

SOME RESEARCH AND DEVELOPMENT OF THORIUM FUEL CYCLE IN RUSSIA*

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Abstract. There has recently been increasing interest in implementation of thorium into nuclear power. Among the new areas of that interest research in the field of nuclear fuel resources, safety improvement of nuclear reactors and their ecological acceptability should be mentioned. Many specialists consider the proliferation resistance to be the main advantage of thorium fuel cycle. In particular, this advantage leads to some proposals to use thorium-plutonium-based reactor fuel for plutonium utilization. This paper presents an overview of investigation, which are under way at the State Scientific Center - Institute of Physics and Power Engineering in Obninsk, Russia.

1. INTRODUCTION

Discussed in this paper are some researches on thorium fuel cycle which are always supported in IPPE at least a little. The authors do not intend to make a comprehensive overview of the problem but present their personal vision of the problem as well as the way how to solve it.

2. ^{232}U ISSUE

First experiments on ^{233}U accumulation in thorium irradiated in graphite-moderated reactor revealed some problems with ^{233}U handling. The decay of ^{232}U accumulated together with ^{233}U during irradiation leads to formation of some daughter isotopes, which are the source of intensive γ -radiation. ^{233}U can be worked with for a rather long time in glove boxes only if the presence of ^{232}U in thorium is less than 1 g/kg thorium, the nuclear content of ^{232}U being about 5 ppm in this case. If ^{232}U content is more than 10 ppm, either frequent extractions of decay products (approximately within 4 months) or installation of a remote control equipment is required for safe handling with irradiated materials.

At the initial stage of ^{233}U -Th fuel cycle implementation pure ^{233}U would be better to use for manufacturing samples and targets, experimental fuel pins and subassemblies. However huge amounts of ^{233}U "contaminated" by ^{232}U would appear. For example, ^{233}U with ^{232}U content of 2000-3000 ppm would be produced in a core of a conventional PWR with thorium for typical fuel burnups.

The production of pure ^{233}U can be started in fast reactors with rather favorable economics characteristics of nuclear power plant (NPP). Thus, in the thorium blanket of fast reactor the hard component of neutron spectrum is cut at the distance of 15-20 cm from the core, so ^{233}U accumulation in thorium of ~ 2.5 g/kg would correspond to the content of ^{232}U a few ppm. Compared to thermal reactors, utilization of thorium in fast reactors does not require a short irradiation time and special reloading schemes of thorium subassemblies.

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For example, in the BN-800-type fast reactor with a thorium radial blanket it is possible to extract the following quantities of ^{233}U per year from different rows (kg): 82 (42 ppm of ^{232}U), 48 (11 ppm), 30 (3 ppm), 16 (0.7 ppm).

There have been some experiments on BN-350 fast reactor where the thorium samples were irradiated in the radial blanket up to ^{233}U accumulation in thorium of 1.3 g/kg. The ^{232}U content was in the range of 2-11 ppm depending on the position. It is worth noting here that the content of ^{232}U in thorium samples placed in the whole uranium blanket is higher than in the whole thorium blanket.

Among the experiments on ^{232}U problem we should mention here laboratory works on ^{233}Pa extraction from irradiated thorium. ^{233}Pa decay leads to formation of pure ^{233}U . These experiments require a quick delivery of irradiated thorium to the radio-chemical laboratory. IPPE possesses approximately 100 mg of ^{233}U extracted from irradiated thorium samples obtained as a result.

Some specialists think it's possible to make the isotope separation of ^{232}U from ^{233}U using a centrifuge process. Obviously, in case of a significant progress of works on thorium fuel cycle trade-off evaluations between the two fuel cycle technologies would be required: either to work with highly radioactive fuel with a special remote control equipment or to use rather simple equipment but with preliminary separated ^{232}U from ^{233}U .

3. THORIUM FUEL CYCLE TECHNOLOGIES

No special problems are expected in manufacturing technology of mixed oxide thorium-uranium (or plutonium) pelletized fuel. High quality experimental fuel pellets of mixed (U-Th) O_2 were manufactured at both IPPE and other research laboratories.

Highly thermal-conductive dispersion fuel compositions characterized by lower temperature in fuel pins and lower accumulated internal heat are very attractive from the point of view of reactor safety.

