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Original Article

NEUTRONICS INVESTIGATION OF CANADA DEUTERIUM URANIUM 6 REACTOR FUELED (TRANSURANIC–TH) O_2 USING A COMPUTATIONAL METHOD

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ABSTRACT

Background: ^{241}Am, ^{243}Am, and ^{237}Np isotopes are among the most radiotoxic components of spent nuclear fuel. Recently, researchers have planned different incineration scenarios for the highly radiotoxic elements of nuclear waste in critical reactors. Computational methods are widely used to predict burnup rates of such nuclear wastes that are used under fuel matrixes in critical reactors.

Methods: In this work, the *Monte Carlo N-particle transport code* was used to calculate the neutronic behavior of a transuranic (TRU)-bearing CANada Deuterium Uranium 6 reactor.

Results: The computational data showed that the 1.0% TRU-containing thorium-based fuel matrix presents higher proliferation resistance and TRU depletion rate than the other investigated fuel Matrixes. The fuel matrix includes higher negative temperature reactivity coefficients as well.

Conclusion: The investigated thorium-based fuel matrix can be successfully used to decrease the production of highly radiotoxic isotopes.

1. Introduction

In contrast to natural uranium that contains fissile isotope ^{235}U, thorium is naturally monoisotopic. Utilization of thorium in the initial stage requires the assistance of fissile materials such as ^{235}U or ^{239}Pu from the processing of fuel cycles. Because of the limited availability of uranium resources in contrast to the high availability of thorium resources, countries using nuclear energy are encouraged to pursue thorium-based fuel applications. In addition, thorium-based fuel-proliferation resistance and its breeder capability in thermal and fast nuclear reactors are other vital advantages

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of this fuel. In addition, thorium-based fuel has higher thermal conductivity, lower fission-gas-release characteristics, better dimensional stability at high burnups, stable stoichiometry, and lower thermal expansion coefficient than uranium-based fuel, which shows that its operating performance will be superior to that of uranium fuel [1,2]. Long half-life alpha-emitter isotopes of nuclear reactor spent fuel can properly be transmuted to short half-life isotopes under thorium-based fuel matrixes that are used in a fast or thermal nuclear reactor.

Hyland and Gihm [3] described many of the CANada Deuterium Uranium (CANDU) reactor design features, such as small and simple fuel bundles, that make it uniquely adaptable for actinide transmutation. They indicated that online refueling allows for precise management of core reactivity and separate insertion of the actinides and fuel bundles into the core. In addition, the high neutron economy of the CANDU reactor can result in high transuranic (TRU) destruction-to-fissile loading ratio. They used *WIMS-AECL* for neutronic calculations of the CANDU 6 core-fed minor actinide-containing fuel. They planned two transmutation schemes of several different partitioning schemes consisting of americium and group-extracted TRU elements, in which all of the TRU elements, plutonium (Pu), neptunium (Np), americium (Am), and curium (Cm), are extracted together and then transmuted. They investigated heterogeneous and homogeneous actinide loading in CANDU reactors. The data obtained by Hyland and Gihm [3] show that the limitation in the homogeneous method reduces the destroyed fraction of actinides during the irradiation in the core. The discharged actinide fuel may require secondary reprocessing and transmutation to achieve high destruction in comparison with the heterogeneous methods.

According to Sahin et al.'s report [4], however, the present generation of CANDU reactors are fueled with natural uranium (approximately 0.71% enrichment), but when using an enrichment grade > 1%, the use of thorium in CANDU reactors becomes viable [4]. They investigated the application of spent fuel of light-water reactors (LWRs) along with some ThO_2 fractions for flux flattening of the CANDU core using transport calculations by solving the Boltzmann transport equation with the transport code *XSDRNPM* using the 238-group neutron data library, which is derived from *ENDF/B–V*.

As the result obtained by Sahin et al. [4] shows, in standard CANDU reactors, the fission power profile, fuel burnup, and fuel exploitation within the bundle are nonuniform due to the strong depression of the very soft thermal neutron flux spectrum in the fuel zone. The nonuniformity can be overcome by increasing the macroscopic fission cross section in the internal fuel rods (FRs). An elegant method of power flattening has been achieved in the bundle by decreasing the LWR spent fuel fraction and increasing the ThO_2 fraction in the mixed fuel in the radial direction, by keeping the FR dimensions unchanged.

Sahin et al. [5] investigated burn up of uranium carbide fuels containing different TRU fractions in a CANDU thorium reactor. Their results indicate that the provided high burnups would drastically reduce the nuclear waste mass per unit energy output. Their study result has clearly shown that TRU in the form of tri-isotropic fuel pellets will provide sufficient criticality as well as reasonable burnup for CANDU reactors to justify their consideration as an alternative fuel [5].

Sahin et al. [6] evaluated the CANDU reactor as a thorium burner. In their study, two different fuel compositions were selected for investigation: (1) 96% thoria (ThO_2) + 4% PuO_2 and (2) 91% ThO_2 + 5% UO_2 + 4% PuO_2. As the computational data obtained by Sahin et al. [6] show, the reactor criticality k_{inf} remains nearly constant between the 4th year and the 7th year of plant operation, and then, a slight increase is observed thereafter, along with a continuous depletion of the thorium fuel; this behavior is more or less identical for both of the investigated fuels. After the 2nd year, the CANDU reactor begins to operate practically as a thorium burner. The reactor criticality would be sufficient until a great fraction of the thorium fuel is burned up, provided that the FRs could be fabricated to withstand such high burnup levels. Fuel fabrication costs and nuclear waste mass for final disposal per unit energy could be reduced drastically.

Sahin et al. [7] studied a CANDU reactor fueled with a mixed fuel made of ThO_2/MAO_2. The MA content fuel was a mixture of ^{237}Np, ^{238}Pu to ^{242}Pu, ^{241}Am, ^{242m}Am, ^{243}Am, ^{244}Cm, and ^{245}Cm. The results of their study indicate utilization of a mixed fuel in a CANDU reactor, consisting of ThO_2/MAO_2 components with variable fractional composition, which will allow for operating the reactor over unprecedentedly long periods without the need for fuel renewal [7].

