Investigation of using U-233 in thorium base instead of conventional fuel in Russian PWR by SERPENT Code

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Abstract. In this work, the fuel assembly of type TVS-2006, which is used in Russian reactor VVER-1200, is considered. A proposed fuel of Thorium and Uranium-233 ((Th\textsuperscript{232}+U\textsuperscript{233})O\textsubscript{2} has been investigated to replace the fuel currently used (U\textsuperscript{238} as a fertile and U\textsuperscript{235} as fissile). This has been done in order to compare the; conversion ratio, reproduction factor (\(\eta\)), burnup, effective delayed neutrons fraction (\(\beta_{eff}\)), Minor Actinides (MA) inventory, and Temperature coefficients of reactivity. The calculations performed using the Monte Carlo code (SERPENT-2.31) to analyze and compare the two fuels. Results showed that using U-233 in the Thorium base will considerably increase the reactivity at the Beginning Of Cycle (BOC), increase the conversion ratio, and burn up. Besides, the safety parameters for the proposed Th-based fuel, in general, lower than that for Low Enriched Uranium (LEU) but still acceptable.

1. Introduction
There is no doubt that nuclear energy is one of the best solutions to face the catastrophe of global warming and change the earth’s atmosphere. One of the most critical problems facing the development of nuclear energy is nuclear wastes, which includes the hazards of long-lived isotopes and proliferation risks [1, 25].

Many countries started to study the Thorium fuel cycle to sharing with [2] or as an alternative to currently used cycles [3]. Thorium fuel has many advantages over Uranium fuel as:

- The more abundance of Thorium.
- Lower radioactive wastes from the extraction process compared to Uranium.
- More possibility of conversion for Th-232 over U-238.
- ThO\textsubscript{2} is chemically stable and has a high radiation resistance, even more, UO\textsubscript{2}, so that it can achieve more burn up than UO\textsubscript{2}.
ThO$_2$ has a higher thermal conductivity and a lower coefficient of thermal expansion than UO$_2$.

ThO$_2$ fuel is lowering the released rate of Pu and fission products as Am and Cm than UO$_2$ fuel, so considered against the proliferation of nuclear weapons more than UO$_2$ [4].

To save the Uranium resources, decrease the radio-toxicity of spent fuel, and resist the dangers of nuclear proliferation, a worldwide interest for the thorium fuel cycle increased since 1960 [4].

Both U-238 and Th-232 are considered fertile materials because they absorb neutrons and turn into fissile materials (Pu$^{239}$ and U$^{233}$, respectively). Besides, U-233, U-235, and Pu-239 are considered fissile materials, because they possess fission with absorbing a neutron. Th-232 is considered better fertile than U-238 because of the high absorption cross section of Th-232 than U-238. U-233 is considered better fissile because it has lower capture cross section than U-235 and other fissile nuclides.

Thorium can be used in a closed or opened fuel cycles. The main difference between both cycles is the chemical reprocessing of irradiated Th-based fuel for separating the bred U-233. After the chemical reprocessing, the collected fissile isotope U-233 can be fed again as a part of the fuel to another reactor to close the fuel cycle. In the open fuel cycle, the bred U-233 is burned in the site and does not reprocess. The main advantage of Th open fuel cycle is to avoid dealing with highly radiotoxic U-233.

The conversion of Th to U$^{233}$ can occur in both thermal and fast reactors. Among these reactors, the high-temperature gas-cooled reactors (HTGR), light water breeder reactors (LWBR), pressurized heavy water reactors (PHWRs), liquid metal cooled fast breeder reactors (LMFBR) [5], supercritical water reactors (SCWR) [6, 7], and molten salt breeder reactors (MSBR) [4, 8, 9]. In India at BARC, high purity U-233, which contains less than five ppm U-232, was successfully obtained starting from the year 1970 [4].

The Russian reactor VVER-1200 is one of Generation III+ reactors. It is the evolution of the well-proven VVER-1000 with an increase in thermal power and additional passive safety features.

