutonium in fuel as function of proliferation resistance of plutonium in feed and ^{237}Np content in "feed" part of fuel. B - bisectrix.

Inherent heat generation of plutonium is considered as a significant factor of its protection. The rates of inherent heat generation for various feed compositions are presented in Table 4. Here, the rates of specific heat generation for weapons-grade plutonium (WGPu) and reactor-grade plutonium (RGPu) are presented as well.

| | | | ^{238}Pu/Pu in fuel and in feed | | |
			(Np/(Np + Pu) in feed)		
Generation	WG Pu	RG Pu	17% (7%)	33% (15%)	44% (19%)
q^{Pu}, W/kg Pu	2.3	13.	97	186	248
n_{sf}^{Pu}, 10^6(n/sec)/kg Pu	0.06	0.38	0.71	1.06	1.30
q^{fuel}, W/kg fuel	---	---	14.9	41.2	99.5
n_{sf}^{fuel}, 10^6(n/sec)/kg fuel	---	---	0.11	0.24	0.53
Feed ^{237}Np/^{238}Pu/^{239}Pu, kg/(GWe*a)	---	---	38 / 82 / 402	103 / 194 / 377	176 / 318 / 421

Table 4. Decay heat generation (q^{Pu}) and neutron generation by spontaneous fissions (n_{sf}^{Pu}) in LWR fuel with equal plutonium protection both in fuel and in feed.

Basing on the results shown above, it can be concluded that denatured fuel plutonium containing more than 25% [238]Pu is characterized by the internal heat generation which exceeds that of RGPu by more than order of magnitude and, by the larger extent, that of WGPu. In addition, denatured fuel plutonium is characterized by the higher neutron background caused by spontaneous fissions. The factors mentioned above enhance plutonium protection against its utilization in NED. The same factors complicate, to certain degree, the handling procedures with such a fuel in nuclear technologies.

Values of specific heat generation and neutron emission due to spontaneous fission of MOX-fuel being loaded for the equilibrium cycle options analyzed are shown in Table 4 also. For comparison, "dry" technology for handling with spent fuel assemblies may be applied if specific heat generation does not exceed 20-35 W/kg fuel. It may be also concluded that plutonium denaturing with [238]Pu is restricted by thermal constraints imposed on permissible specific heat generation of fuel. The same tendency exists in connection with spontaneous neutrons emission. These constraints need to be taken into account in fuel fabrication, fuel rods and fuel assemblies manufacturing and transport operations. These complications of fuel management may be considered as certain "payment" for proliferation resistance of MOX-fuel cycle.

Actually speaking, the protection of plutonium in (Np-U-Pu)-fuel cycle is supposed to be enhanced due to addition [237]Np and [238]Pu into fuel. The degree of fissile nuclides protection depends mainly on magnitude of [238]Pu fraction in plutonium. Meanwhile, [237]Np itself can be also considered as a potential material for NED. For example, critical mass of [237]Np (metal sphere, steel reflector) is about 55 kg (Koch et al., 1997). It's ten times more than that of [239]Pu. The magnitude of critical mass of [237]Np is sensitive with respect of its dilution. For example, minimum critical mass of NpO_2 is as much as 315 kg (Nojiri & Fukasaku, 1997; Ivanov et al. 1997). Besides, in fuel composition [237]Np is present together with plutonium which is characterized by essential neutron source strength due to spontaneous fissions. Therefore, in order to apply extracted [237]Np in NED it is needed to perform effective [237]Np purification from plutonium (plutonium fraction is restricted by value of 10^{-4} - 10^{-3}).

6.3 Increase of fuel burn-up in denatured (Np-U-Pu) fuel cycle

Good neutron-multiplying properties of [238]Pu and its neutron predecessor [237]Np make it possible to extend substantially time period for continuous reactor operation without refuelings. As a consequence, unauthorized extraction of plutonium from SNF becomes practically unfeasible.

Indeed, under reactor irradiation of (Np-U-Pu) fuel it is occurs the following "non-traditional" transition chain (see Fig. 15): [237]Np → [238]Pu → [239]Pu →... A successive transition of these nuclides leads to enhancement of multiplication properties.

Actually, as it can be seen in Fig. 16, excess neutron generation per one absorption (v_{eff}-1) in [237]Np is negative for neutrons of all energy range (excepting fast neutrons), positive for neutrons with E_n > 1 KeV for [238]Pu and, as is known, essential positive one for [239]Pu.

So, for (Np-U-Pu)-fuel the nuclides we are dealing with can be characterized as follows (Table 5).

At the same time, during irradiation in reactor core FP accumulation results in growth of neutron absorption. So, these tendencies can be counterbalanced and such fuel will be characterized by stabilized neutron-multiplying properties over long burning-up.

Burn-up calculations for mono-nitride fuel in cell of PWR-type reactor with heavy water as a coolant were performed by using code GETERA. The cell parameters were similar to that of VVER-1000 cell (see Table 6):
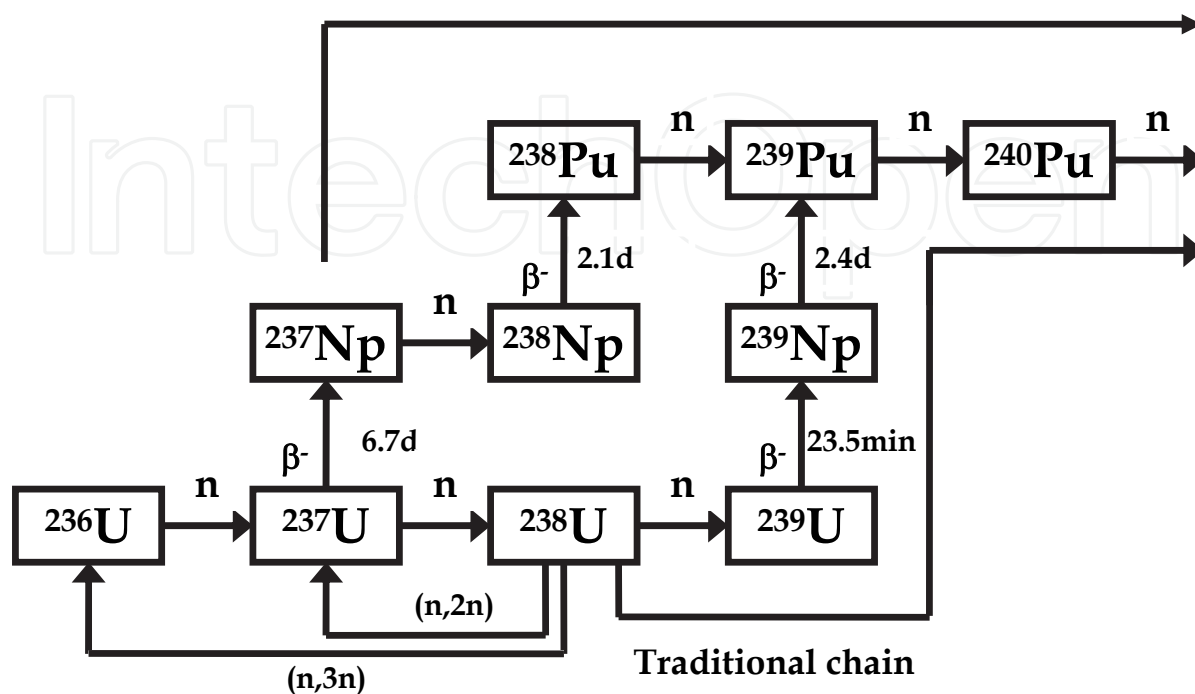