Turkmen and Zabunoğlu [8] investigated the use of U and U–Th fuels in CANDU-type reactors [CANDU-6 and advanced CANDU reactor-700 (ACR-700)] on the once-through nuclear fuel cycle. Their results show that the U–Th fuel had a lower natural uranium requirement than the natural uranium fuel in CANDU-6. In addition, the U–Th fuel had a lower natural uranium requirement than the slightly enriched uranium fuel (2.1 without enriched fuel) in ACR-700. Concisely, once-through use of Th in a straightforward, homogeneous way in CANDU-6 or ACR-700 increases natural uranium requirement and decreases nuclear resource utilization. Then, Th is not a good replacement for ^{238}U in the pressurized heavy-water reactors on the once-through cycle [8].

2. Materials and methods

In this work, *Monte Carlo N-particle transport code* (*MCNPX*) 2.6.0 has been used as a powerful particle transport code with the ability to calculate steady-state reaction rates, normalization parameters, neutronic parameters, as well as fuel burnup using CINDER90 to calculate the time-dependent parameters [9,10].

A square-lattice 380-assembly CANDU 6 core was modeled using the MCNPX 2.6.0 code. Heavy water was selected as a coolant for the assemblies. Heavy water was also selected as a moderator of the modeled core. A three-dimensional neutronic model was set up using the MCNPX 2.6.0 code in cold zero power situations by *ENDF/B-VI* continuous-energy cross section. The cross sections of $S(\alpha, \beta)$ were used for heavy water. The KCODE capability was used for neutronic parameter calculations. A total of 37 concentric fuel pins were used

Table 1 – Core material and dimensions modeled using *MCNPX 2.6.0*.

Unit	Value	Core specifications
g/cm^3	10.60	Fuel Matrix 1 (W%): ^{233}U, 2.0; ^{232}Th, 85.09; ^{243}Am, 0.2547; ^{241}Am, 0.07; ^{237}Np, 0.175; O, remaining balance
g/cm^3	10.60	Fuel Matrix 2 (W%): ^{233}U, 2.3; ^{232}Th, 84.50; ^{243}Am, 0.51; ^{241}Am, 0.14; ^{237}Np, 0.35; O, remaining balance
g/cm^3	6.55	Fuel clad: Zircaloy-4
g/cm^3	6.53	Pressure tube clad: Zircaloy-2
g/cm^3	6.56	Calandria clad: Zr–Nb
g/cm^3	0.001977	Gap: CO_2
Cm	1.2243	Fuel rod diameter
No.	37	Fuel rod number in assembly
No.	380	Fuel assembly number
cm	758 × 600	Core dimension
t	2.11	Fissile mass in Fuel Matrix 1
t	2.42	Fissile mass in Fuel Matrix 2
t	0.073	Total mass of ^{241}Am in Fuel Matrix 1
t	0.268	Total mass of ^{243}Am in Fuel Matrix 1
t	0.184	Total mass of ^{237}Np in Fuel Matrix 1
t	0.147	Total mass of ^{241}Am in Fuel Matrix 2
t	0.535	Total mass of ^{243}Am in Fuel Matrix 2
t	0.368	Total mass of ^{237}Np in Fuel Matrix 2

MCNPX, Monte Carlo N-particle transport code.

in any assembly with CANDU 6 [7] assembly characteristics according to the core specifications presented in Table 1.

Two TRU weight fractions of 0.5% and 1.0% were introduced in the thorium-based fuel matrix separately. Neutronic parameters of the modeled CANDU core were calculated using different TRU-containing fuel matrixes. The cross-sectional view of the modeled CANDU core in MCNPX is depicted in Fig. 1.

To investigate the impact of a TRU-containing thorium-based fuel matrix on neutronic performances of the modeled

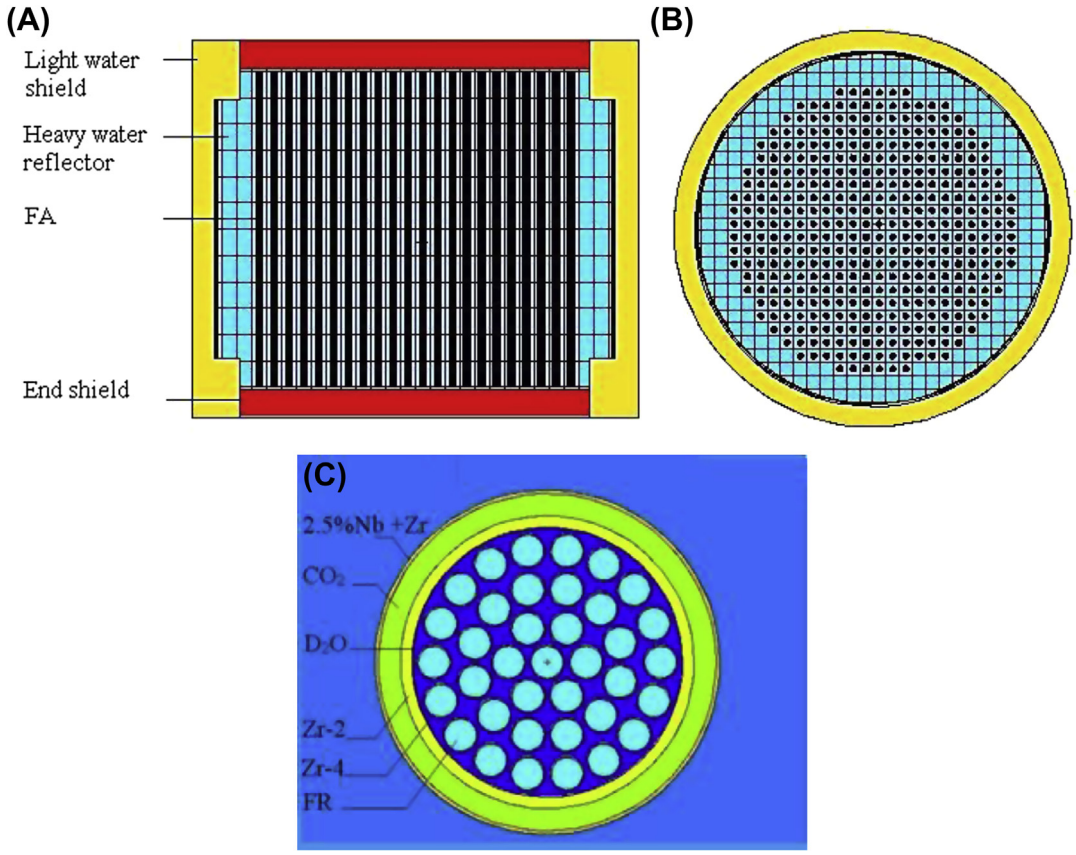

Fig. 1 – Cross-sectional view of the modeled CANDU 6 core. (A) Axial; (B) radial; and (C) fuel assembly containing 37 FRs. CANDU 6, CANada Deuterium Uranium 6; FA, fuel assembly; FR, fuel rod.