Although some works studied the use of Thorium in vver-1200 [10], none has reported specifically the performance (advantages and disadvantages) of the fissile U-233 and fertile Th-232 (Th-based fuel) in Russian VVER-1200 reactor.

This work considers one fuel assembly of reactor VVER-1200 to make a comparison between the performance of LEU and (Th-232 + U-233) fuels without any modifications of assembly and at the same fissile enrichment and working conditions.

This comparison under the same conditions should be able to provide approaches to the future optimization of fuel utilization in VVER-1200.

Section 2 introduces the main characteristics of the fuel assembly. Section 3 describes the simulation software and methodology used in the present study. Section 4 discusses the results. Section 5 gives the conclusion from this study.

2. Model description

The goal of this work is to study the replacement of conventional LEU with Th-232 + U-233 fuel in the Russian VVER-1200 reactor core. The calculations are based on a typical TVC – 2006 fuel assembly used in VVER-1200. The design parameters of fuel assembly are presented in table 1.
Table 1. Active zone and fuel assembly parameters of Russian VVER-1200 reactor [11, 12].

| Parameter                                             | value                  |
|-------------------------------------------------------|------------------------|
| Thermal / Electric Power, MW                          | 3200 / 1200            |
| Coolant temperature $T_{in}$ / $T_{out}$, °C          | 298.1 / 329.5          |
| Number of control rods                                | Up to 121              |
| Average linear power, W / cm                         | 167                    |
| Maximum linear power, W / cm                         | 448                    |
| Fuel assembly height, mm                              | 4570                   |
| Number of fuel rods in assembly                       | 312                    |
| Number of fuel assemblies                             | 163                    |
| Active fuel height, mm                                | 3730                   |
| Fuel mass in assembly, kg                             | 534.1                  |
| Measurement’s tube placement                          | Side tube (central tube replaced with fuel rod) |
| Number of grid spacers                               | 13                     |
| Type of fuel assembly                                 | TVC-2006               |
| Average clad temperature, °C                          | 355                    |
| Cladding outside diameter, mm                         | 9.1                    |
| Fuel rod pitch (center-to-center), mm                 | 12.75                  |
| The dimension of hexagonal, mm                        | 235.1                  |

Figure 1 shows the plane view and side view of the fuel assembly. A homogenous (U-233 + Th-232) O2 and (U-235 + U-238) O2 fuels were used at the same enrichment for U-233 and U-235 (4.95% w/o) without any burnable or soluble poisons.

3. Methodology and simulation software
The Powerful Monte Carlo Code SERPENT (2.1.31) [13] which has been developed at VTT over the last 15 years for various reactor physics applications, such as spatial homogenization, criticality calculations, and fuel cycle studies, was used in this study. Contentious microscopic cross-sections from library ENDF/B-VII [14] were used. 6000-neutron history per burn up step, 50 ineffective cycles, and 5000 effective cycles were used with a neutron energy range between 1e-11 MeV and 2e1 MeV to reduce the relative statistical error to roughly ±20 pcm.

The depletion was performed at a constant power of 18.00 MW (averaged power of one fuel assembly, $P_{f.a} \approx 3200/163$), and 81 nuclides are tracked in trace quantities. For better accuracy, small
burnup steps were applied at the beginning of each cycle to build in the equilibrium concentrations of most fission products isotopes. Under the above conditions, the fuel assembly was depleted for three reactor cycles. Each cycle consists of 480 effective full power days with 16-burn step for each cycle.

To describe the effect of partial reloading of the fuel assembly in the core, or burnup of one assembly in the core for three cycles, the following model was developed.

First, reloads without transposition of fuel assemblies were considered. In this case, in the core, a poly-cell element can be defined as fuel assemblies with different irradiation durations.

Secondly, in the simplest case, the multiplication factor of a poly-cell is equal to the arithmetic average of the entire set of fuel assemblies that form the cell.