Two types of fuel compositions for WWER-type reactors: UO_2 (60%)-Zr (40%) alloy and UO_2 -Al alloy are under investigation and have successfully stood complex tests [1]. The authors suppose oxide UO_2 can be easily substituted by a mixture of UO_2 - ThO_2 . An other unconventional dispersion composition with pyrolytic expanded graphite is under consideration [2]. Technological aspects of these compositions with uranium oxide have already been tested but more complex experiments including reprocessing are required.

The principal technology of ^{233}U extraction from irradiated in thermal reactor samples was tested in IPPE. The cooling period was approximately 3 years. Uranium water extraction method used resulted in 99.4% uranium extraction ratio. The fission product (FP) purification coefficient per one extraction cycle was more than 1000. In a separate reprocessing cycle thorium was extracted from the mixture of Th-FP with the extraction ratio of more than 99% and purification coefficient of 100.

Samples of ThO_2 irradiated in the radial blanket of BN-350 were used to investigate the dissolution process. The technologically accepted rate of ThO_2 dissolution was achieved when the fluoric acid was added to the nitric acid (0.1 mole/liter).

All these experimental results together with some reference data show the principal feasibility of the closed thorium fuel cycle based on water extraction.

Some proponents of dry reprocessing methods advocate the complex technology combining electrochemical reprocessing in salts resulted in oxide grains with subsequent vibropacked technology for fuel manufacturing.

4. SOME REACTOR CONCEPTS UTILIZING THORIUM FUEL CYCLE

4.1. Lightwater reactors with thorium.

As the initial stage of thorium implementation, IPPE proposed utilization of highly enriched uranium (HEU) resulted from weapon disarmament [3], which could help to avoid a special uranium enrichment required for thorium cycle initiation.

This proposition was considered for WWER-1000-type reactor with the dispersion fuel based on the metal thorium matrix.

The annual ^{235}U consumption due to ^{233}U breeding is lower for the considered reactor type compared to the conventional WWER-1000 with UO_2 fuel (735 and 940 kg per year, respectively). The number of consumed subassemblies is 1.8 times lower in the former case. ^{233}U is effectively bred (350 kg/year) instead of plutonium production (250 kg/year).

Safety characteristics of the thorium fuelled reactor such as feedback reactivities, burn-up reactivity swing and reactivity required for compensation during the heating are better. Also the value of energy accumulated in the fuel is lower in this kind of a reactor.

Reactors WWER with $(\text{Th-Pu})\text{O}_2$ fuel have been under study due to the problem of weapon grade plutonium (WG-Pu) utilization [3,4]. As an example, WWER-1000 reactor with mixed oxide thorium and weapon grade plutonium subassemblies in 1/3 of the core was considered. The main reactor hardware was unchanged compared to the conventional WWER.

The annual consumption of WG-Pu in WWER-1000 with the above mentioned core is 355 kg while of the same reactor with 1/3 of the core MOX-fuelled VVER-1000 is only 270 kg. After burn-up of 40 $\text{MWt}\times\text{days/kg}$, the amount of plutonium in subassemblies is 1/3 compared to the initial content. The content of ^{239}Pu in discharged subassemblies with $(\text{Th-Pu})\text{O}_2$ is less than 30%. Thus, only one irradiation cycle in such a reactor is required for all loaded WG-Pu to lose its weapon grade quality completely.

Safety characteristics of the considered reactor are practically the same as for WWER with 1/3 MOX-fuelled subassemblies. The control rod system is unchanged.

Since reactors with $(^{233}\text{U-Th})$ fuel will be implemented only in the future, the core layout of this reactor is not necessarily be the same as in existing WWERs. For $(^{233}\text{U-Th})$ fuel cycle a balance should be found for ^{233}U breeding, safety specific power rating and fuel burn-up. Compared to the existing generation of PWRs, the inherent safety of the new generation of reactors should be higher.

The results of calculations showed the advantage of tighter fuel pin lattices with lower water volume ratio and at least two types of subassemblies in the core. These changes significantly

influence thermal-hydraulics parameters of a reactor, its reactivity characteristics and will require intensive efforts for design developments.

