USE OF THORIUM IN THERMAL-NEUTRON REACTORS: DIFFERENT TYPES OF FUEL COMPOSITIONS IN VVER-1000 REACTOR CELL

A. NAYMUSHIN *, YU. CHERTKOV, I. LEBEDEV, M. ANIKIN AND S. SAVANYUK

National Research Tomsk Polytechnic University (TPU), 634050, Tomsk, Russian Federation

ABSTRACT

This work is dedicated to exploring possibilities of using alternative thorium-based fuel compositions in VVER-type reactor. The article describes comparison between standard uranium dioxide fuel and perspective fuel composition: thorium dioxide, mixed thorium and plutonium dioxides. The calculation result show that using of perspective fuel composition caused to improving of physical and economical characteristics of reactor core. Furthermore, investigated fuel provide effective burnup, that increase fuel cycle length. Moreover, growth of using fertile isotopes is shown in comparison with standard fuel.

INTRODUCTION

The explored resources of thorium in the earth crust are several times higher than the reserves of uranium and this significantly increases nuclear energy's resource base in case of implementing closed nuclear fuel cycle.

30 isotopes of thorium are known as well as 3 excited metastable states of its nuclides. Only one isotope of thorium (thorium-232) has sufficiently long half-life with respect to the Earth's age, therefore, almost all of natural thorium consists of this nuclide only. Th-232 itself is not fission by thermal neutrons, but absorption of neutron by thorium-232 leads to generation of uranium-233, which has high probability to emit neutrons as a result of fission by thermal and intermediate neutron fluxes. Therefore, it plays the same role in a nuclear reactor as U-238: nuclides absorbing neutrons transform into secondary nuclides, which can fission by thermal neutrons (Alekseev, *et al.*, 2011).

Thorium-232 is the best "resource" isotope compared with uranium-238 for the reactors with a thermal neutron spectrum. Uranium-233 emits more than two neutrons per capture of one primary neutron for wide range of reactors with a thermal neutron spectrum.

Thorium dioxide has greater chemical and radiation resistance compared with uranium dioxide, as well as better thermal-physical properties (thermal conductivity, linear expansion coefficient).

Secondary nuclear fuel in thermal-neutron reactors is two isotopes of plutonium: Pu-239 and Pu-241. The former is generated as a result of absorption of thermal and resonance neutrons by U-238 nuclei, the latter is generated as a result of double radiative capture of neutrons by Pu-239 nuclei.

When transitioning from uranium to thorium fuel cycle, production rate of long-lived minor actinides significantly decreases in a nuclear reactor. If thorium reactor operates exclusively in a U-Th fuel cycle, actinides with masses higher 237 will accumulate in negligible amounts in the reactor (Boyko, *et al.*, 2006).

Isotope U-232 draws special attention in thorium-based cycle. It is generated by means of (n, 2n) reaction taking place on Th-232, Pa-233 and U-233 isotopes. Half-life of U-232 is 69 years. Among its daughter products are Tl-208 – isotope with very short lifetime emitting hard gamma particles (2.6 MeV).

Dose rates in thorium fuel will rise due to accumulation of U-232. This creates additional problems when dealing with spent nuclear fuel of thorium reactors, in particular, when re-cyclization

*Corresponding author's email: agn@tpu.ru

of uranium takes place. However, at the same time, presence of U-232 is spent fuel increases proliferation security of this nuclear cycle.

Besides, thorium fuel cycle is more preferable with respect to utilizing weapon-grade plutonium since it does not lead to its reproduction as in case of using U-Pu fuel cycle (Lung and Gremm, 1998).

RESULTS AND DISCUSSION

Let us consider the possibility of using thorium in VVER-1000 thermal-neutron reactors. For this purpose, let us compare multiplication properties of this reactor cells with different types of fuel, including the fuel containing thorium (Galperin, *et al.*, 1997).

To implement thorium fuel cycle it is required to use (at least during the initial loading) traditional fissionable isotopes of uranium and/or plutonium. As such, the following can be chosen:

- Highly enriched uranium;

- Weapon-grade plutonium;

- Reactor-grade plutonium, obtained from irradiated nuclear fuel with high burn-up.