In this case, the change in the infinite multiplication factor of the poly-cell with time can be represented in equation (1).

\[ K_{\text{POLY}}(t) = \frac{K_{\text{INF}}(t)+K_{\text{INF}}(t+T)+K_{\text{INF}}(t+2T)}{3} \]  

(1)

The length of the core life cycle is determined from the condition that at the end of the cycle, the average multiplication factor of the poly-cell is equal to the critical value that provides the value \( K_{\text{critical}} = 1.04 \), as shown in equation (2).

\[ K_{\text{POLY}}(t) = \frac{K_{\text{INF}}(t)+K_{\text{INF}}(t+T)+K_{\text{INF}}(t+2T)}{3} = 1.04 \]  

(2)

4. Results and discussion

The obtained results were used for comparing the performance of Thorium base fuel (\((\text{Th}-232 + \text{U}-233)\text{O}_2\)) and conventional LEU ((\(\text{U}-238 + \text{U}-235\))\text{O}_2) with equal enrichment (4.95% w/o). The calculations were performed at an assembly power equal to 1.8E+07 W.

4.1 Burn up

One of the main characteristics of fuel utilization efficiency in a reactor is fuel burnup. The easiest way to increase fuel burnup is to increase its initial enrichment. Increasing burnup in the fuel causes the increase of concentration of fission products, and heavy nuclei resulting from the radiation capture reaction.

If the cost of the fuel enrichment process were considered, expect that increasing burnup by increasing the initial enrichment of fuel is not always effective.

In this work, the discharged burnup based on three cycles of 480 effective power days for each cycle has founded to be (58,654 and 53,585 MWd/kgHM for Th and LEU fuels respectively), With a 9.45% increase in favor of Th fuel. This increase happened without any increase in enrichment, although results of [7] show that the fissile inventory (\(\text{U}-235\) in this case) must be increased by 11 to 17% in case of using thorium-based fuel to achieve the given burnup, because of the high conversion ratio of Thorium.

This increase in burnup without any increase in fissile inventory in this study because of using \(\text{U}-233\) fissile — also, a noticeable decrease in long-lived fission products and a noticeable increase in the reactor life cycle.

4.2 Infinite multiplication factor \(K_{\text{INF}}\)

Unlike [15] which compared between LEU and (\(\text{Th}-232 + \text{U}-235\)) at BOC. The \(K_{\text{INF}}\) for LEU has been higher than thorium-based fuel, in this study \(K_{\text{INF}}\) for Th-based fuel is higher as shown in figure 2 which represent the change of \(K_{\text{INF}}\) (for a poly-cell) and time.

The \(K_{\text{INF}}\) for Th-based fuel is higher than that for LEU fuel at the same enrichment due to the exchanging a fissile \(\text{U}-235\) by \(\text{U}-233\), which has lower capture cross section compared with that for \(\text{U}-235\) (capture cross section for \(\text{U}-233\) and \(\text{U}-235\) are 54 and 100 barns respectively) and more neutrons produced per neutron absorbed in fuel (\(\eta\)) or reproduction factor (\(\eta\) for \(\text{U}-233\) and \(\text{U}-235\) is 2.26 and 2.08 at thermal neutrons).
In addition, K-INF for Th-based fuel always higher than that for LEU because fertile Th-232 with capturing thermal neutrons will convert to U-233 also which will share with existing U-233 in fission. However, in LEU fuel, fertile U-238, which will convert to Pu-239 with large capture cross section (267 barns), regardless of having a high fission cross section as shown in table 2.

In addition, because Th-232 has a higher thermal capture cross section than U-238 (thermal capture cross section for Th-232 and U-238 is 7.6 and 2.7 barns respectively), so more Th-232 will convert to fissile U-233 than U-238 will convert to U-235. [15].

