Closed fuel cycle with increased fuel burn-up and economy applying of thorium resources

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Abstract. The possible role of existing thorium reserves in the Russian Federation on engaging thorium in being currently closed (U-Pu)-fuel cycle of nuclear power of the country is considered. The application efficiency of thermonuclear neutron sources with thorium blanket for the economical use of existing thorium reserves is demonstrated. The aim of the work is to find solutions of such major tasks as the reduction of both front-end and back-end of nuclear fuel cycle and an enhancing its protection against the uncontrolled proliferation of fissile materials by means of the smallest changes in the fuel cycle. During implementation of the work we analyzed the results obtained earlier by the authors, brought new information on the number of thorium available in the Russian Federation and made further assessments. On the basis of proposal on the inclusion of hybrid reactors with Th-blanket into the future nuclear power for the production of light uranium fraction $^{232+233+234}$U, and ^{231}Pa, we obtained the following results: 1. The fuel cycle will shift from fissile ^{235}U to ^{233}U which is more attractive for thermal power reactors. 2. The light uranium fraction is the most "protected" in the uranium component of fuel and mixed with regenerated uranium will in addition become a low enriched uranium fuel, that will weaken the problem of uncontrolled proliferation of fissile materials. 3. ^{231}Pa doping into the fuel stabilizes its multiplying properties that will allow us to implement long-term fuel residence time and eventually to increase the export potential of all nuclear power technologies. 4. The thorium reserves being near city Krasnoufimsk (Russia) are large enough for operation of large-scale nuclear power of the Russian Federation of 70 GWe capacity during more than a quarter century under assumption that thorium is loaded into blankets of hybrid TNS only. The general conclusion: the inclusion of a small number of hybrid reactors with Th-blanket into the future nuclear power will allow us substantially to solve its problems, as well as to increase its export potential.

1. Introduction

Closure of nuclear fuel cycle (NFC), the process under practical implementation now in the Russian Federation, can substantially upgrade efficiency of uranium utilization. Therefore, additional involvement of natural thorium resources into nuclear fuel cycle for further enlargement of practically unlimited natural uranium resources looks now as an option with the slightest validity. This option will require large additional expenses for development and industrial mastering of complicated technologies for thorium mining, primary processing, fabrication and handling with other nuclear fuel material. It would be appropriately to note here that till now no one country had decided yet to undertake wide involvement of natural thorium resources into national nuclear power system (NPS), even India with large stockpiles of natural thorium.

So, involvement of thorium resources into NPS in the process of the NFC closure has to be validated by other and very weighty arguments. As is known, neutron-physical parameters of thermal power reactors can be remarkably improved by transition from uranium (^{235}U-^{238}U) fuel to uranium-thorium (^{233}U-^{232}Th) fuel. Just this consideration has been taken into account when "The Strategy on Development of Russian Nuclear Power System in the First Half of the XXI century" [1] was formed. The Strategy defines that closure of uranium-plutonium NFC will be followed by thorium involvement, and thermal reactors will be gradually transferred from uranium-plutonium fuel to uranium-thorium (^{233}U-^{232}Th) fuel.

2. Thorium application for generation of "light" uranium fraction

When the problem of thorium involvement into the NFC to be closed is under discussion, it seems reasonable to give attention to the following important (in a practical sense) circumstance. As is known, from the very beginning of nuclear power development and till nowadays the NPS is based on utilization of "heavy" uranium fraction $^{235+236+238}$U, where ^{236}U is a component of the regenerated uranium.

As far as thorium will be involved into the NFC, then a new (on its isotopic composition) uranium material appears in spent fuel, and the following neutron reactions (Fig. 1) are the channels for its generation:

1) Radiative neutron capture by thorium with generation of fissile isotope ^{233}U.
2) Threshold reactions of high-energy neutrons:
    ^{232}Th$(n,2n)^{231}$Th$(\beta^-)^{231}$Pa; ^{232}Th$(n,3n)^{230}$Th$(n,\gamma)...^{231}$Pa.
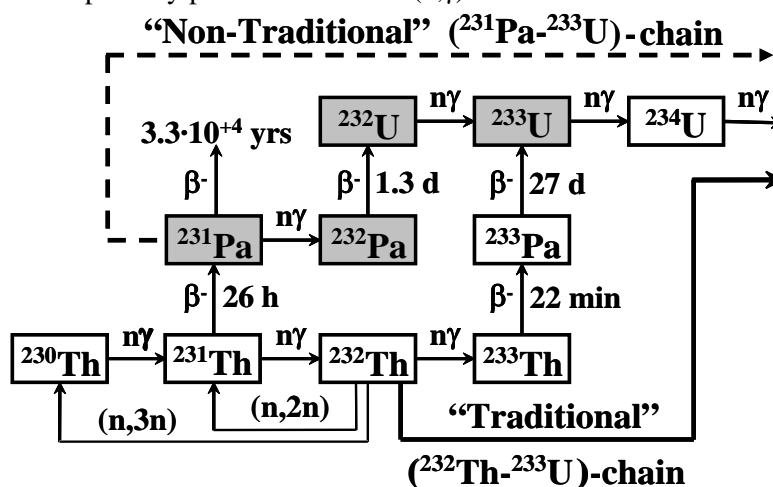3) Radiative neutron capture by protactinium: ^{231}Pa$(n,\gamma)...^{232}$U.

