CONTROLLED THERMONUCLEAR FUSION: POTENTIAL ROLE OF A JOINT (TH-U-PU) NUCLEAR FUEL CYCLE

V.A. APSE
National Research Nuclear University MEPhI
Moscow, Russian Federation

N.I. GERASKIN
National Research Nuclear University MEPhI
Moscow, Russian Federation

E.G. KULIKOV
National Research Nuclear University MEPhI
Moscow, Russian Federation
Email: egkulikov@mephi.ru

G.G. KULIKOV
National Research Nuclear University MEPhI
Moscow, Russian Federation

V.I. SAVANDER
National Research Nuclear University MEPhI
Moscow, Russian Federation

A.N. SHMELEV
National Research Nuclear University MEPhI
Moscow, Russian Federation

Abstract

The paper aims at proposing an innovative nuclear fuel cycle based on ^{231}Pa and thorium. The method is including fusion neutron sources with thorium blanket into future nuclear power system. In addition to production of light uranium fraction consisting of ^{233}U and ^{234}U, high-energy 14-MeV neutrons emitted in the process of fusion (D,T)-reaction can generate ^{231}Pa and ^{232}U through (n,2n)- and (n,3n)-reactions. It has been demonstrated that admixture of ^{231}Pa into fresh fuel composition can stabilize its neutron-multiplying properties thanks to two well-fissile consecutive isotopes ^{232}U and ^{233}U, products of radiative neutron capture by ^{231}Pa. Coupled system of two well-fissile isotopes can allow us to reach the following goals: the higher fuel burn-up and, as a consequence, the longer fuel lifetime; the shorter scope and the lower number of technological operations in nuclear fuel cycle; the better economic potential of nuclear power technologies. Such a fuel cycle presumes shifting from ^{235}U to ^{233}U as more attractive fuel material for thermal nuclear reactors. Uranium component will be protected from unauthorized proliferation by the presence of light uranium isotope ^{232}U. The use of well-mastered traditional uranium-based fuels in power LWR will be preserved. The idea suggests fresh fuel fabrication for power LWR without applications of isotope separation technologies. Proposed innovative nuclear fuel cycle based on ^{231}Pa and thorium has such issue as availability of important quantities of ^{231}Pa that may be accumulated in thermonuclear fusion reactor. However, since the purpose of such a thermonuclear fusion reactor would be accumulation of ^{231}Pa rather than production of energy, requirements for its construction are lower and therefore it is more realistic project comparing to fusion reactor aimed at production of energy.

1. INTRODUCTION

The further development of nuclear power system with introduction of the hybrid “fusion-fission” facilities looks as a very promising option. Main mission of the hybrid facilities may be production of fissile isotopes for nuclear power reactors, not production of thermal and electrical energy. Here it does not matter what types of nuclear power reactors will be main consumers of fuel produced by the hybrid facilities. One else potential mission of the hybrid facilities may consist in neutron transmutation of radioactive wastes (long-lived fission products and minor actinides). The waste transmutation by high-energy fusion neutrons makes it possible to reduce significantly the risk for accidental radioactive contamination of the environment. So, thermonuclear

synthesis of light nuclides becomes an active participant of the world-wide energy market. This circumstance opens broad perspectives for new scientific studies on effective ways towards intense generation of fusion neutrons [1].

Only mutually profitable combination of two nuclear technologies (fusion-based and fission-based) within the frames of the joint nuclear power system can result in steady-state and large-scale usage of nuclear energy. Independent self-development of these two technologies can lead only to their gradual recession.

Evidently, in the nearest future the further development of nuclear power systems will be based on the conception of open nuclear fuel cycle. Gradual involvement of the hybrid "fusion-fission" facilities can extend the fuel resources of nuclear power reactors for any long time period.

The following three properties of the joint "fusion-fission" nuclear power system can define this option as a major strategic pathway [1]:

— Production of artificial fissile isotopes for nuclear power reactors can be organized in the scales high enough for any potential structures of the power system (fast or thermal reactors, large-size or small-size modular reactors, etc.).

— Fraction of the fusion facilities in the joint nuclear power systems can be kept well below 10%. Such a small fraction provides economical efficiency of the system even if the fusion facilities are remarkably more cost-expensive than the fission facilities.

