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THORIUM-BASED FUELS PRELIMINARY LATTICE CELL STUDIES FOR CANDU REACTORS

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Applying once-through Thorium (OTT) cycle in existing and advanced CANDU reactors might be seen as an evolutive concept for sustainable development both from economic and waste management points of view. Using the Canadian proposed scheme - loading mixed ThO_2-SEU CANFLEX bundles in CANDU 6 reactors - simulated at lattice cell level led to promising conclusions on higher burnup, minor actinide content reduction in spent fuel, reduction of spent fuel radiotoxicity, presence of radionuclides emitting strong gamma radiation for proliferation resistance benefit. The calculations were performed by using WIMS and DRAGON codes (together with corresponding nuclear data library based on ENDF/B-VII).

Keywords: *Thorium-based fuels, CANDU reactors, spent fuel radiotoxicity, proliferation resistance, sustainable development.t*

INTRODUCTION

The choice for nuclear power as a major contributor to the future global energy needs must take into account acceptable risks of nuclear weapon proliferation, in addition to economic competitiveness, acceptable safety standards, and acceptable waste disposal options. The main reasons for our interest in Thorium-based fuel cycles have been, globally, to extend the energy obtainable from natural Uranium and, locally, to provide a greater degree of energy self-reliance.

The Thorium terrestrial reserves are estimated to be about 4 times larger than Uranium ones (India, Brazil, Turkey, India, United States, Norway boast of large and very rich Thorium beds); Thorium is widely distributed in nature as an easily exploitable resource in many countries and has not been exploited commercially so far. Unlike natural Uranium, which contains ~0.7% *fissile* U^{235} isotope, natural Thorium does not contain any fissile material being made up of the *fertile* Th^{232} isotope only. Hence, Thorium and Thorium-based fuel has been utilized in combination with fissile U^{235} or Pu^{239} in nuclear research and power reactors for conversion to fissile U^{233}. U^{233} is by far the best fissile isotope for thermal neutron spectrum and can be used for breeding in both thermal and fast reactors. Thorium fuels, therefore, complement Uranium fuels and ensure long term sustainability of nuclear power.

The initial enthusiasm on Thorium fuels and fuel cycles was not sustained among the developing countries later, due to new discovery of Uranium deposits and their improved availability. However, in recent times, the need for proliferation-resistance, longer fuel cycles, higher burnup and improved waste form characteristics, reduction of Plutonium inventories and in situ use of bred-in fissile material has led to renewed interest in Thorium-based fuels and fuel cycles in several developed countries (see Table 1 for Thorium-based fuels and fuel cycles benefits and challenges). Two main international projects, namely Innovative Nuclear Reactors and Fuel Cycles Programme (INPRO) initiated by IAEA and US-led Generation IV International Forum (GIF), are also considering Thorium fuels and fuel cycles [1].

Table 1: Main benefits and challenges of Thorium-based fuels and fuel cycles, [1].

Benefits	**Challenges**
Thorium is 3 - 4 times more abundant than Uranium in earth's crust; is widely distributed in nature as an easily exploitable resource in many countries; has not been exploited commercially so far.	ThO_2 melting point much higher compared to UO_2 one, leads to much higher sintering temperature (> 2,000 °C) in order to produce high density ThO_2 and ThO_2–based mixed oxide fuels.
Thorium fuel cycle is an attractive way to produce long term nuclear energy with low radiotoxicity waste. The transition to Thorium could be also done through the incineration of weapons grade Plutonium or civilian Plutonium.	The process of separation of Uranium, Plutonium and Thorium from spent $(Th, Pu)O_2$ fuel, though viable, is yet to be developed.
Absorption cross-section for thermal neutrons of Th^{232} is nearly 3 times higher than the U^{238} one, leading to a higher conversion with Th^{232} (to U^{233}) than with U^{238} (to Pu^{239}). Breeding in U^{238}–Pu^{239} cycle can be obtained only with fast neutron spectra; Th^{232}–U^{233} fuel cycle can operate with fast, epithermal or thermal spectra.	Database and experience of Thorium fuels and Thorium fuel cycles are very limited, as compared to UO_2 and $(U, Pu)O_2$ fuels, and need to be augmented before large investments are made for commercial utilization of Thorium fuels and fuel cycles.
ThO_2 is chemically more stable and has higher radiation resistance than UO_2. ThO_2 is relatively inert and does not oxidize, unlike UO_2 which oxidizes easily to U_3O_8 and UO_3.	ThO_2 and ThO_2–based mixed oxide fuels are relatively inert and, unlike UO_2 and $(U, Pu)O_2$ fuels, do not dissolve easily in concentrated nitric acid. Addition of small quantities of HF in concentrated HNO_3 is essential, but causes corrosion of stainless steel.
Th–based fuels and fuel cycles have intrinsic proliferation-resistance due to formation of U^{232} via (n,2n) reactions with Th^{232}, Pa^{233} and U^{233}. Half-life of U^{232} is only 73.6 years and daughter products have very short half-life, some like Bi^{212} and Tl^{208} emit strong gamma radiations.	There is a significant buildup of radiation dose with storage of spent Th–based fuel or separated U^{233}, necessitating remote and automated reprocessing and refabrication in heavily shielded hot cells and increase in the cost of fuel cycle activities.
Th^{232}–U^{233} fuel cycle produces much lesser quantity of Plutonium and long-lived Minor Actinides (Np, Am and Cm) as compared to the U^{238}–Pu^{239} fuel cycle, thereby minimizing the spent fuel associated radiotoxicity. However, in the back end of Th^{232}–U^{233} fuel cycle, there are other radionuclides such as Pa^{231}, Th^{229} and U^{230}, which may have long term radiological impact.	In the conversion chain of Th^{232} to U^{233}, Pa^{233} is formed as an intermediate, having a relatively longer half-life (~27 days) as compared to Np^{239} (2.35 days) in the Uranium fuel cycle, thereby requiring longer cooling time of at least one year for completing the decay of Pa^{233} to U^{233}. It is essential to separate Pa from the spent fuel solution prior to solvent extraction process for separation of U^{233} and Thorium.