Fig. 15. Chain of isotopic transformations in uranium-plutonium fuel cycle

^{237}Np	^{238}Pu	^{239}Pu
"Burnable poison" nuclide	Moderate fissile nuclide ($E_n > 1$ KeV)	Well-known fissile nuclide

Table 5. Characteristics of nuclides for (Np-U-Pu)-fuel

Fuel rod diameter	9.1 mm
Thickness of stainless steel cladding	0.4 mm
Coolant (heavy water)	D_2O
Water volume / fuel volume	1.6
Fuel	Mono-nitride (porosity - 30%)
Specific heat generation	110 kW/l

Table 6. Cell parameters of PWR-type reactor

In Fig. 17 it is shown the dependence of K_∞ on fuel burn-up for various fuel compositions. For comparison it is demonstrated also a curve of K_∞ for LWR-UOX. It can be seen that, actually, there is possibility to attain fuel burn-up of 25-30%HM (corresponding residence time is about 20-25 years.). It is worth-while mentioning that, according to papers (Ivanov et al. 1997; Bychkov et al. 1997) presented at the International Conference "GLOBAL'97", vibro-packed MOX fuel in stainless steel cladding was irradiated in fast reactor BOR-60 (Russia) and it was obtained burn-up of 26% HM on standard fuel assemblies and burn-up

of 32% HM in experimental fuel rods. No thermal-mechanical and physical-chemical fuel-cladding interaction was observed in any of the analyzed cross-sections.
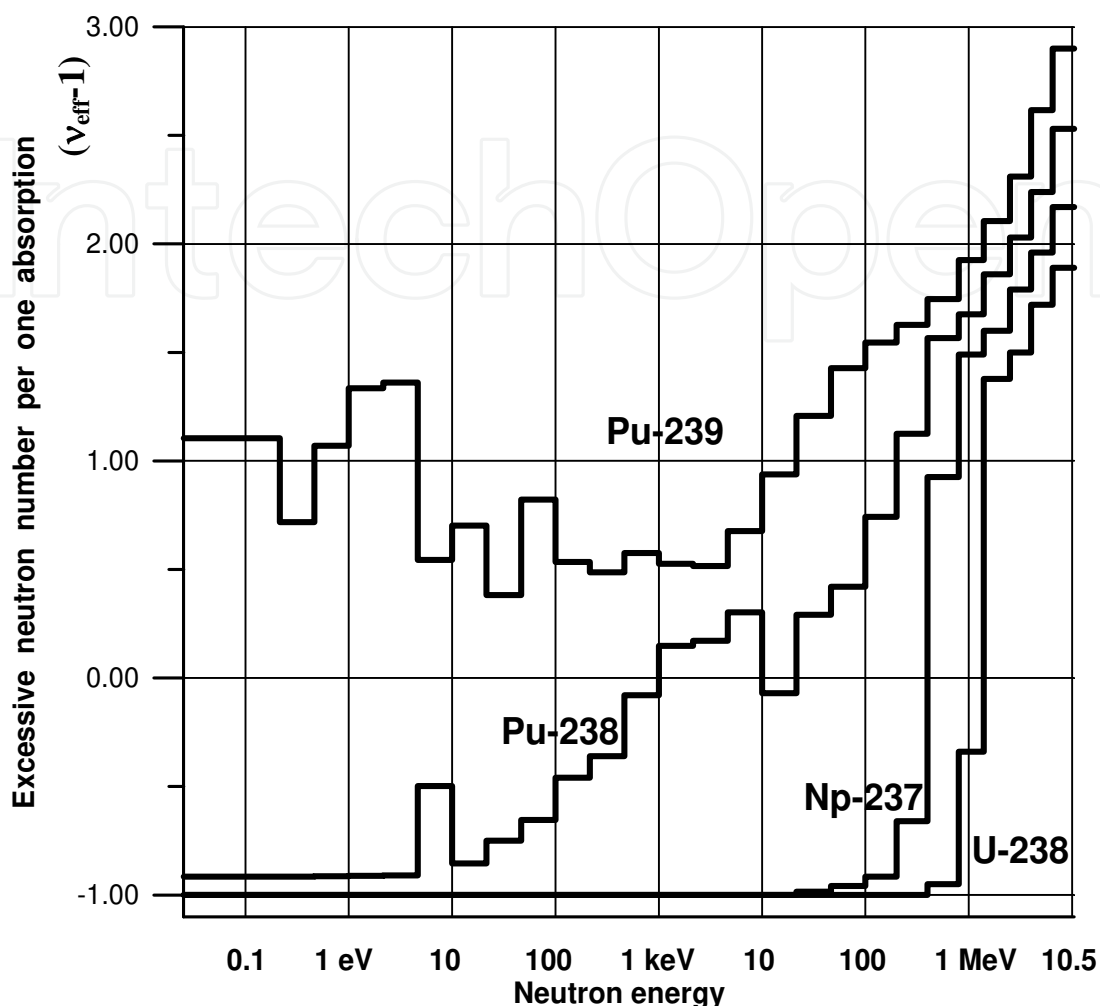

Fig. 16. Dependencies of excessive neutron number per one absorption (ν_{eff}-1) on neutron energy for nuclides of uranium-plutonium fuel cycle

The results mentioned above referred to so-called "ultimate" fuel compositions which didn't contain [238]U. Actually speaking, these results can be considered as preliminary ones to demonstrate scale of benefit. Undoubtedly, it is needed to analyze impact of wide fuel compositions (including [238]U) on stabilized multiplication properties of ultra long-life cores taking into consideration reactor safety in both critical and sub-critical regime of operations.

Anyway, application of ultra long-life core concepts will lead to essential decrease of SNF flow rate, reduction of reprocessing, remanufacturing and shipping operations. It's a factor for internationalization of Nuclear Energy System fuel cycle. Since fuel cycles been discussed are "rich" with respect to excess neutron generation in CFR, there is no necessity to perform fine purification of fuel being reprocessed. It's a factor of enhancement of the fuel cycles protection.