— The risks of accidental radioactive contamination of the environment by the joint systems can be decreased by two-three orders of magnitude as compared with one-component fission-based power system. The joint two-component power system can exclude, in principle, reprocessing of spent nuclear fuel with high values of fuel burn-up and radioactivity.

2. CONTROLLED THERMONUCLEAR FUSION (CTF) CAPABILITY TO PRODUCE FISSILE MATERIALS WITH STABLE NEUTRON-MULTIPLYING PROPERTIES AND WITH THE HIGHER VALUES OF FUEL BURN-UP

The "fusion-fission" CTF concept presumes that high-energy (14.1 MeV) neutrons from thermonuclear reactions in (D,T)-plasma are used to irradiate either uranium or thorium blanket. Neutron irradiation of thorium blanket is a preferable option [1] because only these neutrons are able to initiate threshold ^{232}Th(n,2n) and ^{232}Th(n,3n) reactions. Just these neutron reactions can open a possibility for production of such fuel compositions which are able to maintain long-term operation of nuclear power reactors without frequent refuelings.

If fuel rods can withstand deep fuel burn-up, then one-two refuelings are only required per full lifetime of the reactor operation. Moreover, under certain conditions, the reactor may operate without refuelings at all, in the "Black Box" operation mode. Till now, a certain experience was acquired in designing of fuel rods for fast reactors with increased values of fuel burn-up (above 32% HM): vibro-compacted MOX-fuel pellets in stainless steel cladding [2, 3]; fuel rods with micro particles for high-temperature gas-cooled reactors; rods with composite ceramics-metal fuels [4]. A possibility of principle for the long-term reactor operation without refuelings is based on specific features of the fuel compositions produced in thorium blanket of hybrid "fusion-fission" facility. One of these features consists in stable neutron-multiplying properties during long-term operation of nuclear power reactors.

Neutron irradiation of ^{232}Th in the blanket of hybrid "fusion-fission" facility results in initiation of the following reactions (Fig. 1):

1) Radiative neutron capture by thorium with production of well-fissile isotope ^{233}U.
2) Threshold reactions ^{232}Th (n,2n) ^{231}Th (β) ^{231}Pa and ^{232}Th (n,3n) ^{230}Th (n,γ) ... ^{231}Pa.
3) Radiative neutron capture by protactinium ^{231}Pa (n,γ) ^{232}U.

In addition to traditional isotope ^{233}U, these neutron reactions can produce some non-traditional isotopes, namely ^{231}Pa and ^{232}U.

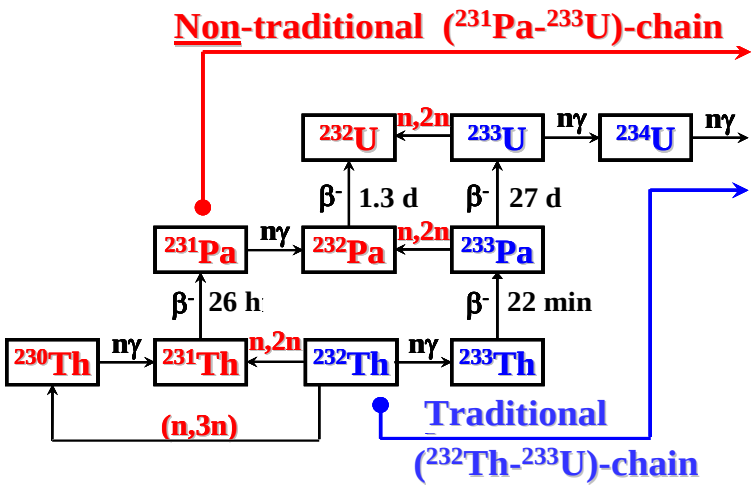

FIG. 1. Chains of isotopic transformations in (Th-U) fuel cycle.

Neutron irradiation of thorium blanket of hybrid "fusion-fission" facility can initiate two chains of isotopic transformations, namely traditional chain (^{232}Th – ^{233}U – ^{234}U) and non-traditional chain (^{232}Th – ^{231}Pa – ^{232}U – ^{233}U – ^{234}U). Rates of threshold (n,2n) and (n,3n) reactions are quite comparable with rates of radiative neutron capture reaction, as is seen from Table 1. This table presents experimental and calculated values of neutron reaction rates in thorium assembly irradiated by fusion neutrons [5-7].