4.2. Mixed fuel cycle with ^{233}U and plutonium breeding in fast reactors

The idea of this cycle was proposed and developed in IPPE [5]. ^{233}U , plutonium and ^{238}U are used in the core of a fast reactor. Thorium is considered only as fertile material of a radial blanket. ^{233}U with ^{238}U is loaded into the inner core and plutonium with ^{238}U – into the outer core. As a result ^{233}U is bred only in blankets and plutonium - only in the core. The material for axial blankets should be chosen for any particular case.

Separated loading of ^{233}U and plutonium into the core gives some advantages:

- Having the same value of Doppler reactivity coefficient, sodium void effect has a significant negative component and as whole can be negative.
 - The β_{eff} in the case of fertile ^{238}U is larger than that in the case of fertile thorium.
 - The protactinium reactivity effect can be eliminated by thorium loading in blankets.
 - The content of ^{232}U in ^{233}U is the lowest as compared with any other reactors.
- Authors would like to attract the attention to this problem for further discussion.

4.3. U-233 for space reactors

The HEU-fuelled “TOPAZ” reactors with thermoionic direct conversion of energy have successfully been tested in space [6]. The thermoionic elements fuel inventory can be reduced in case of ^{233}U employment. This advantage can be used for increasing reliability of fuel elements with higher porosity and thicker cladding. As a result, the reactor lifetime can be significantly prolonged, which is a very important figure-of-merit for space reactors. Some preliminary results showed that 7-10 years of reactor lifetime could be achieved utilizing ^{233}U fuel.

However, the acceptable radiation condition at sites of fuel elements manufacturing, reactor assembling and launch is a provision for successful utilizing ^{233}U in space reactors.

Thermoionic elements manufacturing is a very complicated technology, absolutely excluding high radiation background. According to some evaluations, small contents of ^{232}U in ^{233}U (4-5 ppm) and a short technological cycle provide acceptable radiation conditions for the personnel. As is mentioned above, the production of ^{233}U with this content of ^{232}U is possible.

5. NUCLEAR DATA FOR THORIUM FUEL CYCLE

Increased interest to thorium fuel cycle has recently become the motivation for comprehensive nuclear data provision evaluations, similar to those existing for uranium-plutonium fuel cycle. These evaluations has been made at IPPE [7], where the detailed requirements for various cross sections measurement accuracy and comparisons with the obtained ones are presented. The fulfillment of the requirements will probably take a long time and obviously, the work activity will depend on the development of thorium cycle itself.

It should be mentioned that the present day works on thorium fuel cycle are supported either on the concept level or on the level of a particular problem and the existing nuclear data are

quite sufficient. Two examples show it. First, the uncertainty in evaluation of K_{eff} for systems with ^{233}U is higher (1.0%) than for plutonium-systems (0.5%) but is quite enough. Second, ^{232}U generation is evaluated nowadays with not less than 20% uncertainty. 10% uncertainty would be desirable but the values of thorium inelastic scattering and (γ, n) cross section on ^{231}Pa are to be studied better.

At present integral experiments support the studies of thorium cycle [8,9].

Thus, multiplication properties of media containing thorium, HEU and hydrogen were studied on COBRA critical installation at IPPE. Four assemblies with different composition central regions had K_{∞} of these regions close to unity. The value of average absorption cross section of thorium was determined from the neutron balance conditions. The correction on heterogeneity is the main interpretation problem.

Two other assemblies of the same facility were used for the evaluation of critical parameters of conventional cores. The materials of the one were thorium and enriched uranium, the other one contained the same material and hydrogen. Preliminary results of this set of critical experiments showed 3% uncertainty in thorium absorption cross section.

The ratios of average cross sections to the fission cross section of ^{235}U were determined and the value of thorium absorption cross section was confirmed.

Various samples were irradiated in the core of BN-350 fast reactor. Many samples have not been investigated yet, but some of the results related to the thorium fuel cycle have already been obtained.

- The average uncertainty of 3% thorium absorption cross section is confirmed;
- The sum of $(n, 2n)$ and (γ, n) cross sections for thorium with 5% experimental uncertainty corresponds to data usually taken;
- The data of ENDF/B-V library for (γ, n) cross section on ^{231}Pa are higher ($50 \pm 5\%$) compared to experimental results.

All this work is continued and the efforts will be increased depending on the financial support.

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