![Figure 2. Changing K-INF with time.](image)

**Table 2.** Basic nuclear data for Th-232, U-233, U-235, U-238, Pu-239 and Pu-241 in thermal neutron region.

| Nuclear data | Th-232 | U-233 | U-235 | U-238 | Pu-239 | Pu-240 |
|--------------|--------|-------|-------|-------|--------|--------|
| $\sigma_a$   | 4.62   | 364   | 405   | 1.37  | 1045   | 1121   |
| $\sigma_f$   | -      | 332   | 346   | -     | 695    | 842    |
| $\sigma_c$   | 7.6    | 54    | 100   | 2.7   | 267    | -      |
| $\varepsilon$| $\frac{\sigma_c}{\sigma_f}$ | - | 0.096 | 0.171 | - | 0.504 | 0.331 |
| $\eta_{th}$  | -      | 2.26  | 2.08  | -     | 1.91   | 2.23   |
| $\nu$        | -      | 2.48  | 2.43  | -     | 2.87   | 2.97   |
| $\beta$      | -      | 0.0031| 0.0069| 0.0157| 0.0026 | 0.0050 |

Where: $\sigma_a$, $\sigma_f$ and $\sigma_c$ are the absorption, fission, and capture cross-sections respectively in barns, $\eta$, $\nu$ and $\beta$ are neutron yield per fission to neutron absorbed, neutron yield per fission and delayed neutron fraction respectively.

The descending slope for Th-based fuel is slightly more than that for LEU because of the fission cross-section for U-233 (332 barns) is lower than that for U-235 and Pu-239 (346 and 695 respectively). In addition, the fission in Th-232 in thermal regions is absent because the fission cross section for Th-232 is equal to zero until 1.0 MeV, and only from 1.4 MeV the fission is a bit significant (0.01 barns). However, for U-238, the fission cross section will be significant at energies below 1 MeV so that U-238 will take part in total fission. Still, the participation of Th-232 is of minor importance, as shown in figure 4. The fission cross section for Th-232 and U-238 are shown in figure.
3. From figure 4, it is clear that fission in Th-232 in Th-based fuel is almost constant and does not exceed 1.5% of total fission in fuel. However, in LEU fuel fission in U-238 increases from 5% at BOC to 8.5% at the end. Also, clear that in Th-based fuel, only U-233 is responsible for fission. Nevertheless, in LEU, beside U-235 and after 5 days, Plutonium also takes part in fission.

![Figure 3. Fission cross sections for Th-232 and U-238.](image1)

![Figure 4. Isotopic fission percent in (a) Th-based fuel, (b) LEU.](image2)
4.3. Conversion ratio

The conversion ratio is the more important neutronic measurement of the fuel cycle economy. The conversion ratio is the ratio of fissile bred to fissile consumed. Figure 5 represents the conversion ratio for both fuels.

\[
\% \text{ of fission in isotope} = \frac{\text{number of fission in isotope}}{\text{Total number of fissions in fuel}}
\]  

(3)

From figure 5, the conversion ratio (CR) did not exceed 0.9 for both fuels; this means the reactor is a converter (i.e. \(1 > CR > 0\)) for both cases. CR for Th-based fuel is higher than that for LEU and at the End Of Cycle (EOC), Th-based fuel achieved 15\% higher than LEU. This can be explained on the basis of the capture rate in fertile and fissile for both fuels. As shown in figure 6, most of the capture in Th-based fuel occurred in fertile Th\(^{232}\) (67\% at BOC to 53\% at EOC), which means more U\(^{233}\) will produce, and less in fissile (about 10\%) which means less losing of fissile U\(^{233}\). However, in LEU fewer captures will take place in U\(^{238}\) (58\% at BOC to 36\% at EOC), which means less fissile material will produce from radiative capture in fertile material, and more radiative capture will take place in fuel which means more loss of fissile content than Th-based fuel.
Figure 6. Isotopic fission percent in (a) LEU, (b) Th-based fuel.