Application of NPP with ultra long-life core concepts is expected to be profitable for electricity generation in developing countries which have not improved nuclear technology infrastructure.
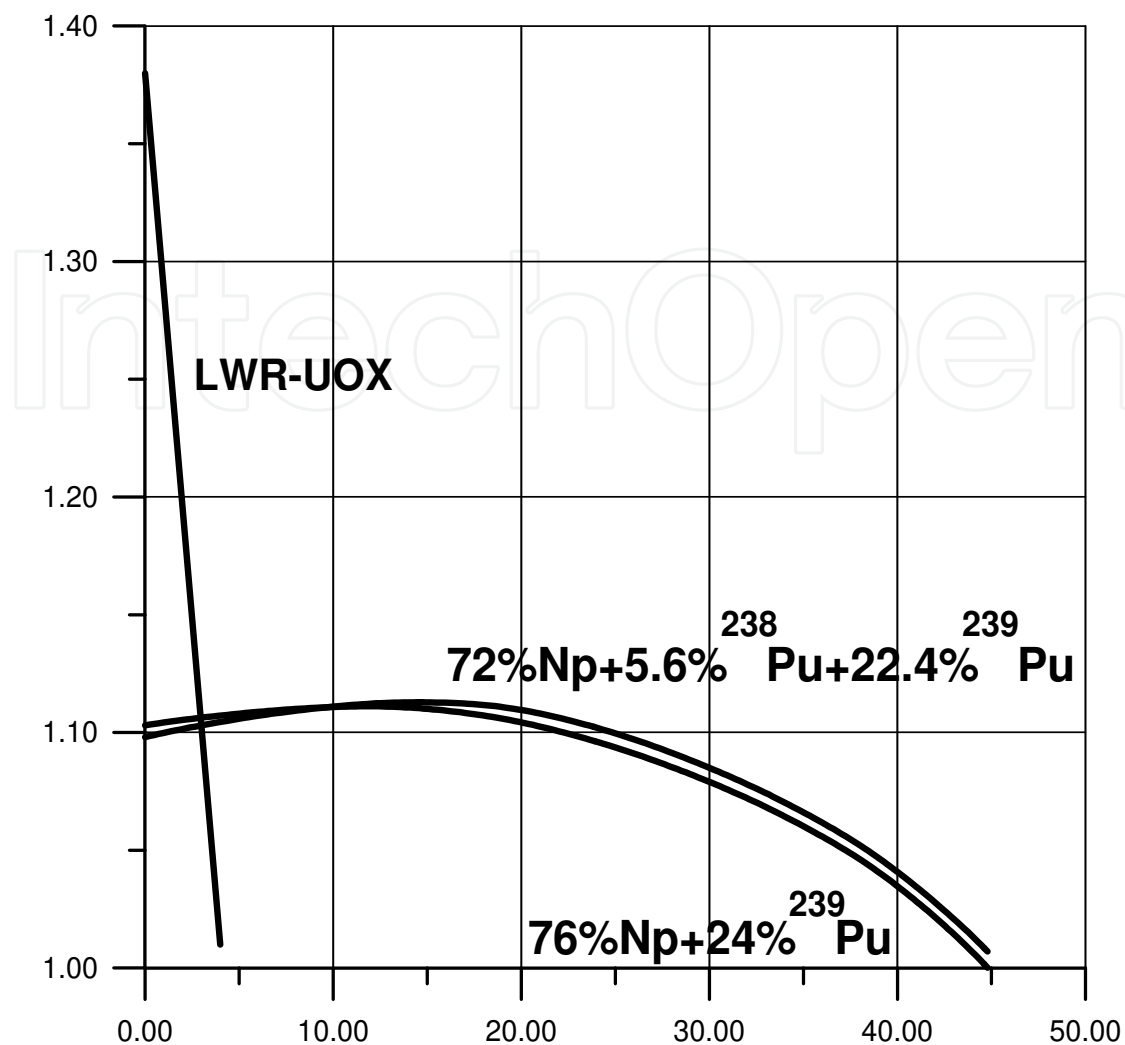
Fig. 17. Dependencies of K_∞ on fuel burn-up for various fuel compositions

7. Mixed (Th – U - Pu) fuel cycle

Plutonium has no its own "fertile" isotope. So, it is impossible to protect plutonium by isotopic dilution, like uranium. Upon exhaustion of cheap ^{235}U resources, the isotope dilution principle can be applied to ^{233}U-^{238}U mixture. So, it seems reasonable to consider the following proliferation resistant fuel - (^{232}Th-^{233}U-^{238}U) [23]. If ^{238}U content is small but sufficient for low content of ^{233}U in uranium fraction, then plutonium build-up may be suppressed.

In other words, the mixed (^{232}Th-^{233}U-^{238}U-Pu) fuel cycle should be studied along with "classical" (^{232}Th-^{233}U) and (^{238}U-Pu) cycles. In both "classical" cycles, fissile materials (^{233}U or Pu) may be figuratively called by "highly-enriched" fuel. In the mixed cycle, on the contrary, fissile isotope ^{233}U is diluted with ^{238}U in uranium fraction, and thus (^{233}U-^{238}U) mixture may be regarded as a "low-enriched" fuel. It is noteworthy that homogeneous mixture of two fertile isotopes ^{238}U and ^{232}Th is a more effective neutron absorber than both separate isotopes. This effect can improve neutron-physical properties of the mixed fuel because it can increase fuel burn-up and thus reduce flow rate of spent fuel assemblies for reprocessing (Kulikov, 2007).

In the mixed fuel cycle, the following double-strata structure may be estimated as an effective and proliferation resistant option (Figs. 18, 19): the top stratum includes full-scale reprocessing of spent fuel assemblies in the International nuclear technology centers with complete incineration of plutonium and minor actinides, the bottom stratum includes a simplified thermal-chemical (DUPIC-type) re-fabrication of fresh fuel with feeding by proliferation resistant ^{233}U. Such a closed nuclear fuel cycle may be equally effective in power reactors of PWR and CANDU types.

So, if fuel contains homogeneous mixture of two fertile isotopes ^{238}U and ^{232}Th, the following new qualities do appear:

- Fissile isotope ^{233}U produced in neutron irradiation of thorium is diluted with fertile isotope ^{238}U. So, ^{233}U-^{238}U mixture represents, in essence, a kind of "low-enriched" uranium.
- Reduced content of ^{238}U suppresses build-up rate of plutonium.
- Mixed fuel is highly effective not only in thermal but in resonant neutron spectrum too because fissile isotope ^{233}U has sufficiently good neutron-multiplying properties both in thermal and resonant neutron spectra.
- Fissile isotope ^{239}Pu converts rapidly into heavier plutonium isotopes with low neutron-multiplying properties because of larger $\alpha = \sigma_c/\sigma_f$. So, plutonium loses its attractiveness as a material suitable for NED manufacturing.