TABLE 1. REACTION RATES PER ONE INCIDENT FUSION NEUTRON (E_n = 14.1 MeV) IN EXPERIMENTAL THORIUM-BEARING ASSEMBLY

Reaction	Experiment	Calculation
^{232}Th (n,f)	0.174±0.010	0.193
^{232}Th (n,γ)	1.63 ± 0.10	1.58
^{232}Th (n,2n)	0.42 ± 0.04	0.58
^{232}Th (n,3n)	0.30 ± 0.05	0.15
Neutron leakage	0.78 ± 0.04	0.762

It may be seen that rates of threshold ^{232}Th(n,2n) and ^{232}Th(n,3n) reactions constitute above 40% from rate of radiative neutron capture reaction ^{232}Th(n,γ)^{233}U.

If isotopes ^{231}Pa, ^{232}U and ^{233}U produced in thorium blanket of hybrid "fusion-fission" facility are introduced into fresh fuel composition of nuclear power reactor, then the following chain of consecutive isotopic transformations with gradual improvement of neutron-multiplying properties can be initiated: ^{231}Pa (burning absorber) → ^{232}U (moderate-fissile isotope) → ^{233}U (well-fissile isotope).

3. ACHIEVEMENT OF ULTRA-HIGH FUEL BURN-UP THANKS TO ^{231}PA

Isotope ^{231}Pa, as a burning absorber, can decrease initial reactivity margin because of large micro cross-sections of neutron absorption. The reactivity effect of ^{231}Pa is quite similar to the reactivity effects produced by gadolinium, which is being widely used as a burning absorber in thermal light-water reactors. Fortunately, the reactivity effect of ^{231}Pa differs profitably from that of gadolinium.

Firstly, micro cross-sections of neutron absorption by ^{231}Pa are not so large as the same micro cross-sections of gadolinium. Therefore, the stabilizing effect of ^{231}Pa on neutron-multiplying properties of the reactor core acts during the longer time interval in comparison with gadolinium. At the same time, micro cross sections of neutron absorption by ^{231}Pa are substantially higher than those of ^{238}U. So, it is unnecessary to introduce extremely large fraction of ^{231}Pa into fresh fuel composition in order to achieve its favorable effect.

Secondly, the neutrons absorbed by ^{231}Pa have a possibility to come back to the chain fission reaction through fissions of its daughter isotopes. The first of them, ^{232}U is a moderately fissile isotope for thermal and

intermediate neutrons. The next isotope ^{233}U, product of $^{232}U(n,\gamma)$ reaction, is a well-known and well-fissile isotope.

Numerical studies were carried out with application of the computer code SCALE-6.1 [8] and the evaluated nuclear data file ENDF/B-VII for elementary cell of light-water VVER-1000 reactor: fuel – uranium dioxide, density of light-water coolant – 0.72 g/cm^3, fuel cladding – martensitic stainless steel MA956 with the following composition: 74.5% Fe, 20% Cr, 4.5% Al, 0.5% Ti and 0.5% Y_2O_3.

Time evolution of neutron multiplication factor was analyzed for two cases, with and without introduction of ^{231}Pa-based neutron absorber into fresh fuel composition, and presented in Fig. 2.

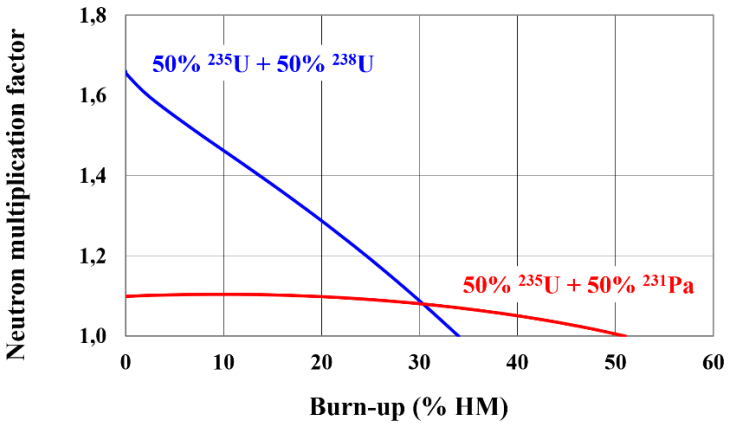

FIG. 2. Achievement of ultra-high fuel burn-up in VVER-1000 thanks to the application of ^{231}Pa as a burning absorber