Figure 1. Chains of nuclide transformations in the thorium-uranium fuel.

These reactions can generate, in addition to fissile uranium isotope ^{233}U, protactinium isotope ^{231}Pa and uranium isotope ^{232}U in the neutron-irradiated thorium. The chains of isotopic transitions initiated by neutron-thorium reactions can be grouped by such a way:

1) "Traditional" chain that started from radiative neutron capture by ^{232}Th (the capture channel).
    ^{232}Th$(n,\gamma)^{233}$Th$(\beta^-, T_{1/2}=22')^{233}Pa(\beta^-, T_{1/2}=27$ d) ^{233}U$(n,\gamma)^{234}$U$(n,\gamma)...$

2) "Non-traditional" chain that started from threshold ^{232}Th$(n,2n)$ and ^{232}Th$(n,3n)$ reactions (the threshold channel):
    ^{232}Th$(n,2n)^{231}$Th$(\beta^-, T_{1/2}=26$ h)^{231}Pa$(n,\gamma)^{232}$U$...$
    ^{232}Th$(n,3n)^{230}$Th$(n,\gamma)^{231}$Th$(\beta^-, T_{1/2}=26$ h)^{231}Pa$...$
    ^{231}Pa$(n,\gamma)^{232}$Pa$(\beta^-, T_{1/2}=1.3$ d)^{232}U$(n,\gamma)^{233}$U$(n,\gamma)^{234}$U$(n,\gamma)^{235}$U$...$

So, introduction of natural thorium in fresh fuel composition will result in production of the "light" uranium fraction, namely mixture of light uranium isotopes ^{232}U, ^{233}U, ^{234}U with dominant role of well-fissile isotope ^{233}U.

If the "light" uranium fraction is used as a component of fresh nuclear fuel, then the NFC of nuclear power reactors (both thermal and fast reactors) may be based on utilization of either uranium-based fuel or mixed uranium-plutonium fuel compositions for a long time, even for an ultra-long time period taking into account practically unlimited resources of natural uranium and natural thorium. Besides, plutonium and minor actinides can be utilized within the frames of the NFC. Isotope composition of uranium fraction in fresh fuel will contain mixture of light uranium isotopes ^{232}U, ^{233}U, ^{234}U with uranium regenerated from spent LWR fuel.

If the "light" uranium fraction is generated by thorium-based fertile materials in relatively small number of highly efficient producers of fissile materials, then the main park of nuclear power reactors will be fuelled by uranium as before. The hybrid thermonuclear "FUSION-FISSION" reactors with thorium blankets and with plasma parameters already achieved in contemporary experimental fusion installations can be regarded, in the first turn, as such producers. In the second turn, accelerator-driven facilities and fast reactors with thorium blankets can be also used to produce the "light" uranium fraction but they are characterized by relatively low production potential.

Thus, the closed (^{232}Th-^{233}U-^{235}U-^{238}U-Pu) NFC with multiple fuel recycle and with feeding by the "light" uranium fraction can form the closed (^{232}Th-^{232}U-^{233}U-^{234}U-^{235}U-^{236}U-^{238}U-Pu) NFC with multi-isotope (^{232}U-^{233}U-^{234}U-^{235}U-^{236}U-^{238}U) uranium fraction containing practically all uranium isotopes. As a consequence, the NFC "center of gravity" can be shifted from the currently used ^{235}U towards the use of ^{233}U in the future.

The "light" uranium fraction, in addition to content of very attractive fissile uranium isotope ^{233}U, can offer the following significant feature from nuclear non-proliferation point of view. If the "light" uranium fraction would be mixed with uranium regenerate extracted from spent LWR fuel, then some kind of low-enriched uranium fuel could be produced for nuclear power reactors. This is a very auspicious factor for weakening the problem of unauthorized proliferation of fissile materials. By the way, radioactivities of three light uranium isotopes (^{232}U, ^{233}U and ^{234}U) are more intense than those of other uranium isotopes. The more intense radioactivity of such uranium fuel can additionally enhance its proliferation resistance.