As is known (Benedict et al., 1981), fissile isotope ^{233}U can be additionally protected by its denaturing with ^{232}U because this isotope has the following proliferation-resistance properties (Fig. 19):

1. ^{232}U is an intense source of high-energy γ-radiation emitted by its decay products.
2. ^{232}U is an intense source of spontaneous neutrons, i.e. spontaneous fission neutrons plus neutrons from (α,n)-reactions with light impurities.
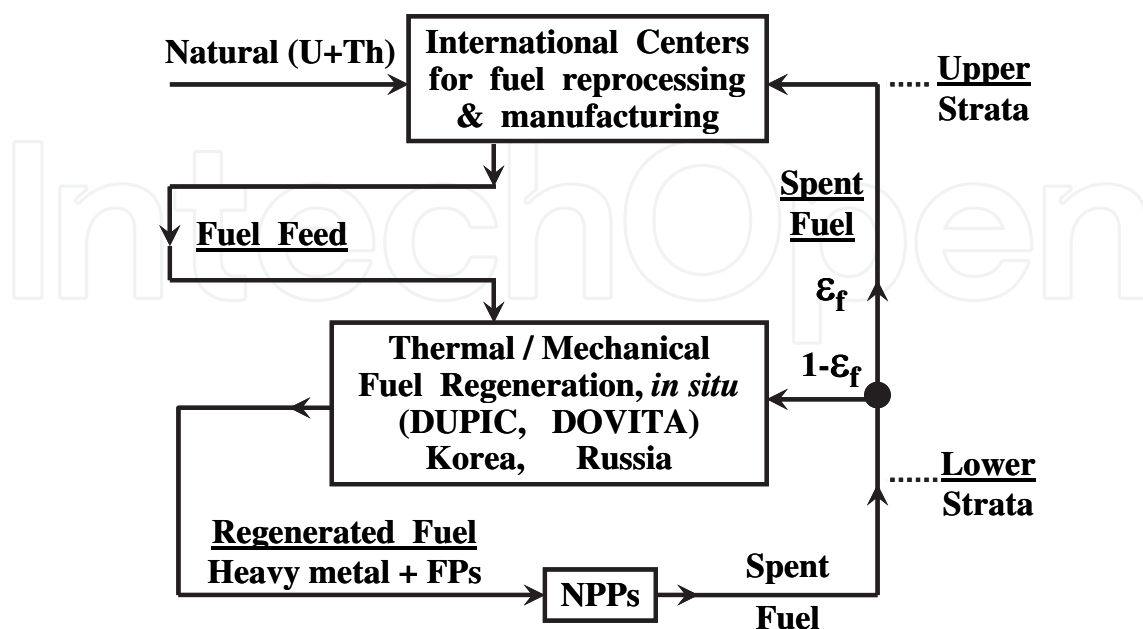3. ^{232}U is an intense heat source from its own α-decays and from decays of its daughter products.

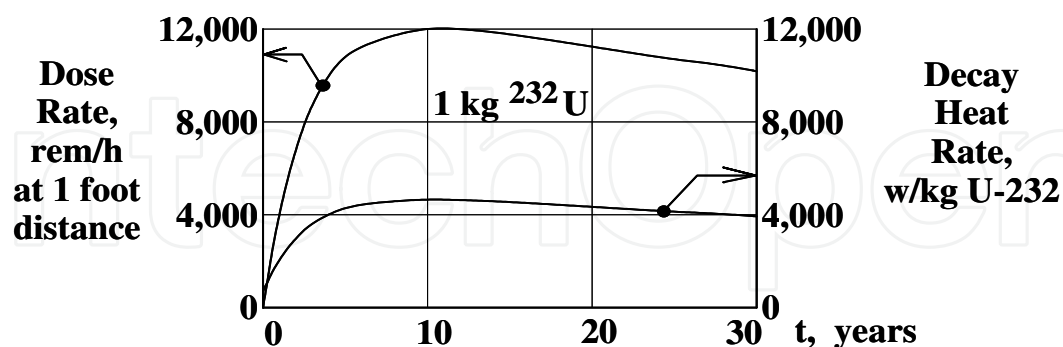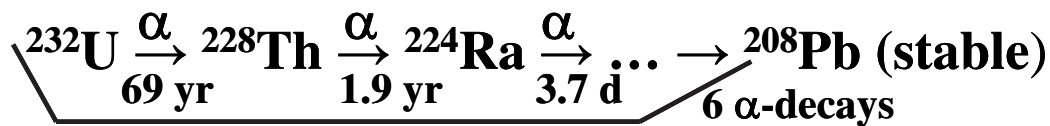Fig. 18. Double-Strata closed fuel cycle protected

$$^{232}U \xrightarrow[69 \text{ yr}]{\alpha} {}^{228}Th \xrightarrow[1.9 \text{ yr}]{\alpha} {}^{224}Ra \xrightarrow[3.7 \text{ d}]{\alpha} \dots \rightarrow {}^{208}Pb \text{ (stable)}$$

$$6 \text{ } \alpha\text{-decays}$$

Fig. 19. ^{232}U as a Spikant

Q_{sf} (Spontaneous Fission Neutrons) = $1.3 \cdot 10^3$ n/(s·kg ^{232}U);
$Q_{\alpha,n}$ (Uranium Dioxide) = $15 \cdot 10^6$ n/(s·kg ^{232}U) (·20 – equilibrium);
^{232}U–leader among U isotopes as a spontaneous neutrons generator.

7.1 Proliferation protection of multi-isotope fuel containing uranium generate and protactinium-uranium mixture produced by Hybrid Fusion Facility

Neutron irradiation of natural thorium in blanket region of Hybrid Fusion Facility (HFF) based on (D,T)-plasma can produce many thorium, protactinium and uranium isotopes. High-energy (14 MeV) thermonuclear neutrons are able to initiate some threshold (n,xn)-reactions leading to intense generation of ^{230}Th, ^{231}Pa, ^{232}U, ^{233}U and ^{234}U. The longer irradiation time, the larger content of these isotopes in irradiated thorium. Content of ^{232}U, for example, can reach a value of several percents.

NFC closure and SNF reprocessing can release huge amounts of fissionable materials: about 210 000 tons of uranium regenerate, RGPu and minor actinides, where uranium regenerate is a dominant fraction. Uranium regenerate may be regarded as a fertile material suitable for further use by nuclear power industry. Uranium regenerate will be released in the amounts large enough to feed NPP of total electric power at the level of 1500 GWe, i.e. 4 times higher that total power of global nuclear energy system today.

Uranium regenerate contains the following isotopes: ^{232}U, ^{233}U, ^{234}U (minor fraction) and ^{235}U, ^{236}U, ^{238}U (main fraction). Uranium produced in thorium blanket of HFF contains only isotopes of minor fraction, i.e. ^{232}U, ^{233}U and ^{234}U. So, if HFF-produced uranium is admixed to uranium regenerate, content of only minor fraction increases. Content of minor fraction can be made comparable with content of main fraction. In the extreme case, minor fraction becomes a dominant one, and NFC shifts towards ^{233}U-based fuel.