As is seen in Fig. 2, if fuel does not contain the burning absorber (50% ^{235}U + 50% ^{238}U), then initial reactivity margin is sufficiently large (K_∞ (0) ≈ 1.7) to reach maximal fuel burn-up about 34% HM. If fuel contains the burning absorber (50% ^{235}U + 50% ^{231}Pa), then initial value of neutron multiplication factor is close to unity and remains nearly the same during fuel lifetime. Such time evolution of neutron multiplication factor can be only explained by the fact that neutron absorption by fission products and incineration of the primary fissile isotopes are almost completely compensated by reproduction (breeding) of the secondary fissile isotopes, daughter products of the burning absorber ^{231}Pa.

Neutron spectrum shifts towards resonance range because of intense neutron absorption by ^{231}Pa. Significant ^{231}Pa fraction (about 80%) converts to the secondary fissile isotopes (^{232}U and ^{233}U), whose fission neutrons can sustain the reactivity margin and, thus, provide ultra-high fuel burn-up (about 51% HM). Under operation conditions of the VVER-1000 reactor (fuel load ~ 66 tons, thermal power ~ 3000 MW) this value of fuel burn-up corresponds to the fuel lifetime at the level of 35 years.

4. PROLIFERATION RESISTANCE OF THE PROPOSED MULTI-ISOTOPE FUEL COMPOSITION

Here the term “proliferation resistance” should be understood as a protection of fissile materials against their switching over to any non-energy-producing purposes.

In addition to the systems of nuclear materials physical protection, control and accountability, proliferation resistance of fissile materials in the currently existent nuclear fuel cycle (mainly, once-through NFC with uranium fuel in light-water reactors) is defined by the following two factors:

1) Relatively low uranium enrichment makes it impossible to use such uranium directly, i.e. without preliminary isotope separation, as a weapon-grade material.

2) Spent Pu-bearing fuel is protected by high level of residual heat generation rate and by intense radiation of fission products and minor actinides until they are removed by technologies of spent fuel reprocessing.

In “traditional” uranium-thorium fuel isotope ^{233}U is, in essence, a weapon-grade material. Extraction of ^{233}U from U-Th fuel makes this fuel cycle vulnerable from the standpoint of nuclear proliferation. It was proposed to dilute ^{233}U with other uranium isotope ^{238}U but, in this case, ^{238}U acts as a source for plutonium

build-up. Accumulation of weapon-grade plutonium in ^{238}U-bearing fuel requires to undertake appropriate protective measures.

A possibility exists to upgrade proliferation resistance of ^{233}U in U-Th fuel by the fuel denaturing with one else uranium isotope ^{232}U [9]. This isotope is an intense heat source from α-decays (half-life 68.9 years, specific heat generation rate 740 W/kg) and a source of spontaneous fission neutrons. Besides, ^{232}U decay products are intense sources of high-energy γ-rays. If charge of a nuclear explosive device is made of ^{232}U-containing uranium, then some additional means are required for heat removal and for distant management. It is important to note here that α-decays of ^{232}U are able to complicate significantly application of isotope separation technologies for removal of isotope ^{232}U.

In mixed (Th-U-Pu) fuel plutonium may play an auxiliary role in comparison of the dominant role played by ^{233}U. So, Pu fraction in such a fuel may be relatively small. In this case, plutonium can be removed from the world-wide nuclear power and utilized in the dedicated nuclear facilities, as it was foreseen in the Global Nuclear Energy Partnership (GNEP) initiative proposed by the US President [10]. By the way, in this case, plutonium will be additionally denatured by its isotopes ^{238}Pu and ^{240}Pu.