core, the following neutronic parameters were calculated: radial and axial neutron flux distributions were calculated using the mesh tally card of the computational code. Deposited power distributions were calculated using the mesh tally card for the hottest fuel assembly. Neutron spectra and average fission per absorption ratio were calculated using F4 tally. Reactivity coefficients of fuel, coolant, and moderator were calculated using the TMP card module and temperature-related cross-section libraries of .70c, and .71c from ENDF70 in MCNPX. Void reactivity variations of the coolant and moderator were calculated for the different fuel loads in the modeled CANDU core. Delayed neutron fraction and effective delayed neutron fraction were calculated separately for the CANDU 6 cores fueled with different TRU-containing thorium matrixes. Burnup calculation was performed at a power of 1,500 MW for 3 years using the BURN card module for the different fuel matrixes separately. Depletion rates of $^{241/243}$Am and ^{237}Np after the burnup time were investigated. Production of long half-life alpha-emitter radioisotopes after the burnup time was investigated.

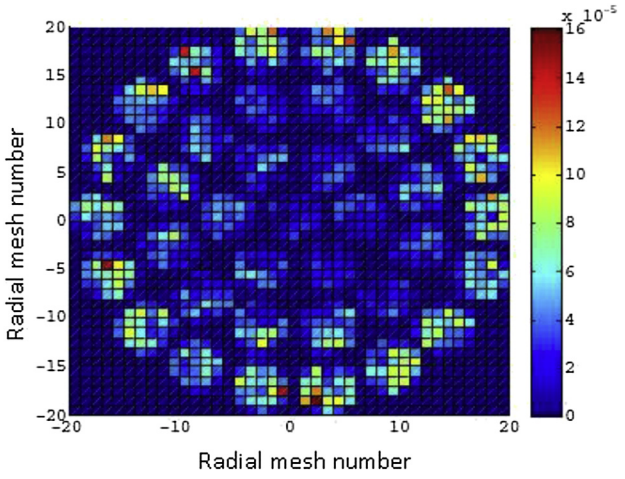

Fig. 3 – Power distribution in the hottest fuel assembly.

3. Results and discussion

Neutron flux distribution calculations showed that the central fuel assemblies experience more neutron flux than the external fuel assemblies (Fig. 2). Therefore, the hottest fuel assembly is obviously the central one.

Power distribution in the hottest fuel assembly was calculated using the mesh tally card of the computational code. The calculations showed that the external ring containing FRs experienced more power deposition in comparison with the inner rings (Fig. 3). Therefore, power density was calculated for the hottest FR.

The radial power peaking factor of the hottest fuel assembly was 2.15 and the axial power peaking factor of the hottest fuel assembly was 1.12 for the 0.5% TRU-containing fuel matrix loads. The radial and axial power peaking factor values were 2.11 and 1.10 for the 1.0% TRU-containing fuel matrix loads. Neutron spectra were calculated separately for the CANDU 6 cores, which fueled the different TRU-containing fuel matrixes. The calculations showed that the neutron spectra of both TRU-containing fuel matrixes investigated are approximately overlapped (Fig. 4).

The results presented in Table 2 show that the higher TRU-containing fuel loads in CANDU 6 decrease neutron-generation time compared with the lower TRU-containing fuel loads with 6.1% relative discrepancy. However, enhancement of TRU concentration in the thorium-based fuel matrix favorably increased the delayed and effective delayed neutron fractions of the CANDU 6-modeled core. In addition, the higher TRU-containing fuel experiences a higher fission per absorption ratio in the modeled thermal CANDU 6 core than the other investigated fuel matrix with 3.2% relative discrepancy. The calculations carried out showed that the higher TRU-containing fuel experiences a fairly higher temperature fuel reactivity coefficient than the other investigated fuel matrix with 10.4% relative discrepancy. The higher TRU-containing fuel resulted in noticeably higher temperature moderator and coolant reactivity coefficients than the lower TRU-containing thorium-based fuel matrix. Therefore, 1.0% weight fraction of TRUs in the thorium-based fuel matrix enhanced safety

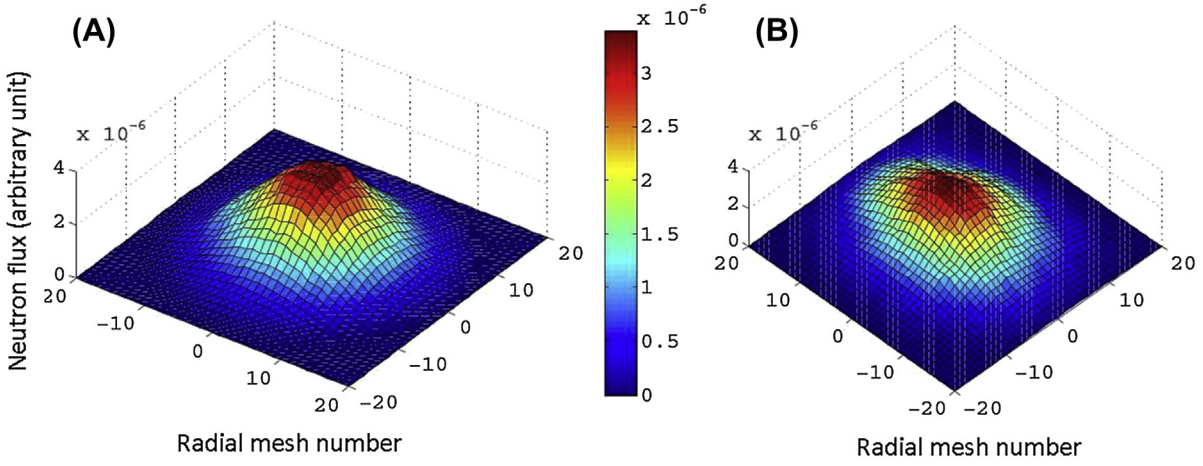

Fig. 2 – Neutron flux distribution. (A) Radial. (B) Axial.