When natural ^{230}Th thorium is irradiated by high-energy neutrons, the lighter nuclides ^{231}Pa and ^{230}Th [2, 3] are generated in addition to the "light" uranium fraction (^{232}U, ^{233}U, ^{234}U). Protactinium isotope ^{231}Pa can be directly extracted from neutron-irradiated thorium with application of radiochemical technologies. Under further neutron irradiation light thorium isotope ^{230}Th captures neutron, converts to ^{231}Pa that can be also extracted from neutron-irradiated thorium.

It would be wrong to estimate build-up of ^{231}Pa under neutron irradiation of thorium as a generation of useless by-product and neutron poison. ^{231}Pa, being admixed to fresh uranium fuel of nuclear power reactors, can initiate the chain of isotopic transformations ^{231}Pa $\rightarrow$ ^{232}U $\rightarrow$ ^{233}U. As a result, ^{231}Pa-bearing fuel acquires a new valuable quality, namely stable neutron-multiplying properties under high and ultra-high values of fuel burn-up [3]. The opportunity arises to provide long and ultra-long fuel lifetimes, strengthen autonomy of nuclear energy sources and heighten export potential of nuclear power technologies.

Involvement of natural thorium into the closed NFC can promote resolving some principal problems with which nuclear power will face in the foreseeable future. Some of these problems have become apparent already, some other bottlenecks of large-scale NPS with the closed NFC are already foreseen.

There are three, at least, difficulties in further development of global nuclear power. The first problem is a necessity to reduce scope of operations in back-end NFC part via substantially higher values of fuel burn-up in nuclear power reactors. The second problem is a necessity to enhance NFC protection against unauthorized proliferation of fissile materials and, thus, heighten export potential of nuclear technologies. The third problem is a necessity to make radioactive wastes harmless via neutron

transmutation of long-lived fission products and minor actinides, i.e. their conversion into short-lived and stable isotopes. At present, these problems produce only negative effects on public opinion concerning the future destiny of large-scale nuclear power technologies.

3. Specific features of thorium resource problem in Russia

The following two specific features are usually mentioned in the connection with involvement of thorium, one else natural fertile material in addition to uranium, into the NPS. The first feature consists in the necessity of developing other fuel technologies as compared with the existing uranium technologies due to quite different material properties of thorium and uranium. The second feature is related with thorium mining and primary processing. The question arises: up to what scales the industrial plants on mining and primary processing of natural thorium ores should by deployed? Generally speaking, till now no one country in the world has developed thorium technologies up to an industrial and, therefore, very expensive scale.

Current applications of natural thorium are limited by its usage in chemical and metallurgical industries, in electronics and medicine [4]. According to various publications and evaluations, total annual consumption of natural thorium in the world covers the range from several dozens of tons up to 200–300 tons.

Thorium is being mined now mainly as a by-product of rare-earth elements. Thorium is extracted from mineral ores during their primary processing and stockpiled for potential applications in the future. Besides, thorium is a by-product of mining and primary processing of uranium and mixed uranium-thorium ores [5–7].

However, when considering the problem, some peculiarities in mining, primary processing and energy utilization .of natural uranium and thorium resources should be taken into account. Large scale of uranium mining industry is mainly defined by low content of the only natural fissile isotope ^{235}U in uranium (~0.71%). About 200 tons of natural uranium must be mined in order to produce one ton of ^{235}U with application of isotope separation technologies.

Since effective fissile isotope, analogue of ^{235}U in natural uranium, is absent in natural thorium, thorium involvement into the closed NFC does not require the same scale of thorium mining industry. Scale of thorium mining industry may be defined by thorium consumption needed to produce large enough amount of well-fissile isotope ^{233}U. Anyway, scale of thorium mining industry is mainly defined by the NFC structure and by strategies of thorium utilization.

The following simple assumptions may be used to evaluate minimal thorium consumption in the closed NFC. As is known, one typical LWR with electric power of 1 GWe incinerates annually about 960 kg of fissionable materials. These 960 kilograms include ~580 kg ^{235}U (i.e. about 60%). If useless transformation of ^{235}U into ^{236}U via radiative neutron capture is taken into account, then total amount of the eliminated ^{235}U may be evaluated as ~800 kg ^{235}U/(GWe·year).

Total power of Russian nuclear power plants is planned to be at the level of 50-60 GWe in the middle of the XXI century. If ^{233}U is substituted for ^{235}U, then, under the same other conditions, thorium consumption for production of ^{233}U will be laughably low, at the level of 40-50 tons annually. So small value of thorium consumption will require no additional efforts on exploration of new thorium deposits and deployment of thorium mining plants in Russia.