- For fuel composition:

- Traditional uranium, plutonium and thorium dioxides;

- Promising nitrides (carbides) of these nuclides, which has significantly higher density than dioxides.

Computation model is an elementary cell of the VVER-1000 reactor with infinite height, consisting of a fuel element surrounded by water coolant. During the computation, real hexagon cell is replaced by equivalent cylindrical cell with cross-section of a real one (Fig. 1).
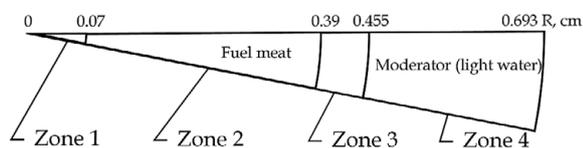

Fig. 1 Computation model.

Fuel element's core (zone 2) with a radius of 0.39 cm has inner hole with a diameter of 0.07 cm (zone 1). Zone 3 represents fuel element's cladding; zone 4 represents water coolant (and moderator).

Fuel element's cladding of the VVER-1000 reactor is made from zirconium alloy with the outer diameter of 0.91 cm and thickness of 0.65 cm. Fuel pellet made from uranium dioxide has the outer diameter of 0.78 cm and axial hole with the diameter of 0.14 cm.

Placement step of fuel elements is 1.275 cm.

Material composition of the basic description of fuel elements is represented by uranium dioxide with density of 10.5 g/cm³ and 4.5% U-235 enrichment. Cladding is made from zirconium alloy with 1% niobium. Water density corresponds to pressure of 16 MPa and temperature of 300°C (Vladimirova, *et al.*, 2012).

Fuel cycle with uranium dioxide

Fig. 2 shows dependencies of the infinite multiplication factor (k_{inf}) for the major Russian fuel for power reactors — uranium dioxide — with 5%, 10% and 20% enrichment in the elementary cell of the VVER-1000 reactor.
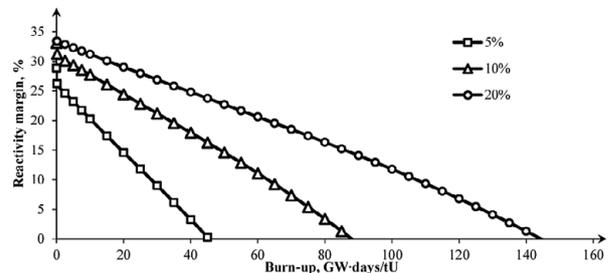

Fig. 2 Multiplication properties of the VVER-1000 reactor cell with fuel of different enrichment vs. burn-up.

Infinite multiplication factor (k_{inf}) in the cell with traditional fuel — with enrichment less than 5% — drops lower than 1 at burn-up of 50 GW·days/tU. When increasing the initial enrichment up to 10% to 20% with respect to U-235 isotope and conserving multiplication properties, the burn-up of 85 and 145 GW·days/tU can be achieved.

The bigger the burn-up, the more amount of U-238 is involved in reactor operation. When using the fuel with less than 5% enrichment, no more than 3.8% of U-238 is involved in the fuel cycle. However, using the fuel with 20% enrichment can lead to both higher values of burn-up achieved and more U-238 used — up to 8.6% (see Fig. 3).
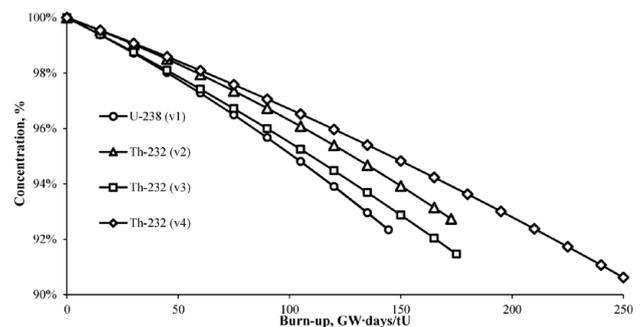

Fig. 3 Relative change of nuclei concentration of reproducing nuclides vs. achieved burn-up.