Thus, uranium fraction of nuclear fuel represents a mixture of practically all significant uranium isotopes: ^{232}U, ^{233}U, ^{234}U, ^{235}U, ^{236}U, ^{238}U. The following three aspects should be noted. Firstly, main fissile isotopes, ^{233}U and ^{235}U, are accompanied by lighter and heavier uranium isotopes, essential neutron absorbers. Secondly, if ^{232}Th and ^{231}Pa are introduced into fuel composition replacing partially uranium regenerate, then plutonium generation rate is suppressed. Thirdly, the presence of ^{236}U in fuel composition can initiate the chain of isotopic transformations leading to accumulation of ^{232}U, ^{233}U, ^{238}Pu, main isotope for plutonium denaturing (De Volpi, 1982):

^{236}U(n,γ)^{237}U(β⁻, $T_{1/2}$ = 7 days)^{237}Np (n,γ)^{238}Np (β⁻, $T_{1/2}$ = 2.1 days)^{238}Pu

So, produced plutonium will contain not only ^{240}Pu, usually accompanying isotope to ^{239}Pu in power reactors, but ^{238}Pu too.

In mixed (Th-U-Pu) fuel cycle, plutonium plays an auxiliary role only while ^{233}U is a main fissile isotope, and plutonium content in fuel composition may be diminished. Finally, plutonium could be removed from global nuclear energy system for peaceful utilization in the dedicated nuclear power facilities. The GNEP initiative advanced by the US President (Sokolova, 2008) foresees just a similar option. This aspect represents a special significance from the standpoint of plutonium protection against unauthorized diversion to non-energy purposes (Mark, 1993).

Uranium fraction consisting of practically all significant uranium isotopes from ^{232}U to ^{238}U is, in essence, low-enriched uranium with rather small content of main fissile isotopes (^{233}U and ^{235}U). Isotopic enrichment of such a multi-isotope composition will be a very difficult problem for potential proliferators in the case of its unauthorized diversion.

The presence of α-emitters (mainly, ^{232}U, ^{233}U and ^{234}U) in uranium fraction can initiate physical and chemical processes leading to α-radiolysis of uranium hexafluoride including molecular dissociation with generation of minor fluorides, exchange reactions of recombination and coagulation. These processes can provoke serious violations in the correspondence between the order in masses of uranium isotopes and the order in masses of uranium hexafluoride molecules. This correspondence is a necessary condition for successful uranium enrichment.

So, closed mixed (^{233}U-^{232}Th-^{238}U) fuel cycle can offer the following advantages in comparison with "classical" (^{238}U-Pu) and (^{232}Th-^{233}U) cycles:

- Fissile isotope ^{233}U is diluted by fertile isotope ^{238}U in uranium fraction of fuel composition.
- ^{238}U content in fuel composition may be diminished thus suppressing plutonium production. As a consequence, load of the International centers on plutonium utilization may be reduced.

General conclusion can be defined as follows: fuel of mixed (Th-U-Pu) cycle contains fissile isotopes with upgraded level of their protection against any unauthorized attempts of their diversion to non-energy purposes.

8. Probability analysis of risk reduction in non-energy applications of denatured uranium

Proliferation protection of uranium and uranium-plutonium fuel can be quantitatively evaluated within the frames of the concept developed for risk assessment in authorized applications of nuclear materials. The concept includes some relationships which can be used to evaluate probability for a certain chain of unauthorized actions (UAA) to occur and to evaluate damage from potential NED applications.

8.1 Scenarios for UAA with nuclear materials and models for UAA detection

One of main directions in nuclear non-proliferation ensuring is a formation of inaccessibility conditions for NM against any UAA. This is a main strategic function of MPC&A system at any nuclear-dangerous objects. However, the following questions arise:

1. What can occur with nuclear materials, if these conditions are violated due to some kind of reasons?

2. How can we estimate the threats?
3. What must we do under these accidental conditions? Answers to the questions are related to the threats of NM diversion including the threat of NED manufacturing from diverted NM and its military application. In order to give a correct response to these questions, two, at least, conditions must be satisfied:

- We must know how to evaluate the threats of NED manufacturing from diverted NM and their military applications.
- We must work out the recommendations on effective countermeasures to be undertaken against any UAA.

An important condition for successful counteraction against the use of diverted NM in NED manufacturing consists in development of the control system over illegal NM trafficking. External UAA monitoring system can apply various strategies of the searching process for potential UAA objects.

Unlike authorized activity, unauthorized actions with NM can be characterized by the following specific features:

- Secrecy of unauthorized works. The secrecy level is defined by NM properties and financial expenses to be paid by potential proliferators.
- Striving for manufacturing of NED with maximal destructive capability.
- Striving for maximal shortening of UAA time which follows from the fact that potential proliferator understands properly the threats from external UAA monitoring system.

These tendencies are the conflicting ones from position of potential proliferator who strives to reach his ultimate purpose. For example, proliferator strives for NED manufacturing with maximal destructive capability but this requires application of sophisticated nuclear technologies for processing of diverted NM. In their turn, nuclear technologies require large financial and long time expenses with appropriate reduction of the secrecy level and rising of the detection probability.

So, when analyzing various scenarios of NM diversion, we presumed a rational behavior of nuclear proliferators, i.e. the proliferator has to accept a certain compromise between his striving for manufacturing of NED with maximal destructive capability and rising of the detection probability caused by application of sophisticated nuclear technologies. In any case rather long chain of technological processes is required to manufacture NED from diverted NM.

8.2 Concept of risk of NM applications in destructive purposes

Potential risk of NM application for NED manufacturing and military use by terrorist groups can be evaluated as follows: $R = P \cdot D$, where P – probability of NED manufacturing and military use; D – potential damage from the use of NED for destructive purposes.

Probability P depends on proliferator capabilities, initial and final NM states. The probability may be written in the following form: $P = P(F, S_I \rightarrow S_F)$, where F – proliferator capabilities (his material and financial funds, available technological basis); S_I – initial NM state (mass, physical form, chemical composition, radioactivity, local position, etc); S_F – final NM state (design of NED, local position, chemical and isotopic compositions, radioactivity, etc). Potential damage D depends on final NM state only, i.e. $D = D(S_F)$.

Assumption on a rational behavior of nuclear proliferator enables us to think that proliferator will follow the well-grounded plan with proper accounting for the detection probability, if sophisticated nuclear technologies are applied for processing of diverted NM