The Indian Thorium-based fuel cycles studies must also be mentioned. Various Thorium-based fuel cycles were examined: Thorium-U^{233} self-sustaining cycle with U^{235} makeup/with reactor grade Plutonium makeup/ with weapon Plutonium makeup; Thorium with U^{235}/with reactor Plutonium/ with weapon Plutonium in high burnup cycle; natural Thorium and SEU in separate channels; natural Thorium and natural Uranium with reactor Plutonium in separate channels/with weapon Plutonium in separate channels; natural Thorium and recovered Uranium with reactor Plutonium in separate channels/with weapon Plutonium in separate channels. The Thorium-based fuel cycles yield better fuel utilization; high burnup cycles help in shrinking spent fuel inventory for direct disposal and are very efficient in constraining Plutonium [2]. Table 2 shows the Indian nuclear power program based on the Thorium cycle. India has almost reached the 2nd stage of this fuel cycle and is preparing for the 3rd stage of the nuclear power program which is expected to begin around 2030, according to [3].

Table 2: Thorium cycle scheme, [2].

	Reactor(s)	**Fuel / Blanket**	**Product(s)**
Stage I	PHWR (CANDU)	natural Uranium	Plutonium
Stage II	Fast Breeder Reactor (FBR)	Plutonium/ Thorium and Uranium	U^{233} and Pu^{239}
Stage III	Advanced Heavy Water Reactors (AHWR)	Th^{232}; U^{233}; Plutonium	U^{233}; Th^{232}; Plutonium

CANDU reactors offer a proven technology, safe and reliable reactor technology, with an interesting evolutionary potential for proliferation resistance, their versatility for various fuel cycles creating premises for a better utilization of global fuel resources. The CANDU reactor has an impressive degree of fuel-cycle flexibility, as a consequence of its channel design, excellent neutron economy, on-power refueling, and simple fuel bundle. These features facilitate the introduction and exploitation of Thorium fuel cycles in CANDU reactors in an evolutionary fashion [4].

The interest in applying Thorium fuel cycle in CANDU reactors is based on several reasons, as follows:

- a significantly increased amount of energy can be extracted from mined Uranium by using Thorium fuel cycles, and the self-sufficient equilibrium Thorium (SSET) cycle is independent from natural Uranium or any external supply of fissile material;
- the once-through Thorium (OTT) cycle in CANDU provides an evolutionary approach to exploiting some of the energy potential of Thorium without recycling;
- ThO_2 thermal conductivity is about 50% higher than the UO_2 one over a large temperature range, and its melting temperature is by 340 °C higher than that of UO_2, leading to lower fuel-operating temperatures and decreased thermally activated processes;
- long-term supply of nuclear fuel based on the 3 to 4 times larger abundance of Thorium relative to that of Uranium;
- fissile U^{233} concentration (very valuable fissile material based on high number of neutron produced per neutron absorbed in CANDU reactors thermal spectrum) in spent Thorium-based fuel is about 5 times higher than Pu^{239} concentration in spent natural UO_2 fuel;
- benefit for normal operation, postulated accidents and waste management, taking into account that ThO_2 is chemically very stable and it does not oxidize like UO_2;
- lower toxicity of the spent Thorium-based fuel because Th^{232} produces fewer minor

actinides than U^{238} does.