Fuel cycle with uranium and thorium dioxide

To fully appreciate all the benefits of thorium compared with U-238, we will use uranium with the maximum enrichment (90%) so that to minimize the content of this isotope. To identify the effect of replacement of U-238 with thorium, uranium dioxide content is chosen such that the concentration of U-235 in the mixture is equal to that of which was in the previous case with 20% enrichment.

Fig. 4 shows infinite multiplication factor (k_{inf}) for the fuel of uranium dioxide with 90% enrichment in the elementary cell of the VVER-1000 reactor.

With the same loading of the fuel element with respect to U-235, the replacement of U-238 with Th leads to decrease of the initial reactivity margin from 34.2% to 33.3%, but the length of the operating cycle increases from 145 to 175 GW·days/tU.

When using this fuel, 8.2% of thorium-232 is involved in the fuel cycle (see Fig. 3), which is somewhat less than in a case of using traditional fuel. Herewith, ~36% of U-238 is involved as well.

At the end of the operating cycle, there is $3.3 \cdot 10^{20}$ cm^{-3} of Pu-239 contained in the fuel cycle with uranium dioxide while in the fuel cycle with thorium the concentration of U-233 reaches up to $5.3 \cdot 10^{20}$ cm^{-3}.

Fuel cycle with plutonium dioxide and thorium dioxide

Isotope composition of plutonium significantly depends of the method of its production: weapon-grade plutonium or reactor-grade plutonium produced during recycling of spent fuel of the VVER reactor. Table 1 shows compositions of plutonium used during computations.

Table 1. Isotope compositions of plutonium in the fuel (Deen, *et al.*, 2004)

| Isotope	Content in weapon-grade plutonium, %	Content in reactor-grade plutonium, %
Pu-238	1.2	2.4
Pu-239	93.8	58.4
Pu-240	5.8	24.0
Pu-241	0.35	11.2
Pu-242	0.022	3.9

To determine advantages of different types of fuel, concentration of fissionable nuclides were chosen such so that to provide approximately equal reactivity margins during computations. When using the fuel based on weapon-grade plutonium or thorium, the value of achieved burn-up is 175 GW·days/tU (with 25% of dioxide plutonium in the mixture) (Halsall, 1996). Fig. 5 shows multiplication properties of the cell vs. burn-up.

When using fuel with weapon-grade plutonium, reactivity margin is 33.4% at the beginning of the reactor operating cycle. Thorium-232 involvement in the fuel cycle rises up to 8.5% (see Fig. 3) and nuclear concentration of U-233 $8.9 \cdot 10^{20}$ cm^{-3} at the end of the operating cycle.

When using reactor-grade plutonium, to provide reactivity margin similar to the previous cases, it is required to raise the content of plutonium dioxide up to 45%. In this case, the initial reactivity margin is 34.4% (see Fig. 6).
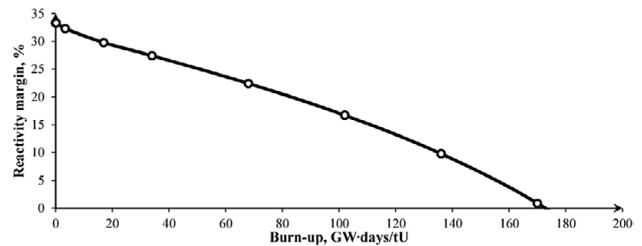
Fig. 4 Multiplication properties of the VVER-1000 reactor cell with UO_2+ThO_2 fuel vs. burn-up.
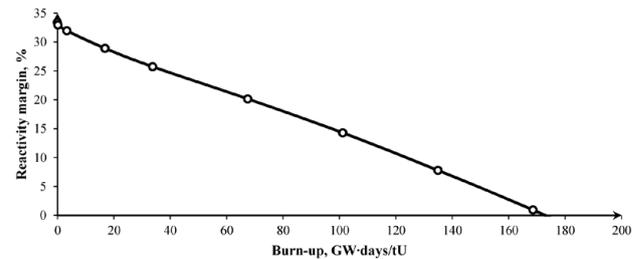
Fig. 5 Multiplication properties of the VVER-1000 cell using weapon-grade plutonium (PuO_2+ThO_2) vs. burn-up.