(for example, fine NM purification with removal of all significant impurities, isotopic re-enrichment and so on). So, the risk of NED manufacturing and military use can reach a maximal point either within or on the boundaries of the domain that includes all potential UAA undertaken by nuclear proliferators. The maximal risk and its location in UAA domain depends on the level of external UAA monitoring and on financial capabilities of nuclear proliferators (see Fig. 20).
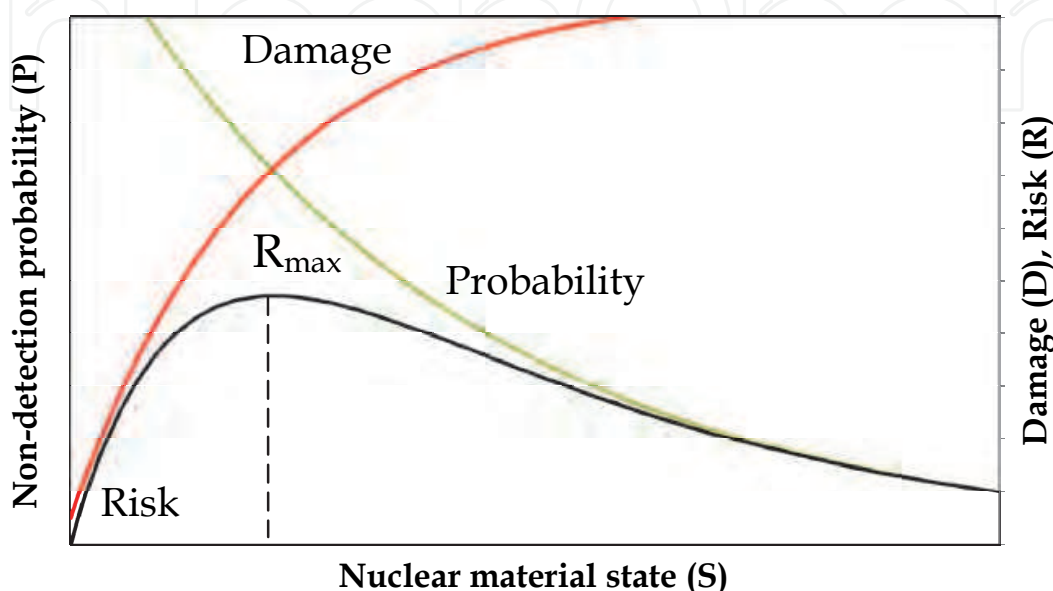

Fig. 20. Variations of the risk related with NM application in destructive purposes when sophisticated nuclear technologies are involved into NM processing

This circumstance can be used to simplify analysis by using a conservative approach to evaluating the maximal risk of NM usage for NED manufacturing. Within the frames of this approach, probability P for successful completion of UAA chain (from initial state S_I to final state S_F) can be replaced by the following maximal evaluation:

$$R_{max}(F,S_I)=\max_{sf}\left(P(F,S_I \rightarrow S_F)\cdot D(S_F)\right) \tag{1}$$

8.3 Probability to avoid UAA detection

The following problem is considered below: it is required to search for UAA object which was created on a certain territory. Let's consider a discrete limited set N consisting of n components each of them may be checked up in one identification step. If the set N contains a closed limited subset S that includes s components and characterizes dimensions of UAA object from the viewpoint of the identification process, then probability for successful identification of any component belonging to the subset S is equal to $P_{det} = s/\text{n}$. Naturally, non-detection probability per one identification step is equal to $P_{undet} = 1 - P_{det} = 1 - s/n$.

Let's assume that UAA object is not moved and UAA can be unambiguously detected by one identification procedure. If the identification rate $V = dn/dt$ is a constant value, and UAA object is a sufficiently concealed object, i.e. $s \ll n$, then time dependency of non-detection probability may be presented as follows:

$$P_{undet}(t) = 1 - \frac{s}{n} V \cdot t = 1 - \lambda \cdot t \tag{2}$$

where $\lambda = s / n \cdot V$, parameter of successful detection, is a product of two multipliers, one of them depends on properties of UAA object only.

If UAA can not be detected for one identification step, or if UAA object moves during the identification process, then a necessity arises to perform a repeat examination of the regions which were checked up previously. In this case, time dependency of non-detection probability may be written in the following form:

$$P_{undet}(t) = e^{-\frac{s}{n} V \cdot t} = e^{-\lambda \cdot t} \tag{3}$$

8.4 UAA chains. Indicators of the searching process for UAA objects

The following main links can be identified in UAA chains resulting in NED manufacturing from diverted uranium-containing NM: NM theft → chemical and physical reprocessing → isotopic re-enrichment → manufacturing of main NED components → military use of NED. Each link of UAA chain is defined by its duration t_i and mean time interval needed to detect the proliferator $1/\lambda_i$, which are the functions of the proliferator capability F, changes of NM properties $(S_{i-1} \rightarrow S_i)$ and efficiency of the searching process. In general case, detection probability is described by exponential function. So, probability P_i for successful completion of the i-th link without detection and suppression can be written in the following form:

$$P_i = P_{undet,i} \cdot P_{unsup,i} = e^{-\lambda_i t_i} \cdot P_{unsup,i}(t_i) \tag{4}$$

where $P_{undet,i}$ – non-detection probability of diverted NM at the i-th link; $P_{unsup,i}$ – non-suppression probability for UAA performed by detected proliferator at the i-th link. So, risk of NED manufacturing and military use is defined by the following equation:

$$R = D \cdot \prod_i P_{unsup,i}(t_i) \cdot e^{-\lambda_i t_i} = D \cdot P_{unsup} \cdot e^{-\sum_i \lambda_i t_i} \tag{5}$$

UAA object can be detected from the really existing indicators including the indicators related with consumption of energy and water resources in the unauthorized activity. The following indicators can be used in the search for UAA aimed at NED manufacturing and military use:

- Emission rate (A).
- Resource consumption rate (W).
- Capital expenses (K).

When searching the UAA-object being to several independent indicators, then total non-detection probability is a product of partial non-detection probabilities for different UAA indicators, i.e.

$$P_{undet}(t) = P_{undet}^K(t) \cdot P_{undet}^W(t) \cdot P_{undet}^A(t) = e^{-\lambda_K t} \cdot e^{-\lambda_W t} \cdot e^{-\lambda_A t} = e^{-(\lambda_K + \lambda_W + \lambda_A)t} = e^{-\lambda t}, \tag{6}$$

where

$$\lambda = \lambda_K + \lambda_W + \lambda_A = \alpha_K \cdot f(K) + \alpha_W \cdot f(W) + \alpha_A \cdot f(A), \tag{7}$$

where α – efficiency of the searching process for appropriate UAA indicators.

Relationship between UAA indicators and detection parameters can be derived from the following models for strategic behavior of nuclear proliferator:

1. The proliferator creates a new infrastructure for his unauthorized activity. According to equation (2), UAA detection parameter in the random searching process for new resources is proportional to the scale of new resources which were put in operation. In the simplest case, the scale is defined by the resource consumption rate W and capital expenses K. So, in this case: $\lambda_W = \alpha_W \cdot W$ and $\lambda_K = \alpha_K \cdot K$.

2. The proliferator applies already available infrastructure to perform UAA. Let's assume that industrial enterprises in the search region consumes resources W in accordance with distribution $N(W)$, and frequency of the inspecting actions $F_{ins}(W)$ depends on the resource consumption rate also. Optimal scheme of the searching process can be found from the following optimality criterion: efficiency of the searching process does not depend on the proliferator strategy, i.e. $\lambda(W) \cdot T_P(W)$ is a constant value for any W, where T_P is proliferation time. Naturally, the larger available resources may be used by nuclear proliferator, the shorter time is needed to modify NM for successful NED manufacturing and military application. So, detection parameter depends on power consumed by a nuclear enterprise. Since power W consumed by nuclear enterprises and proliferation time T_P are linked by the energy E required to modify NM as $T = E/W$, the following equation can be written: $\lambda(W) \cdot (E / W) = \mathrm{const}$, or $\lambda(W) = \mathrm{const} \cdot (W / E) = \alpha_W \cdot W$.