Nowadays, available stockpiles of thorium-bearing monazite concentrate (82 000 tons with mean thorium content of 4.7%, i.e. ~3800 tons of natural thorium, [8]) are stored in the vicinity of Krasnoufimsk (Sverdlovsk Region, Russia). Even so modest resources of natural thorium are large enough to satisfy demands of the large-scale Russian NPS (50-60 GWe in the middle of century) during the next 70 years.

In addition to thorium consumption for ^{233}U generation in nuclear power reactors, it is necessary to evaluate thorium amounts needed to fill up blankets of hybrid fusion facilities, and thorium amounts stored at spent fuel reprocessing plants. As is known, radiochemical reprocessing of irradiated fertile materials can be performed without cooling-time at all, as it was shown for hybrid fusion facility with

molten-salt blanket [9]. This means that thorium amounts stored at reprocessing plants can be evaluated as negligibly small.

As to thorium amounts needed to fill up thermonuclear neutron source (TNS) blankets, they can be sufficiently large. Some relevant information was taken from the project of pilot hybrid fusion facility with uranium blanket developed by the National Research Center – Kurchatov Institute in the 1980s as a plutonium-production plant [10]. Metal uranium blanket (20-cm thick) was placed just behind the first wall of plasma chamber, and contained molybdenum alloy with depleted uranium. Main parameters of the fusion facility: thermonuclear power – 578 MWt; neutron load on the first wall - 0.8 MW/m^2; plutonium production rate - 4.0 tons per year. Total uranium amount loaded into the TNS blanket was evaluated as 1040 tons.

If the same thorium amount is taken to load into the TNS blanket, then currently available resources of natural thorium (~3800 tons) with accounting for thorium consumption for ^{233}U production (~40-50 tons annually) are large enough to provide operation of two TNS for about 30 years. Afterwards, either additional thorium mining or thorium purchasing will be required.

However, these evaluations are correct only for the case when no thorium amounts are introduced into fresh fuel compositions of nuclear power reactors. Thorium must be loaded into blankets of highly efficient producers of ^{233}U. Just this variant of Russian NPS structure may be realized, if conceptual design of hybrid TNS with thorium blanket will be worked out. Then, involvement of so effective producers of ^{233}U (even at relatively low number of these producers) into the closed NFC can provide Russian NPS with artificial ^{233}U-based fuel for a very long time period. In this case, multi-isotope uranium compositions with dominant content of ^{233}U and ^{238}U can be used as a fuel of thermal nuclear reactors.

Nuclear nonproliferation problem is considered in [11]. If several percents of ^{231}Pa and ^{237}Np are introduced into fresh (^{233}U-^{238}U) fuel compositions, then, under neutron irradiation in nuclear power reactors, uranium isotope ^{232}U can be produced in amounts comparable with amount of ^{233}U via the following chain of isotopic transitions ^{231}Pa(n,γ)^{232}Pa(β)^{232}U [12]. Similarly, plutonium isotope ^{238}Pu can be produced in amounts comparable with amount of ^{239}Pu via the chain of isotopic transitions ^{237}Np (n,γ)^{238}Np(β)^{238}Pu [13]. Both these isotopes (^{232}U and ^{238}Pu) are able to enhance proliferation resistance of fissile, weapon-usable materials.

4. Conclusion

The paper analyzes potential involvement of hybrid fusion facilities with thorium blanket into the closed (U-Pu) NFC of future Russian NPS for production of the "light" uranium fraction (^{232}U-^{233}U-^{234}U) and ^{231}Pa that can lead to the formation of advanced (^{231}Pa-^{232}Th-^{232}U-^{233}U-^{234}U-^{235}U-^{236}U-^{238}U-Pu) fuel cycle. The following main results are listed below:

1. Nuclear fuel cycle will be shifted from fissile isotope ^{235}U towards ^{233}U, more attractive isotope for thermal nuclear reactors.
2. The "light" uranium fraction is the most proliferation-resistant fuel component. The "light" uranium fraction, being mixed with uranium regenerate, becomes some kind of low-enriched uranium, i.e. proliferation resistance can be additionally strengthened.
3. Introduction of ^{231}Pa into fresh fuel compositions can stabilize their neutron-multiplying properties and, thus, can create conditions for ultra-long fuel lifetimes, upgrade autonomy of nuclear energy sources and heighten export potential of nuclear power technologies.
4. Natural thorium resources currently available in Russia are large enough to provide operation of large-scale (50-60 GWe) Russian NPS for about 30 years under assumption that thorium is loaded into blankets of hybrid TNS only.

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