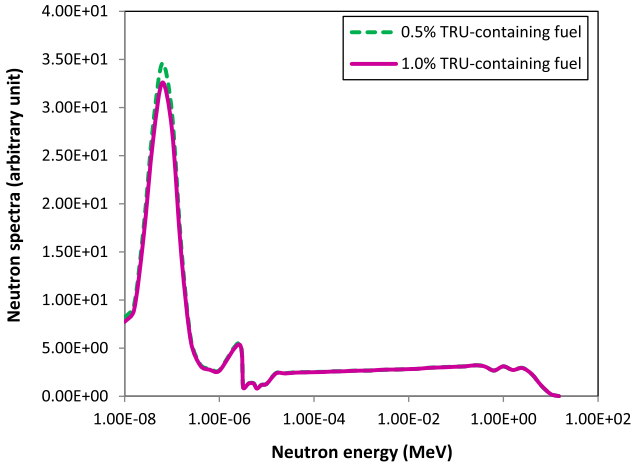

Fig. 4 – Total neutron spectra in the modeled core with different fuel matrixes. TRU, transuranic.

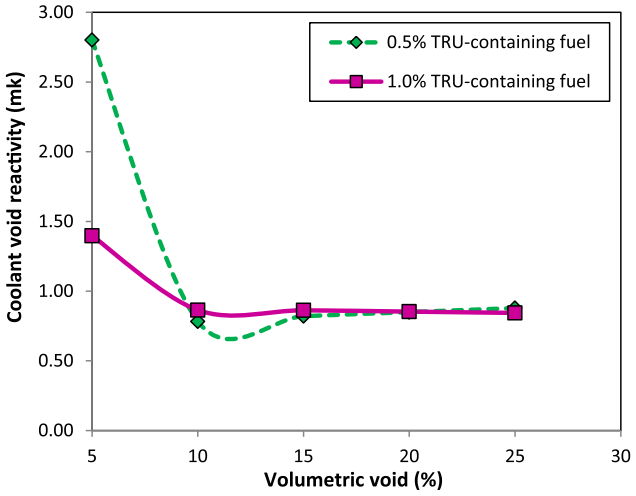

Fig. 5 – Coolant void reactivity variations in the modeled core with different fuel matrixes. TRU, transuranic.

operation of the CANDU 6-modeled core due to the more negative temperature reactivity coefficients.

The computational data show that the hottest FR containing higher TRU concentrations receives higher power density (1,177 W/cm^3) than the other investigated fuel matrix (1,135 W/cm^3) with 3.7% relative discrepancy. The radial power peaking factor of the modeled CANDU 6 reactor fueled with a 0.5% TRU thorium-based matrix was 2.49, which is higher than the value obtained by 1.0% weight fractions of TRU in the CANDU 6 thorium-based matrix fuel (2.39) with 4.1% relative discrepancy. The axial power peaking factor of the CANDU 6 core-fed higher TRU-containing fuel was lower than the other investigated fuel matrix loading with about 1.0% relative discrepancy. Therefore, enhancement of TRU concentration in the thorium-based fuel matrix reduced power peaking factors of the modeled core.

Void reactivity coefficients related to void formation in the coolant and moderator were calculated separately for the different TRU-containing fuel matrix loads. As Fig. 5 shows, the higher TRU-containing fuel experiences less coolant void reactivity coefficient variation during volumetric void fraction alteration of 5–10% in comparison with the 0.5% TRU-containing fuel loads of the modeled CANDU 6 core. During enhancement of void volumetric fractions from 10% to higher values, the coolant void reactivity coefficients are approximately constant for both TRU-containing fuel matrix loads (~0.85 mk). The calculations carried out show that the 0.5% TRU-containing fuel matrix load in the CANDU 6 core resulted in noticeably positive enhancement of the moderator void reactivity coefficient during void volumetric fraction growth (from −4.55 to −2.45 mk), whereas the 1.0% TRU-containing fuel matrix load in the CANDU 6 core resulted in reductions in the reactivity coefficient during void volumetric fraction growth (from −2.7 to −9.87 mk; Fig. 6). The calculation errors were < 80 pcm.

According to the calculations, an integrated neutron flux on the order of 10^{14} (n/s·cm^2) is available in the modeled core.

Table 2 – Comparison of different neutronic parameters of the modeled CANDU core-fed different fuel matrixes.

Neutronic parameters	0.5% TRU-containing fuel	1.0% TRU-containing fuel	Statistical errors (%)
Neutron-generation time (μs)	538	507	< 0.2
Delayed neutron fraction (pcm)	408	652	< 50 pcm
Effective delayed neutron fraction (pcm)	552	616	< 50 pcm
Fission per absorption ratio	0.92	0.95	< 0.2
Fuel temperature reactivity (mk/K)	−0.0239	−0.0268	< 50 pcm
Coolant temperature reactivity (mk/K)	−0.0085	−0.0148	< 50 pcm
Moderator temperature reactivity (mk/K)	−0.0409	−0.0543	< 50 pcm
Deposited power in the hottest rod (kW)	66.8	69.3	< 1.0
The hottest fuel rod power density (W/cm^3)	1,135	1,177	< 2.0
Radial power peaking factor	2.49	2.39	< 0.5
Axial power peaking factor	1.98	1.96	< 0.5
Fraction of neutrons with E_n < 1 eV in total core (%)	66.50	65.20	< 0.2
Fraction of neutrons with 1 eV < E_n < 1 keV in total core (%)	12.10	12.40	< 0.2
Fraction of neutrons with E_n > 1 keV in total core (%)	21.40	22.30	< 0.2

The variation was calculated during transit from 293 K to 599 K.
CANDU, CANada Deuterium Uranium; TRU, transuranic.

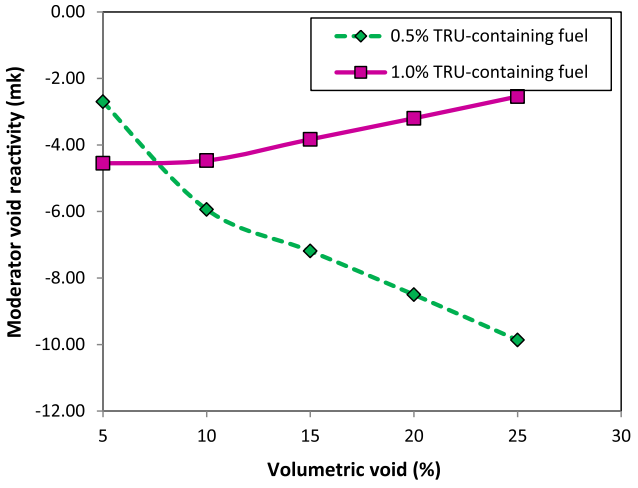

Fig. 6 – Moderator void reactivity variations in the modeled core with different fuel matrixes. TRU, transuranic.