In both models the emission rate parameter is proportional to the territorial area where abnormal emission level was observed, i.e. $\lambda_A \sim S(A) \sim R^2(A) \sim A$, or $\lambda_A = \alpha_A \cdot A$. So, each addend in equation (7) can be written as a product of two multipliers:

$$\lambda = \alpha_K \cdot K + \alpha_W \cdot W + \alpha_A \cdot A \qquad (8)$$

For example, detection parameter λ^W is equal to the mean UAA detection frequency on the resource consumption rate W. Of course, the UAA detection frequency depends on the sensitivity of the detecting devices to the resource consumption rate W, or to W-indicator. The sensitivity defines efficiency a^W of the searching process.

8.5 Comparative evaluations of external UAA monitoring efficiency and enhancement of inherent proliferation protection

The following problems are considered below: it is required to analyze dependency of metal uranium proliferation protection on uranium enrichment at different efficiencies of the searching process, and it is required to analyze the effects of uranium denaturing on its proliferation resistance, if uranium is denatured by admixing small amounts of ^{232}U that intensifies inherent neutron background. Nuclear proliferator does not resort to uranium re-enrichment up o the weapon-grade level, his main goal consists in a NED manufacturing.

Relative values of uranium proliferation protection were calculated for different efficiencies α of the searching process including the case when $\alpha = 0$, i.e. the case of uranium self-protection.

Mark-Hippel-Lyman model (Mark, 1993) of CFR initiation and propagation was used to evaluate damage from NED manufacturing and military use. CFR parameters were calculated by direct mathematical simulation of neutron multiplication process with application of Monte Carlo code MCNP-4B (Briesmeister, 1997) and evaluated nuclear data

file ENDF/B-VI (National Nuclear Data Center, 2001). Mathematical model and algorithm for determination of the model parameters correspond to the approach described in paper (Kryuchkov et al., 2008).

The results obtained in calculations of relative proliferation protection (inverse value to the risk) for different monitoring efficiencies and for different levels of uranium denaturing by ^{232}U are presented in Fig. 21.
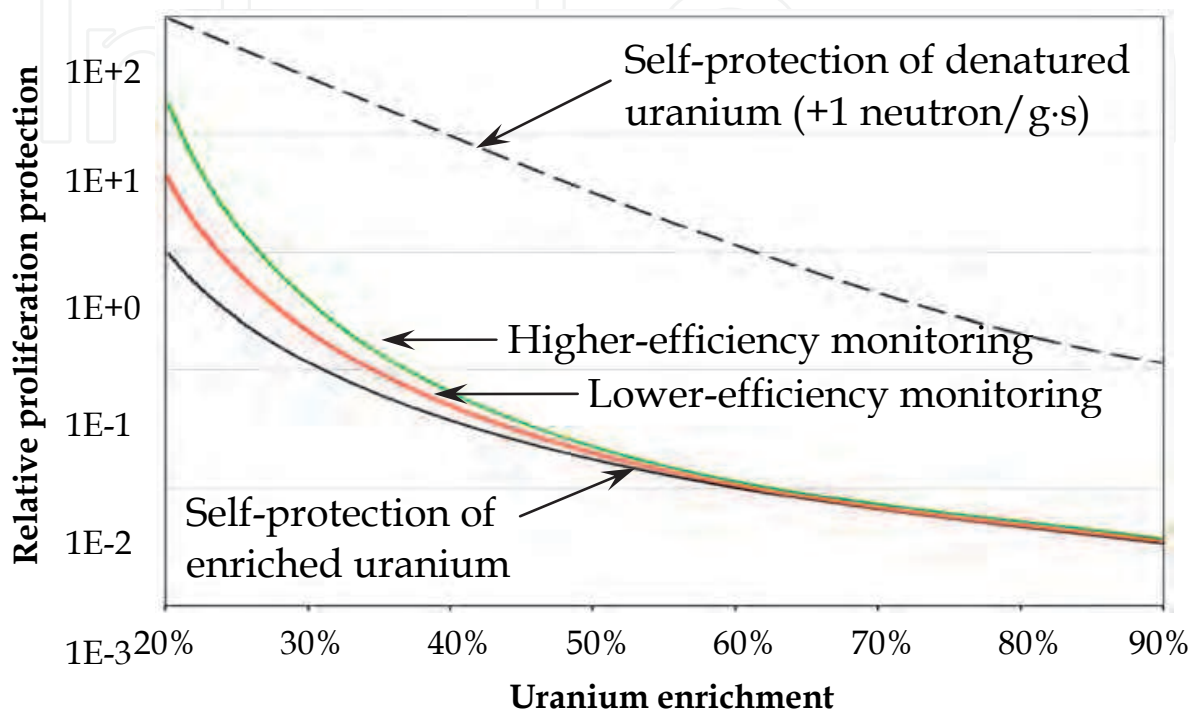

Fig. 21. Proliferation protection of metal uranium as a function of its enrichment

The following conclusions can be derived from numerical evaluations of metal uranium proliferation protection:

1. Measures of external monitoring (outside of MPC&A system) are ineffective ones in comparison with the measures aimed at upgrading of uranium self-protection for highly-enriched compositions.
2. Efficiency of external monitoring can excel efficiency of inherent self-protection for uranium enriched below 20% ^{235}U.
3. Upgrading of uranium self-protection by its denaturing, i.e. by formation of internal neutron source, weakly depends on uranium enrichment and provides approximately the same effect in a rather wide range of uranium enrichments.

So, nuclear non-proliferation requires maximal restrictions to be imposed on any transactions of pure HEU while free material must be maintained in a self-protected state produced by isotopic denaturing, for instance.

9. References

Babichev, A.P.; Babushkina, N.A.; Bratkovsky, A.M. et al. (1991) *Physical values*. Energoatomizdat, ISBN 5-283-04013-5

Belousov, N.; Bychkov, S.; Marchuk, Yu. et al. (1992). The Code GETERA for Cell and Polycell Calculations. Models and Capabilities, *Proceedings of the 1992 Topical Meeting on Advances in Reactor Physics*, Charleston, SC, USA, March 1992

Benedict, M.; Pigford, T.H. & Levi, H.W. (1981). *Nuclear Chemical Engineering* (2nd Edition), McGraw-Hill, ISBN 0-07-004531-3, New York

Briesmeister, J.F. (1997). MCNP - A General Monte Carlo N-Particle Transport Code - Version 4B. *Los Alamos National Laboratory Report, LA-12625-M*

Bychkov, A.V.; Vavilov, S.K.; Skiba, O.V. et al. (1997). Pyro-Electrochemical Reprocessing of Iradiated FBR MOX fuel. III. Experiment on High Burn-up Fuel of the BOR-60 Reactor, Proceedings *of the International Conference on Future Nuclear Systems "GLOBAL'97"*, Yokohama, Japan, October 1997

Cunningham, P.T.; Arthur, E.D.; Wagner, R.L. Jr. & Hanson, E.M. (1997). Strategies and Technologies for Nuclear Materials Stewardship, *Proceedings of the International Conference on Future Nuclear Systems "GLOBAL'97"*, Yokohama, Japan, October 1997