Uranium fraction in mixed (Th-U-Pu) will contain practically full spectrum of all significant uranium isotopes: ^{232}U, ^{233}U, ^{234}U, ^{235}U, ^{236}U and ^{238}U. Such a composition represents a low-enriched uranium with small content of main fissile isotopes ^{233}U and ^{235}U. If any unsanctioned attempts to upgrade uranium enrichment up to the weapon-grade level are undertaken, then large, maybe insuperable difficulties can arise.

As is known, manufacturing of even primitive nuclear explosive device requires availability of a weapon-grade uranium. According to the IAEA regulations [9], content of ^{235}U in uranium suitable for this purpose must be above 20%. So, the proposed multi-isotope uranium must undergo the process of isotope separation. If the most advanced gas-centrifuge technology is used for uranium isotope separation, then the process encounters the following barriers, difficult to overcome.

Firstly, main fissile isotopes ^{233}U and ^{235}U are surrounded from both sides by other uranium isotopes (neutron absorbers) and ^{232}U. Their separation requires applying the sophisticated scheme of a single separative cascade or several separative cascades in a common scheme [11]. The separation efficiency of these schemes will be low because of small differences between molecular weights of uranium hexafluorides $^{232}UF_6$, $^{233}UF_6$, $^{234}UF_6$, $^{235}UF_6$ and $^{238}UF_6$ (ΔM = 1 a.m.u.) while, in the case of natural uranium enriching, this difference is equal to 3 a.m.u. As is known, total mass of the gas that circulates in the separative cascades and total scope of separative works are inversely proportional to the squared single-stage enrichment gain which, in its turn, directly proportional to the difference ΔM [12].

Secondly, the presence of uranium isotope ^{232}U, the most powerful α-emitter, even in a very small amounts (decimal percent fractions), as well as the presence of other uranium isotopes ^{233}U and ^{234}U, α-emitters too, in uranium to be enriched can initiate some physical and chemical processes which are able to disorder the uranium enriching procedures. These disorders can take place during full time of the separative cascade operation including a preparatory period.

Uranium isotope ^{232}U is a dominant contributor to emission of high-energy α-particles (half-life $T_{1/2}$ = 68.9 years, mean energy E_α = 5.3 MeV). During the braking time of α-particles in uranium hexafluoride one α-particle is able to destroy nine, in average, UF_6 molecules per 1 keV of its energy loss [13, 14]. So, one α-particle is able to destroy about 48000 UF_6 molecules during full braking time. As a result, the lower uranium fluorides (UF_3, UF_4 and UF_5) and free fluorine will appear in uranium hexafluoride and collide with each other, mainly with UF_6. The energy necessary to tear fluorine atom out of UF_6 molecule is relatively small (1.97 eV) while the energy necessary to join fluorine atom to the lower uranium fluorides is substantially larger, about 5-6 eV (Table 2). That is why the lower uranium fluorides UF_3 and UF_4 are quickly transformed to uranium pentafluoride UF_5 [15, 16].

TABLE 2. ENERGIES FOR TEARING FLURINE ATOMS OUT OF URANIUM FLUORIDES

Fluoride molecule	$UF_4 \rightarrow UF_3+F$	$UF_5 \rightarrow UF_4+F$	$UF_6 \rightarrow UF_5+F$
Tearing energy, eV	6.56	5.38	1.97

If molecules of uranium pentafluoride UF_5 collide with molecules of uranium hexafluoride UF_6, then the isotope exchange reactions can be initiated. For example:

$$^{235}UF_5 + {}^{238}UF_6 \rightarrow {}^{235}UF_6 + {}^{238}UF_5.$$

As is seen, before collision molecular weight of $^{235}UF_5$ was substantially lighter than that of $^{238}UF_6$ (330 a.m.u. versus 352 a.m.u.) but after collision molecular weight of $^{238}UF_5$ became remarkably lighter than that of $^{235}UF_6$ (333 a.m.u. versus 349 a.m.u.). This isotope exchange reaction can disorder a correspondence between atomic weights of uranium isotopes and molecular weights of uranium fluorides. The lighter $^{238}UF_5$ molecules will go to the enriching branch of the separative cascade while the heavier $^{235}UF_6$ molecules will go to the depleting branch of the separative cascade.