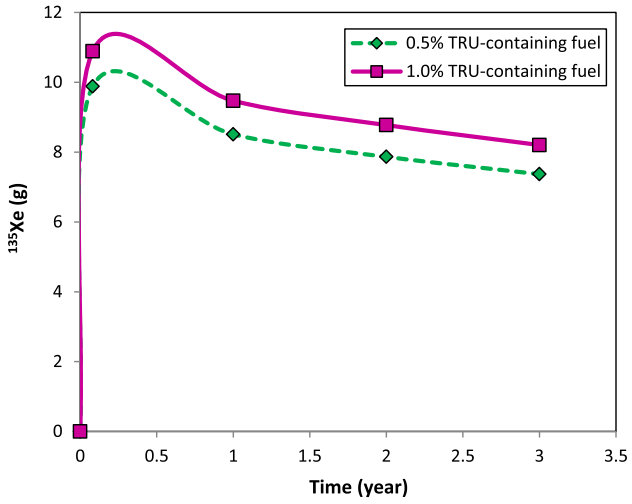

Fig. 8 – Dependence of ^{135}Xe buildup on burnup time at 1,500-MW power. TRU, transuranic.

As Fig. 7 shows, the core effective multiplication factor decreased to approximately 1.00085 pcm after 1,642.5 GWd in the case of 1.0% TRU-containing fuel; there is roughly parallel behavior of multiplication changes for both TRU-containing fuel matrixes during the burnup process.

The ^{135}Xe buildup after 3 years' burn up of the higher TRU-containing fuel matrix is approximately 1.1 times higher compared with the other fuel matrix investigated. The ^{149}Sm buildup after the burnup time is approximately 1.2 times higher in the case of the 1.0% TRU-containing fuel core compared with the other fuel matrix investigated (Figs. 8 and 9).

The estimated concentration buildup of the poisons by the used computational code contained calculation errors < 0.1%.

3.1. *Inventory of thorium isotopes*

The presented burnup calculations in Table 3 show that ^{231}Th and ^{233}Th isotopes were produced during burning of the thorium fuel matrixes.

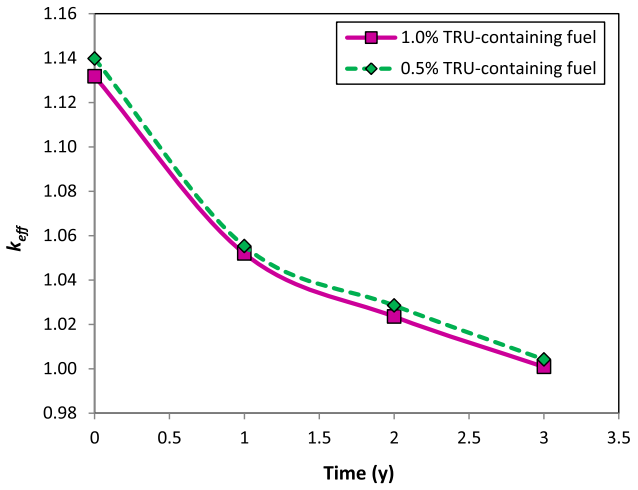

Fig. 7 – Effective multiplication factor variations during burnup time at a power of 1,500 MW. TRU, transuranic.

The ^{231}Th beta-emitter isotope was produced in both fuel matrixes investigated. The radioisotope decays to ^{231}Pa (long half-life alpha-emitter: 32,760 years) after 25.52 hours. Its buildup concentration is fairly high (5.73 and 5.53 g) in the fuel matrixes, whereas the 0.5% TRU-containing fuel matrix bears higher buildups of ^{231}Th.

The ^{233}Th inventory occurred in the both burnt fuel matrixes. The produced beta-emitter concentration is 30.3 g in the 0.5% TRU-containing fuel and 27.3 g in the 1.0% TRU-containing fuel. It converts to ^{233}U through the $^{233}\text{Th} \xrightarrow{22.5\ m} {}^{233}\text{Pa} \xrightarrow{26.967\ d} {}^{233}\text{U}$ decay chain.

3.2. *Inventory of protactinium isotopes*

Three isotopes of ^{232}Pa and ^{233}Pa were produced in the burnt fuel matrixes. The ^{232}Pa beta-emitter isotope was produced in

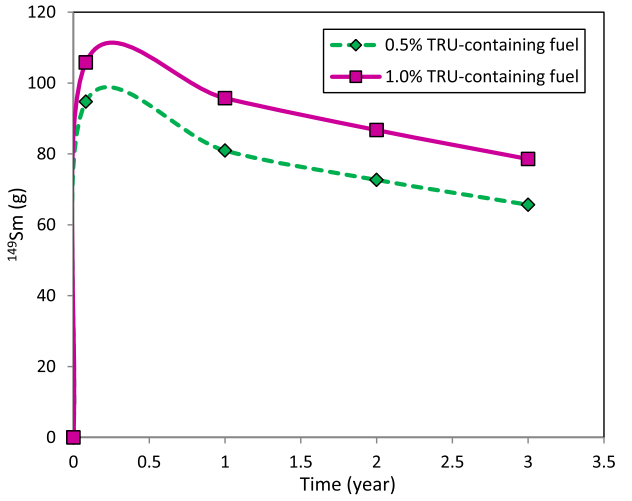

Fig. 9 – Dependence of ^{149}Sm buildup on burnup time at 1,500-MW power. TRU, transuranic.

Table 3 – Comparison of actinide mass inventory of the modeled CANDU core-fed different fuel matrixes (power = 1,500 MW, burnup time = 3 y).