De Volpi, A. Denaturing Fissile Materials. (1982). *Progress in Nuclear Energy*, Vol.10, No.2, (November 1981), pp. 161-220, ISSN 0149-1970

Feiveson, H.A. (2005). In Memoriam – Ted Taylor. *Science & Global Security*, Vol.13, pp. 117–128, ISSN 0892-9882

Ganesan, S.; Sharma, A.R. & Wienke, H. (2002). New Investigations of the Criticality Property of Pure ^{232}U. *Annals of Nuclear Energy*, Vol.29, No.9, pp. 1085-1104, ISSN 0306-4549

Gilfoyle, G.R. & Parmentola, J.A. (2001). Using Nuclear Materials to Prevent Nuclear Proliferation. *Science & Global Security*, Vol.9, No.2, pp. 81-92, ISSN 0892-9882

Grachev, A.F.; Mayorshin, A.A.; Golovanov, V.N.; Tsykanov, V.A.; Bychkov, A.V. & Shishalov, O.V. (2003). Experience and prospects for the use of fuel rods made of vibro-packed oxide fuel, *Proceedings of the International Conference "Nuclear Power and Nuclear Fuel Cycles"*, Dimitrovgrad, Russia, December 2003

Ivanov, V.B.; Mayorshin, A.A.; Skiba, O.V. et al. (1997). The Utilization of Plutonium in Nuclear Reactors on the Basis of Technologies, Developed in SSC RIAR, Proceedings *of the International Conference on Future Nuclear Systems "GLOBAL'97"*, Yokohama, Japan, October 1997

Koch, L.; Betti, M.; Cromboom, O. & Mayer, K. (1997). Nuclear Material Safeguards for P&T, Proceedings *of the International Conference on Future Nuclear Systems "GLOBAL'97"*, Yokohama, Japan, October 1997

Kryuchkov, E.F.; Shmelev, A.N.; Masterov, S.V. et al. (2008). An Approach to Quantitative Evaluation of Inherent Proliferation Resistance of Uranium Enriched up to 20% ^{235}U, *Proceedings of the 30-th International Meeting on Reduced Enrichment for Research and Test Reactors*, Washington, D.C., USA, October 2008

Kulikov, E.G.; Shmelev, A.N. & Kulikov, G.G. (2007). Neutron-physical properties of (^{233}U-Th-^{238}U) fuel in LWR with supercritical coolant parameters. *Communications of Higher Schools. Nuclear Power Engineering*, No.2, pp. 27-39, ISSN 0204-3327

Mark, J.C. (1993). Explosive Properties of Reactor-Grade Plutonium. *Science & Global Security*, Vol.4, pp. 111-128, ISSN 0892-9882

Massey, J.V. & Schneider, A. (1982). The Role of Plutonium-238 in Nuclear Fuel Cycles. *Nuclear Technology*, Vol.56, No.1, pp. 55-71, ISSN 0029-5450

National Nuclear Data Center (NNDC). (2001). http://www.nndc.bnl.gov/nndcscr/ documents/endf/endf201/

Nojiri, I. & Fukasaku, Y. (1997). Calculational Study for Criticality Safety Data of Fissionable Actinides, Proceedings *of the International Conference on Future Nuclear Systems "GLOBAL'97"*, Yokohama, Japan, October 1997

Oak Ridge National Laboratory. (1995). *SCALE: A Modular Code System for Performing Standardized Computer Analyses for Licensing Evaluation*, Vols. I-III, NUREG/CR-0200, Rev. 4 (ORNL/NUREG/CSD-2/R4), Available from Radiation Shielding Information Center at Oak Ridge National Laboratory as CCC-545.

OECD Nuclear Energy Agency. (1997). *JEF-PC, Version 2.0, A Personal Computer Program for Displaying Nuclear Data from the Joint Evaluated File Library*

Reilly, D.; Ennslin, N. & Smith, H.Jr. (1991). *Passive Nondestructive Assay of Nuclear Materials*. Los Alamos National Laboratory, ISBN 978-999-6344-45-9

Rolland-Piegue, C. (1995). Safeguards and Non-Proliferation for Advanced Fuel Cycles. IAEA Safeguards on Plutonium and HEU, *Proceedings of the International Conference on Evaluation of Emerging Nuclear Fuel Cycle Systems "GLOBAL'95"*, Versailles, France, September 1995

Selle, J.E.; Angelini, P.; Rainey, R.H. & Federer, J.I. (1979). Technical Consideration of the Use of Nuclear Fuel Spikants for Proliferation Deterrence. *Nuclear Technology*, Vol.45, No.3, (October 1979), pp. 269-286, ISSN 0029-5450

Sokolova, I.D. (2008). Program of Global Nuclear Energy Partnership (GNEP). *Atomic Techniques Abroad*, No.3, (March 2008), pp. 3-13, ISSN 0320-9326

USA National Academy of Sciences. (2000). *The Spent-Fuel Standard for Disposition of Excess Weapon Plutonium. Application to Current DOE Option,* National Academy Press, ISBN 0-309-07320-0, Washington, D.C.

Willrich, M. & Taylor, T.B. (1974). *Nuclear Theft: Risks and Safeguards: A report to the Energy Policy Project of the Ford Foundation*, Ballinger Pub.Co., ISBN 0884102076, Cambridge
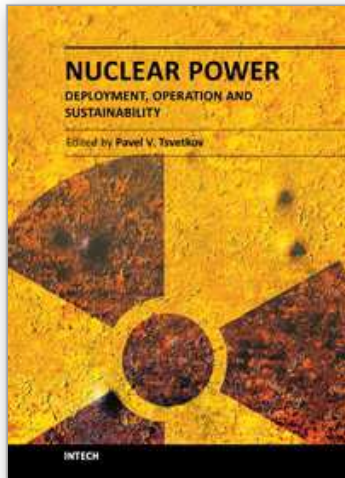
Nuclear Power - Deployment, Operation and Sustainability

Edited by Dr. Pavel Tsvetkov

ISBN 978-953-307-474-0

Hard cover, 510 pages

Publisher InTech

Published online 09, September, 2011

Published in print edition September, 2011

We are fortunate to live in incredibly exciting and incredibly challenging time. Energy demands due to economic growth and increasing population must be satisfied in a sustainable manner assuring inherent safety, efficiency and no or minimized environmental impact. These considerations are among the reasons that lead to serious interest in deploying nuclear power as a sustainable energy source. At the same time, catastrophic earthquake and tsunami events in Japan resulted in the nuclear accident that forced us to rethink our approach to nuclear safety, design requirements and facilitated growing interests in advanced nuclear energy systems. This book is one in a series of books on nuclear power published by InTech. It consists of six major sections housing twenty chapters on topics from the key subject areas pertinent to successful development, deployment and operation of nuclear power systems worldwide. The book targets everyone as its potential readership groups - students, researchers and practitioners - who are interested to learn about nuclear power.

INTECH

open science | open minds