Molecules of uranium pentafluorides (with different uranium isotopes) can collide with each other, coagulate with formation of the heavier aggregates, move towards the centrifuge periphery and precipitate on the inner centrifuge surface. Moreover, these coagulates can seriously contaminate the connecting pipelines of the separative cascade. Just such a radioactive contamination takes place in the uranium enriching process by means of the MLIS (Molecular Laser Isotope Separation) technology [17]. Here the isotope exchange reactions between gaseous UF_6 phase and solid UF_5 phase lead to a remarkable reduction of the MLIS efficiency. The same negative effects can occur in the gas-centrifuge technology. It should be noted here that the gas ionization process induced by α-particles is able to intensify remarkably the isotope exchange reactions [18].

Specialists from the National Research Center “Kurchatov Institute” (Moscow, Russia) have recently worked out the methodology that is able to weaken negative effects from UF_6 radiolysis induced by incident α-particles [19]. The “carrier” inert gas $C_8H_3F_{13}$ (freon) is admixed to gaseous uranium hexafluoride UF_6. Mission of the “carrier” gas is to remove main α-emitter $^{232}UF_6$ out of the circulating gas flow. As the developers have said, “application of the carrier gas may be helpful in the separation process of radioactive gaseous mixtures for decreasing the radiolysis effects by dilution with inert material”.

As hexafluoride of uranium isotope ^{232}U is a main α-emitter, the “carrier” gas must be selected by such a way that molecular weight of $^{232}UF_6$ (346 a.m.u.) and molecular weight of the “carrier” gas – freon $C_8H_3F_{13}$ (346 a.m.u.) coincided. Radioactive component of the gas mixture is, as if, immersed into large volume of the “carrier” gas.

The acceptable levels for dilution of uranium hexafluoride by the “carrier” gas can be determined from the following consideration. Any admixture of the “carrier” gas increases the scope of separative works needed to produce enriched uranium. Large dilution can cause so significant increase of the cascade dimensions and the scope of separative works that it becomes unfeasible to enrich ^{232}U-bearing uranium. It might be simpler and cheaper to enrich natural uranium.

Indeed, real situation with radiolysis of uranium hexafluoride is much more complicated because of the following processes: ion-molecule interactions followed by formation of freon fragments with broken carbon links, recombination of freon molecules and freon fragments with molecules and fragments of the lower uranium fluorides. Presently, no quantitative evaluations of these processes in the existent gas centrifuges are available.

In conclusion the authors would like to note that any content of uranium isotope ^{232}U and its decay products in the separative cascade leads to radioactive contamination of the involved equipment units [20].

5. INNOVATIVE CLOSED NUCLEAR FUEL CYCLE

The global nuclear power by now has already accumulated a large amount of irradiated fuel, which contains quite a lot of plutonium. There are concerns of this plutonium and its subsequent fate for various reasons, among which an important place is occupied by the problem of its proliferation. In order to reduce the risk of uncontrolled proliferation of plutonium, the concept of its burning under international control was developed at LANL (USA) and announced by US Department of Energy (DOE) in 2006. The concept has the abbreviation GNEP, meaning Global Nuclear Energy Partnership. In 2009 the DOE announced the cancellation of the US domestic component of GNEP.

Within this approach the new improved scheme of innovative closed nuclear fuel cycle includes fusion neutron source with Th-blanket to generate (^{231}Pa-^{232}U-^{233}U)-fuel for nuclear reactors (Fig. 3).