Radioisotope	0.5% TRU-containing fuel		Half-life	Emitted particle	1.0% TRU-containing fuel	
	Mass (g)	Activity (Ci)			Mass (g)	Activity (Ci)
^{231}Th	5.73×10^{00}	3.05×10^{06}	7.538×10^{4} y	β^-	5.53×10^{00}	2.94×10^{06}
^{232}Th	8.84×10^{07}	9.69×10^{00}	25.52 h	α	8.77×10^{07}	9.61×10^{00}
^{233}Th	3.03×10^{01}	1.10×10^{09}	1.405×10^{10} y	β^-	2.73×10^{01}	9.86×10^{08}
^{232}Pa	3.71×10^{00}	1.59×10^{06}	22.3 m	β^-	3.25×10^{00}	1.40×10^{06}
^{233}Pa	5.22×10^{04}	1.08×10^{09}	1.31 d	β^-	4.70×10^{04}	9.75×10^{08}
^{233}U	8.10×10^{05}	7.80×10^{03}	26.967 d	α	7.62×10^{05}	7.34×10^{03}
^{234}U	5.93×10^{04}	3.69×10^{02}	68.9 y	α	5.03×10^{04}	3.12×10^{02}
^{235}U	6.87×10^{05}	1.49×10^{00}	1.592×10^{5} y	α	9.03×10^{05}	1.95×10^{00}
^{236}U	2.23×10^{05}	1.44×10^{01}	2.455×10^{5} y	α	2.39×10^{05}	1.55×10^{01}
^{237}U	1.08×10^{02}	8.87×10^{06}	7.038×10^{8} y	β-	1.09×10^{02}	8.91×10^{06}
^{238}U	5.23×10^{01}	1.76×10^{-05}	2.342×10^{7} y	α	4.58×10^{01}	1.54×10^{-05}
^{236}Np	1.94×10^{-01}	2.55×10^{-03}	6.75 d	β^-	4.41×10^{-01}	5.81×10^{-03}
^{237}Np	1.26×10^{05}	8.90×10^{01}	2.144×10^{6} y	α	2.59×10^{05}	1.83×10^{02}
^{238}Np	1.58×10^{02}	4.10×10^{07}	2.117 d	β^-	2.87×10^{02}	7.44×10^{07}
^{239}Np	5.49×10^{-01}	1.27×10^{05}	2.3565 d	β^-	9.45×10^{-01}	2.19×10^{05}
^{237}Pu	3.57×10^{-02}	4.35×10^{02}	45.2 d	α, β^-	7.12×10^{-02}	8.68×10^{02}
^{238}Pu	6.64×10^{04}	1.14×10^{06}	87.7 y	α	1.27×10^{05}	2.18×10^{06}
^{239}Pu	1.66×10^{04}	1.03×10^{03}	24,110 y	α	2.93×10^{04}	1.81×10^{03}
^{240}Pu	7.18×10^{03}	1.63×10^{03}	6563 y	α	1.26×10^{04}	2.86×10^{03}
^{241}Pu	2.65×10^{03}	2.74×10^{05}	14.35 y	α	4.03×10^{03}	4.17×10^{05}
^{242}Pu	8.51×10^{03}	3.36×10^{01}	3.733×10^{5} y	α	1.60×10^{04}	6.34×10^{01}
^{243}Pu	7.14×10^{-01}	1.86×10^{06}	4.956 h	β^-	1.24×10^{00}	3.22×10^{06}
^{244}Pu	1.28×10^{01}	2.35×10^{-04}	8.08×10^{7} y	α	2.47×10^{01}	4.52×10^{-04}
^{241}Am	1.42×10^{04}	4.88×10^{04}	432.2 y	α	3.51×10^{04}	1.20×10^{05}
^{242}Am	7.00×10^{01}	7.34×10^{02}	16.02 h	EC, β^-	1.74×10^{02}	1.83×10^{03}
^{243}Am	1.54×10^{05}	3.08×10^{04}	7370 y	α	3.34×10^{05}	6.67×10^{04}
^{244}Am	3.49×10^{01}	4.44×10^{07}	10.1 h	EC, β^-	6.24×10^{01}	7.94×10^{07}
^{242}Cm	6.00×10^{03}	1.99×10^{07}	162.8 d	α	1.23×10^{04}	4.07×10^{07}
^{243}Cm	2.20×10^{02}	1.13×10^{04}	29.1 y	EC, α	4.12×10^{02}	2.13×10^{04}
^{244}Cm	1.01×10^{05}	8.20×10^{06}	18.10 y	α	1.81×10^{05}	1.47×10^{07}
^{245}Cm	2.79×10^{03}	4.78×10^{02}	8500 y	α	4.60×10^{03}	7.89×10^{02}
^{246}Cm	7.00×10^{02}	2.15×10^{02}	4730 y	α	9.79×10^{02}	3.01×10^{02}

CANDU, CANada Deuterium Uranium; EC, electron capture; TRU, transuranic.

low concentrations (3.71 g and 3.25 g, respectively) after the fuel burning. As the computational data show, its concentration is higher in the case of the spent fuel containing 0.5% TRU. It converts to ^{232}U alpha-emitter long half-life (68.9 years) through the $^{232}\text{Pa} \xrightarrow{1.31\ \text{d}} {}^{232}\text{U}$ decay chain. Its decay chain terminates to ^{208}Pb after some years in which ^{232}U is the highest radiotoxic element of the decay chain.

The produced beta-emitter ^{233}Pa isotope in the spent fuel matrixes will decay to ^{233}U as mentioned previously. Its production concentration is higher in the case of the spent fuel containing 0.5% TRU in comparison with the spent fuel containing 1.0% TRU (52.2 kg and 47.0 kg, respectively).

3.3. Inventory of uranium isotopes

The produced uranium isotopes in the burnt fuel matrixes are ^{233}U, ^{234}U, ^{236}U, and ^{237}U. With the exception of ^{237}U, all are highly radiotoxic alpha-emitter elements.

The ^{234}U buildup (half-life, 2.455×10^5 years) is higher in the spent fuel containing 0.5% TRU than the other investigated fuel (59.3 kg and 50.3 kg, respectively). Its decay chain terminates at ^{206}Pb while the highest radiotoxic element of the chain is ^{234}U.

The ^{236}U buildup (half-life, 2.342×10^7 years) is considerably less in the lower TRU-containing fuel matrix than the other fuel matrix (687 and 903 kg, respectively). Its decay chain terminates at ^{208}Pb while the highest radiotoxic element of the chain is ^{232}Th (half-life, 1.405×10^{10} years).