Isotopic reaction (fission or capture) Percentage = \[100 \times \frac{\sum_{isotope}^{reaction}}{\sum_{isotope}^{absorption}}\] (4)

4.4 Average number of neutrons produced per fission (\(\nu\)) and reproduction factor (\(\eta\)).

Not all neutrons that absorbed in fuel cause fission; some do not [16]. Reproduction factor \(\eta\) is the number of neutrons produced after one neutron absorbed by fission, and it differs from \(\nu\) the total number produced per fission [17, 26]. \(\eta\) plays a significant role in determining if the reactor is converter or breeder, so if \(\eta\) is bigger than one the reactor will be converter and if \(\eta\) more than two the reactor will be breeder [18, 19]. \(\eta\) in thermal neutrons region for U-233 and U-235 are (2.26 and 2.08 respectively)[20]. This meaning that one neutron needed to make fission and 1.26 neutrons can be
absorbed in Th-232 to convert it to fertile U-233, but only 1.08 neutrons available for U-238 to convert to Pu-239.

This gives to U-233 superior over U-235; this superiority will increase when fertile Th-232 (which have a larger capture cross section than U-238) works together with fissile U-233, which help to increase the conversion ratio. Values of $\eta$ for fissile only will change in case of fuel contains several fissionable materials, and will change with time because of the change of isotopic composition. Figure 5 shows the changing of $\eta$ and $\nu$ with time.

![Figure 7](image_url)

**Figure 7.** Change of reproduction factor ($\eta$) and the number of neutrons per fission ($\nu$) with time.

Figure 7 shows that $\eta$ for LEU will be higher than that for Th-based fuel, which can be explained by the contribution of Pu-239, Pu-241, and U-238 in total fission where in Th-based fuel only U-233 can make fission and the contribution of Th-232 can be neglected. $\nu$ for Th-based fuel will be almost constant all over the time of burning because the fissile produced and consumed in Th-based fuel are the same (U-233). While at the beginning of the first cycle, the $\nu$ for Th-based fuel was higher than that for LEU because at this time (until day 180), the fissile will be mainly U-233 in Th-based fuel and U-235 for LEU which has $\nu$ equal (2.48 and 2.43 respectively). After that, Pu-239 and Pu-241 (which have $\nu$ equal 2.87 and 2.97 respectively) will build up to significant quantities.

### 4.5 The effective delayed neutron fraction ($\bar{\beta}_{eff}$)

$\bar{\beta}_{eff}$ is the thermal neutrons fraction, which borne delayed [21]. It is a very important factor in nuclear reactor safety where it plays an important role in determining the reactor period ($\tau$) (the period required to the reactor to change its power by a factor of e, where $e = 2.718$).

$\bar{\beta}_{eff}$ depends on the type of fuel, with burnup, change in isotopic composition, dimensions and type of reactor. Figure 6 shows the change $\bar{\beta}_{eff}$ with time for both fuels. $\bar{\beta}_{eff}$ for LEU always higher than that for Th-based fuel because the fissile in LEU is U-235, which has $\beta = 0.0069$, but in Th-based fuel, the fissile is U-233, which has $\beta = 0.0031$. $\bar{\beta}_{eff}$ for LEU decreased with time because of the
involution of Pu-239 (which has relatively low $\beta$, $\beta = 0.0026$) in the fission process, which lowers the average delayed neutrons and effective delayed neutrons.

On the other hand, for Th-based fuel, $\bar{\beta}_{\text{eff}}$ slightly increases with time because of the participation of U-235 in fission at the end of the second cycle.

4.6. U-232 contamination and Minor Actinides (MA) inventory

One of the most important reasons behind using Thorium is lowering MA, which produces from the burning process. Lowering MA reduces the level of radiotoxicity in spent fuel. Nevertheless, using Thorium increases U-232 ($\alpha$-particles emitter, half-life = 73.6 years) production and Th-228 (possess $\alpha$-decay and produce very strong $\gamma$ - emitters Pb-212, Bi-212, and Ti-208) which cannot be chemically separated (until this moment).