In the case of using this type of fuel, the burn-up significantly increases (up to 250 GW·days/tU). Such increase is caused by high concentration of even isotopes of plutonium and high initial concentration of Pu-239, which transforms into fissionable nuclides after a neutron capture.

To understand which of the variants better uses thorium, the following variants of fuel compositions (with the same reactivity margin at the beginning of the operating cycle) were considered:

- v1–initial variant of fuel with UO_2 used in the VVER reactor but with 20% enrichment;

- v2–mixture of UO_2 with ThO_2;

- v3–mixture of ThO_2 with PuO_2, which uses weapon-grade plutonium;

- v4–mixture of ThO_2 with PuO_2, which uses reactor-grade plutonium;

Major criteria of thorium involvement in energy release are decrease of thorium concentration during reactor operation and achieved energy release. Fig. 3 shows computation concentrations of nuclei of reproducing nuclides vs. achieved burn-up for these variants.

The variant with reactor-grade plutonium shows the best results. Thorium-232 involvement is 9.4% at burn-up of ~250 GW·days/tU. Nuclear concentration of U-233 at the end of the operating cycle in this case is $8.4 \cdot 10^{20}$ cm^{-3}.
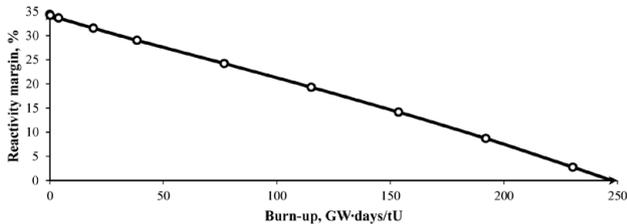

Fig. 6 Multiplication properties of the VVER-1000 reactor cell using reactor-grade plutonium (PuO$_2$+ThO$_2$) vs. burn-up.

CONCLUSION

All the considered variants of using thorium-containing fuel show better value of the achieved burn-up than in case of using pure uranium dioxide. For the mixture of reactor-grade plutonium with thorium dioxide, the burn-up is ~1.7 times higher.

Even better results can be achieved when using carbides and nitrides of uranium and thorium (instead of its oxides), which, compared to the oxides, have significantly higher density and this may allow using uranium with less enrichment and plutonium with less volume content for the initial loading.

ACKNOWLEDGMENT

REFERENCES

Alekseev, N.I. and Gomin, E.A. 2011. MCU-PTR program for high-precision calculations of pool and tank type research reactors. *At. Energy.* 109: 149.

Boyko, V.I., Vlasov, V.A., Zherin, I.I., Maslov, A.A. and Shamanin, I.V. 2006. Thorium in nuclear fuel cycle. Ruda I Metally Publishing. Moscow.

Deen, J.R., Woodruff, W.L., Costescu, C.I. and Leopando LS. 2004. *WIMS-ANL* User Manual. Rev.6. ANL/TD/TM99-07. Argonne National Laboratory. Argonne.

Galperin, A., Reichert, P. and Radkowsky, A. 1997. Thorium fuel for light water reactors--reducing proliferation potential of nuclear power fuel cycle. *Sci. Global Secur.* 6: 265-290.

Halsall, M.J. 1996. WIMSD A neutronics code for standard lattice physics analysis, distributed by the NEA Databank. AEA Technology. Harwell.

Lung, M. and Gremm, O. 1998. Perspectives of the thorium fuel cycle. *Nucl. Eng. Design.* 180: 133-146.

Vladimirova, S.V., Zyabkin, M.V., Klochko, G.G., Koryagin, S.L., Levchenko, V.M., Merzlikin, V.G., Khabarov, V.S., Shevchenko NN. and Novikov, M. 2012. Export control. GNC RF-FEI. Obninsk.