The ^{237}U buildup in spent fuel matrix 1 (0.5% RTU containing) was approximately identical with the burnt fuel matrix 2 (1.0% RTU containing); approximately 109 g of ^{237}U beta-emitter isotope was produced in spent fuel matrixes after 1,642.5-GWd burn up. It decays to ^{237}Np through the $^{237}\text{U} \xrightarrow{6.75\ \text{d}} {}^{237}\text{Np}$ decay chain. Its decay chain terminates at stable ^{209}Bi while the highest radiotoxic element of the chain is ^{237}Np. The ^{238}U buildup in the spent fuel was 52.3 g and 45.8 g in the 0.5% TRU-containing and 1.0% TRU-containing fuel matrixes, respectively.

3.4. Inventory of neptunium isotopes

Four neptunium isotopes were produced in the spent fuel matrixes after 3 years. Among them, ^{237}Np has the highest production concentrations during the burnup process. The produced radioisotope concentration in the 1.0% RTU-containing spent fuel matrix is approximately 2.05 times more

than that of the other investigated fuel matrix—126 kg and 259 kg, respectively (Table 3). It converts to ^{233}U according to the following decay chain: $^{237}\mathrm{Np} \xrightarrow{2.144\times10^{6}\ y} {}^{233}\mathrm{Pa} \xrightarrow{26.967\ d} {}^{233}\mathrm{U}$.

The produced concentrations of ^{236}Np and ^{239}Np beta emitters in the fuel matrixes are less than 1 g. The produced neptunium isotopes decay to ^{236}Pu—^{236}U and ^{239}Pu, respectively.

The concentration of ^{238}Np beta-emitter isotope in spent fuel 1 was 158 g, which is higher than the value in spent fuel matrix 2 (287 g). The isotope decays to ^{238}Pu after 2.117 days.

3.5. Inventory of plutonium isotopes

Eight isotopes of plutonium were produced in the fuel matrixes during the burnup process (from ^{237}Pu to ^{244}Pu). Among them, ^{237}Pu concentrations were insignificant in both the investigated spent fuels (< 0.08 g). The amount of ^{238}Pu produced in the 1.0% RTU-containing spent fuel matrix was approximately 1.92 times more than that of the other fuel matrix (66.4 kg and 128 kg for the 0.5% and 1.0% spent fuel matrixes, respectively). It decays to ^{234}U and ^{230}Th high radiotoxic elements through the $^{238}\mathrm{Pu} \xrightarrow{87.7\ y} {}^{234}\mathrm{U} \xrightarrow{2.455E5\ y} {}^{230}\mathrm{Th} \xrightarrow{7.538E4\ y} {}^{226}\mathrm{Ra}$ decay chain. The ^{239}Pu alpha-emitter concentration was prominently higher for burnt fuel matrix 2 than burnt fuel matrix 1 (16.6 kg and 29.3 kg for the 0.5% RTU-containing and 1.0% RTU-containing fuel matrixes, respectively).

Another high radiotoxic isotope of plutonium produced is ^{240}Pu (half-life, 6,563 years). The produced concentrations were 7.18 kg and 12.6 kg for the 0.5% RTU-containing and 1.0% RTU-containing fuel matrixes, respectively. It decays to ^{236}U and ^{232}Th high radiotoxic elements through the $^{240}\mathrm{Pu} \xrightarrow{6,563\ y} {}^{234}\mathrm{U} \xrightarrow{2.342E7\ y} {}^{232}\mathrm{Th} \xrightarrow{1.405E10\ y} {}^{228}\mathrm{Ra}$ decay chain.

The amounts of ^{241}Pu produced were 2.65 kg and 4.03 kg for the 0.5% RTU-containing and 1.0% RTU-containing fuel matrixes, respectively. It decays to ^{241}Am and ^{237}Np high radiotoxic elements through the $^{241}\mathrm{Pu} \xrightarrow{14.35\ y} {}^{241}\mathrm{Am} \xrightarrow{432.2\ y} {}^{237}\mathrm{Np} \xrightarrow{2.144\times10^{6}\ y} {}^{233}\mathrm{Pa}$ decay chain.

The amounts of ^{242}Pu produced were 8.51 kg and 16.0 kg for the 0.5% RTU-containing and 1.0% RTU-containing fuel matrixes, respectively. Its decay chain is $^{242}\mathrm{Pu} \xrightarrow{3.733E5\ y} {}^{238}\mathrm{U} \xrightarrow{4.468E9\ y} {}^{234}\mathrm{Th} \xrightarrow{24.10\ d} {}^{234m}\mathrm{Pa} \xrightarrow{1.17\ m} {}^{234}\mathrm{U}$.

The amounts of ^{243}Pu produced were insignificant (< 1.5 g) for both the spent fuel matrixes. The ^{244}Pu-produced concentrations were 12.8 g and 24.7 g for the 0.5% RTU-containing and 1.0% RTU-containing fuel matrixes, respectively, after 3 years' fuel burning. It decays to ^{240}U after 8.08×10^{7} years.

3.6. Inventory of americium isotopes

The ^{242}Am concentration was 70 g in the 0.5% RTU-containing spent fuel. The isotope concentration was 174 g in the other investigated spent fuel. It decays to ^{242}Pu and ^{242}Cm through the electron capture and β^{-} decay, respectively.

The ^{244}Am concentration was 34.9 g in the 0.5% RTU-containing spent fuel. The isotope concentration was 62.4 g in the 1.0% RTU-containing spent fuel. Its decay chain is $^{244}\mathrm{Am} \xrightarrow{10.1\ h} {}^{244}\mathrm{Cm} \xrightarrow{18.10\ y} {}^{240}\mathrm{Pu}$.

3.7. Inventory of curium isotopes

The amounts of ^{242}Cm produced were 6.0 kg and 12.3 kg in the 0.5% RTU-containing and the 1.0% RTU-containing fuel matrixes, respectively. The alpha-emitter isotope follows chain decay of $^{242}\mathrm{Cm} \xrightarrow{162.8\ d} {}^{238}\mathrm{Pu} \xrightarrow{87.7\ y} {}^{234}\mathrm{U}$.

The amounts of ^{243}Cm produced were 220 g and 412 g in the 0.5% RTU-containing and in the 1.0% RTU-containing fuel matrixes, respectively. The alpha-emitter isotope follows chain decay of $^{243}\mathrm{Cm} \xrightarrow{29.1\ y} {}^{239}\mathrm{Pu} \xrightarrow{24,110\ y} {}^{235}\mathrm{U}$.