The amount of U-232 in spent fuel is one of the most important factors that determine the validity of using Th-base fuel in reactors, and it depends on the fast neutron flux and the time of irradiation. Therefore, a large amount of U-232 will most likely be produced in fast reactors, which use Thorium based fuel. In harder flux, the amount of U-232 in spent fuel will be in the range 2000 – 3000 ppm, and in thermal reactors, the U-232 amount will be in the range 500 – 1000 ppm.

Because of putting the Th-blanket 15 – 20 cm away from the core border in Russian fast neutron reactor BN-350, the amount of U-232 in the blanket was significantly decreased to 2 – 11 ppm[22]. On the other hand, the production of U-232 and Th-228 can help the proliferation resistance effort, because they complicate the chemical reprocessing of spent fuel.

In this work, and at the assumed fissile enrichment, 4.95 % and burn up 58 MWd/kgHM. The amount of U-232 produced in Th-based fuel was 37 gm or 73 ppm in the fuel assembly. Which, in principle, acceptable, but a full core burnup is needed to more accurate prediction of U-232 amount. Nevertheless, in LEU fuel, the amount of U-232 produced can be neglected. Masses inventory of MA in fuel assembly after burnup for 1440 days are presented in table 3.

| Element vector | Th-based fuel | LEU          |
|----------------|---------------|--------------|
| Pu             | 1.56E+00      | 6.14E+03     |
| Np             | 5.88E+00      | 4.26E+02     |
| Am             | 9.31E-04      | 1.46E+02     |
| Cm             | 2.04E-04      | 6.61E+01     |

5. Safety-related neutronic parameters

In PWR, the most important neutronic safety parameter is the temperature coefficient of reactivity. In this study, fuel and moderator temperature coefficients for both fuels were studied, and the results are presented in Table 4.

To calculate the fuel temperature feedback, the fuel temperature increased by a step of 100 K, and to calculate the moderator temperature feedback, the water temperature increased by a step of 30 K, and its density decreased from 0.7001 to 0.6069 gm/cm$^3$.

| Fuel temperature reactivity coefficient, [pcm / K] | Th-based fuel | -1.745 ± 0.198 |
| Moderator temperature reactivity coefficient, [pcm / K] | LEU fuel       | -1.726 ± 0.199  |

| Fuel temperature reactivity coefficient, [pcm / K] | Th-based fuel | -22.08 ± 0.204 |
| Moderator temperature reactivity coefficient, [pcm / K] | LEU fuel       | -47.37 ± 0.20  |
As shown in the Table, temperature reactivity coefficients have negative feedback. However, the moderator temperature coefficient for LEU is higher and safer, all values within the range of PWRs temperature coefficients (-4 to -1 pcm / K, for fuel temperature reactivity coefficient) and (-50 to -8 pcm / K, for moderator temperature coefficient of reactivity)[23].

6. Conclusion
This work aimed to a better understanding of the effect of replacing the fuel used now (LEU) by (Th-232 + U-233)O2 fuel without any assembly modifications.

• The results showed that Th-based fuel could achieve higher burnup by about 10 % over LEU for the same enrichment.

• At the same level of enrichment, (Th-232 + U-233)O2 fuel has higher excess reactivity at BOC and conversion ratio than (U-238 + U-235)O2 fuel, which reflects the possibility of Th-based fuel to extend the core life cycle after that value for LEU. So more burn up and lowering the amount of fuel remaining in spent fuel. At the same time, more researches requested to find a good cladding material that can withstand deeper burnup.

• Th-based fuel significantly reduced the Minor Actinides (MA) production especially Pu, which can play a significant rule in proliferation resistance and lowering the radiotoxicity of spent fuel. In spite of this, Th-based fuel produces small amounts of U-232 and Th-228, which increases the activity of spent fuel and makes it difficult to processing. This can be considered as a double-edged weapon where it will increase the proliferation resistance.

• From a safety point of view, the results showed that LEU had a higher amount of $\beta_{eff}$ along the cycle’s time, and had a higher moderator temperature reactivity coefficient, but the values for Th-based fuel still comparable to the LEU.

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