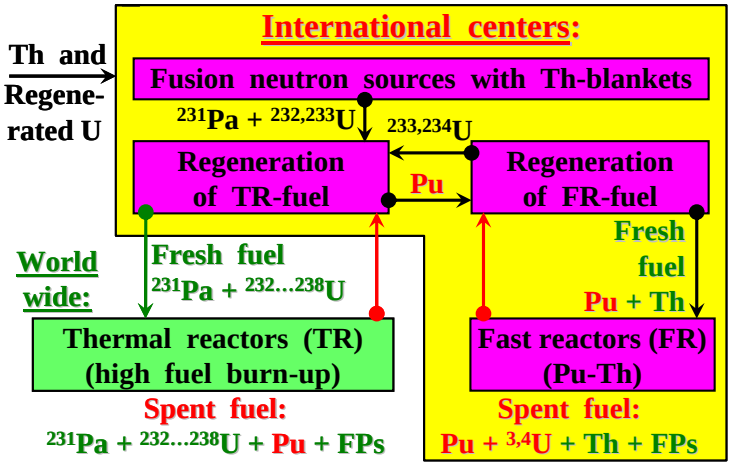

FIG. 3. Innovative nuclear fuel cycle based on ^{231}Pa and Th

To simplify the scheme of the innovative closed fuel cycle a fuel cycle of fusion neutron source for tritium supply is not included into the figure. It is assumed that tritium generation would be realized by traditional way into lithium blanket of fusion neutron source by neutron irradiation. Then accumulated tritium is extracted, from irradiated lithium, and injected into plasma of fusion neutron source.

The proposed innovative closed fuel cycle has a whole number of very important advantages:

- ✓ Such a fuel cycle presumes shifting from ^{235}U to ^{233}U as more attractive fuel material for thermal nuclear reactors.
- ✓ Light uranium fraction (containing mainly ^{233}U) is the most proliferation-resistant part of uranium component (due to presence of denaturing ^{232}U) and, being mixed with regenerated uranium, becomes to be low-enriched uranium fuel. This option can weaken the problem of unauthorized proliferation of fissile materials and, thus, upgrade the export potential of nuclear power industry.
- ✓ Generation of plutonium fraction in such uranium-based fuel will be suppressed because some part of fertile uranium isotope ^{238}U is replaced by ^{231}Pa. This is an additional factor to upgrade export potential of nuclear power technologies.
- ✓ The use of well-mastered traditional uranium-based fuels in power LWR will be preserved. The lower plutonium quantities can be incinerated in fast reactors placed at international centers (i.e. under international control).
- ✓ Fresh fuel fabrication for power LWR without applications of isotope separation technologies makes it possible to improve potential abilities of nuclear non-proliferation regime and simplify the existing technologies of fresh fuel fabrication.
- ✓ Very moderate requirements for fusion neutron sources, namely: without plasma ignition and without energy generation.

6. CONCLUSIONS

Thorium blanket of the hybrid “fusion-fission” facility is able to produce not only uranium isotope ^{233}U but also multi-isotope mixture of ^{231}Pa with all other significant uranium isotopes (from ^{232}U to ^{238}U). Such a multi-isotope fuel can be profitably used in nuclear power reactors because the proposed fuel composition is characterized by the stable neutron-multiplying properties during full time of the reactor operation.

Nuclear power reactors loaded with such a fuel distinguish profitably by the longer lifetimes, high and ultra-high values of fuel burn-up (about 50% HM) at the reactor operation without refueling, shortened scope of operations with fresh and spent fuel in the outer NFC part.

In mixed Th-U-Pu fuel the multi-isotope uranium fraction represents a continuous series of all significant uranium isotopes, from ^{232}U to ^{238}U. Such a fuel composition can complicate substantially the process of uranium enriching for illegal applications.

Highly active α-emitters (mainly, ^{232}U, ^{233}U and ^{234}U) in the multi-isotope uranium fraction are able to initiate radiolysis of uranium hexafluoride UF_6 followed by destruction of UF_6 molecules, recombination and

isotope exchange reactions, the coagulation and precipitation processes. All these effects can disorder a correspondence between atomic weights of uranium isotopes and molecular weights of uranium fluorides.

Thus, it may be concluded that thorium blanket of hybrid “fusion-fission” facility offers a possibility of principle to produce a new kind of nuclear fuel. The stable neutron-multiplying properties of such a fuel are able to provide ultra-high values of fuel burn-up and ultra-long lifetimes of nuclear power reactors. Significant amounts of the proposed fuel can be only produced in thorium blanket of hybrid “fusion-fission” facility because intense threshold ^{232}Th(n,2n) and ^{232}Th(n,3n) reactions can be only initiated by high-energy fusion neutrons. Mixed Th-U-Pu fuel composition contains fissile isotopes which are characterized by enhanced proliferation protection against any unauthorized attempts of their switching over to non-energy-producing applications.

ACKNOWLEDGEMENTS

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