The amounts of ^{244}Cm produced were 101 kg and 181 kg in the 0.5% RTU-containing and in the 1.0% RTU-containing fuel matrixes, respectively. The alpha-emitter isotope follows chain decay of $^{244}\mathrm{Cm} \xrightarrow{18.10\ y} {}^{240}\mathrm{Pu} \xrightarrow{6,563\ y} {}^{236}\mathrm{U}$.

The amounts of ^{245}Cm produced were 2.79 kg and 4.60 kg for the 0.5% RTU-containing and the 1.0% RTU-containing burnt fuel matrixes, respectively. The alpha-emitter isotope follows chain decay of $^{244}\mathrm{Cm} \xrightarrow{8,500\ y} {}^{241}\mathrm{Pu} \xrightarrow{14.35\ y} {}^{241}\mathrm{Am}$.

According to Table 3, concentrations of ^{246}Cm were 700 g and 979 g after the burnup process in the investigated fuel matrixes. It decays to ^{242}Pu after 4,730 years.

Overall, 495 kg of long half-life alpha-emitter isotope was produced in the 0.5% RTU-containing spent fuel matrix after 1,642.5-GWd burn up. The value was 678 kg in the case of the 1.0% RTU-containing spent fuel matrix. Among the investigated produced high radiotoxic isotopes, the total masses of the isotopes with a half-life greater than 100 years were 318 kg and 353 kg at end of cycle (EOC) for the 0.5% RTU-containing and the 1.0% RTU-containing burnt fuel matrixes, respectively. The data presented in Table 4 indicate that 232 kg of the initially loaded TRUs depleted off the 0.5% RTU-containing burnt fuel matrix after 1,642.5-GWd burn up. This value was 424 kg in the case of the 1.0% RTU-containing burnt fuel matrix. Therefore, regardless of alpha emitters with half-life < 100 years, the 1.0% RTU-containing fuel matrix is a net long half-life high radiotoxic TRU burner (353 kg inventory < 424 kg depletion). Therefore, it can be said that loading of 1.0% TRU in the thorium-based fuel matrix resulted in reduction of radiotoxic effects of the spent fuel matrix in the CANDU 6 reactor at EOC.

According to Table 4, 826 kg of fissile isotopes consisting of ^{239}Pu and ^{233}U were produced in the 0.5% RTU-containing fuel at 1,500-MW burn up after 3 years, while 1.42 t of ^{235}U was consumed during this time. In the case of the 1.0% RTU-containing fuel, the produced and consumed values were 791 kg and 1.52 t, respectively.

The burnup data error of the actinides was < 2.0% on average, and the largest errors belonged to ^{236}Pu (4.49%) and ^{241}Am (2.79%).

According to the obtained computational data with respect to initial loads, 67.46%, 1.57%, 31.61%, 80.72%, and 42.55% of ^{235}U, ^{232}Th, ^{237}Np, ^{241}Am, and ^{243}Am, respectively, were consumed in the 0.5% RTU-containing burnt fuel matrix. In the case of the 1.0% RTU-containing burnt fuel matrix, the depleted values were 62.67%, 1.41%, 29.74%, 76.20%, and 37.67% for ^{235}U, ^{232}Th, ^{237}Np, ^{241}Am, and ^{243}Am, respectively.

As the data presented in Table 5 show, TRU loading in the CANDU 6 core enhanced the safety parameters of the core in

Table 4 – Comparison of consumption, depletion, and inventory of the most important isotopes of the modeled CANDU 6 core-fed different fuel matrixes (power = 1,500 MW, burnup time = 3 y).

Fuel type	Consumption (g)		Depletion (g)			Inventory (g)	
	^{235}U	^{232}Th	^{241}Am	^{243}Am	^{237}Np	^{239}Pu	^{233}U
0.5% TRU-containing fuel	1.42×10^{06}	1.41×10^{06}	5.96×10^{04}	1.14×10^{05}	5.84×10^{04}	1.66×10^{04}	8.09×10^{05}
1.0% TRU-containing fuel	1.52×10^{06}	1.26×10^{06}	1.12×10^{05}	2.02×10^{05}	1.10×10^{05}	2.92×10^{04}	7.62×10^{05}

CANDU, CANada Deuterium Uranium; TRU, transuranic.

Table 5 – Comparison of some safety parameters of the CANDU core-fed different fuels [11].

Safety parameter	0.5% TRU CANDU 6	1.0% TRU CANDU 6	$^{Nat}UO_2$ fuel CANDU 6	ACR-700
Fuel temperature effect (mk/K)	0.0239	0.0268	Small negative	0.0044
Coolant temperature effect (mk/K)	0.0085	0.0148	Positive	0.0033
Moderator temperature effect (mk/K)	0.0409	0.0543	Slightly positive	0.0072
Void effect (mk)	0.175	0.203	+10 to 15	3.0
Delayed neutron fraction (pcm)	408	652	580	560
Prompt neutron lifetime (ms)	0.61	0.62	0.92	0.33

ACR-700, advanced CANDU reactor-700; CANDU, CANada Deuterium Uranium; TRU, transuranic.
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comparison with the CANDU 6 core fueled with natural uranium oxide. In comparison with the modified version of the CANDU core (ACR-700), temperature reactivity coefficients of the CANDU 6 core bearing the different TRU weight fractions are more negative. In addition, the delayed neutron fraction of the 1.0% TRU-containing fuel matrix loads in the CANDU 6 core is higher than the $^{Nat}UO_2$-fueled CANDU 6 and AVR-700, which ensures higher safety of the core.

3.8. Conclusion

The investigated thorium-based fuel matrix can be successfully used to decrease the production of highly radiotoxic isotopes. According to the computational data, TRU loading in thorium-based fuel matrixes keeps temperature reactivity coefficients as negative, which ensures safe operation of the system. A 1.0% TRU-containing fuel matrix offers higher transmutation potentials than a 0.5% TRU-containing fuel matrix due to the fact that 424 kg of the TRU initial load is depleted during 1,642.5-GWd burn up, while approximately 353 kg of long half-life (> 100 years) alpha-emitter isotopes were produced at EOC, which is noticeably less. However, its use resulted in high plutonium isotopes' inventories with total concentrations approximately 123 kg higher than the 0.5% TRU-containing fuel matrix. In addition, the fuel matrix suffers higher buildup of ^{135}Xe and ^{149}Sm neutron poison isotopes.

Conflicts of interest

All contributing authors declare no conflicts of interest.

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