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THE ONCE-THROUGH THORIUM (OTT) CYCLE IN CANDU

Since Thorium itself does not contain a fissile isotope, neutrons must be initially provided by adding a fissile material, either within or outside the ThO_2. Starting from this point two options were considered by AECL ([4]) for Thorium fuel cycle in CANDU reactors, namely: recycling (U^{233} is recycled into fresh fuel) and OTT cycles (Thorium-based fuel once passed through the reactor is either disposed or stored for possible recycling).

In order to introduce the OTT fuel cycle into existing CANDU reactors, AECL proposed both a *mixed-core approach* (a large number of driver channels containing enriched Uranium are used to support a relatively small number of channels dedicated to Thorium irradiation) and a *mixed-fuel bundle approach* (the whole reactor core is loaded with mixed-fuel bundles containing both Thorium and enriched Uranium in the same bundle). The latter illustrates a practical way of using Thorium in existing CANDU reactors, while keeping both the fuel and the reactor operating within the current safety and operating envelopes established for the natural Uranium fuel cycle [4]. The flexible CANFLEX geometry (Figure 1) was the candidate for the proposed studies.

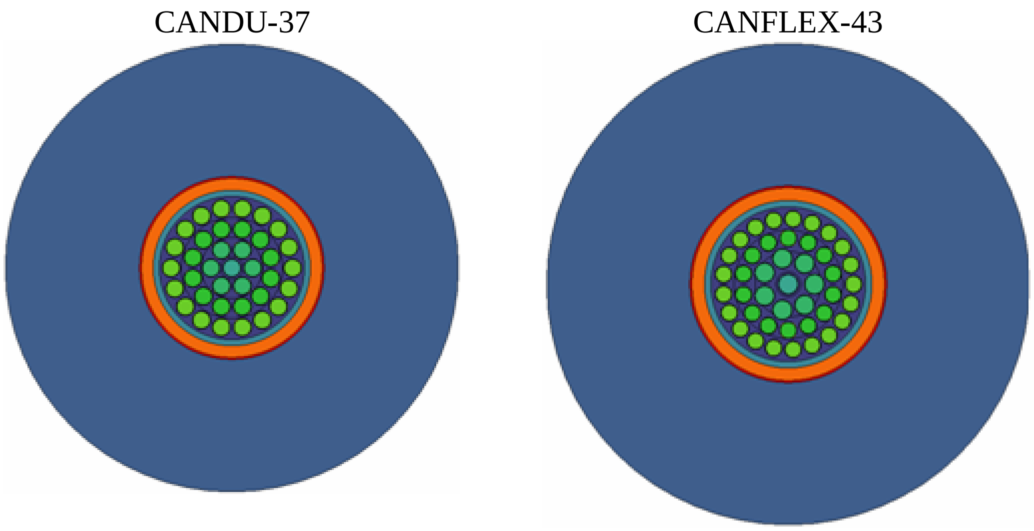

Figure 1: Lattice cell WIMS model.

The first option was to load the entire core with the same fuel type (referred to as *OTT_1*), the reactor adjuster rods being removed from the core. The fuel bundle contains natural ThO_2 in the innermost (1+7=8) elements and 1.8 wt% slightly enriched UO_2 in the outermost (14+21=35) elements.

The second option kept the adjuster rods fully inserted in the core, but three core regions were defined (Figure 2, from [4]), each being loaded with a different fuel type: the innermost 124 channels contain *OTT_2.in* bundles (similar to *OTT_1*, but the central ThO_2 element contains 6.0 wt% of Gd to shape the flux distribution), the surrounding outer 196 channels contain *OTT_1* bundles, and finally the peripheral 60 channels contain bundles with natural ThO_2 in all the 43 elements (referred to as *OTT_2.per* bundles). Since these "peripheral" bundles are assumed to achieve high burnup (> 50 MWd/kg HE), "the initial fissile content in the outer 35 elements is increased from 0 wt% to 1.7 wt% by using 20 wt% enriched Uranium" ([4]). As UO_2 and ThO_2 densities are different (10.7 and 10.0 g/cm^3 respectively), we assumed that the initial fissile content only means U^{235}. Three fuel types were considered for the peripheral channels bundles, namely *OTT_2.per_noU235* (important for studying the natural ThO_2 behavior, though not present in any real fuelling scheme), *OTT_2.per_1.7U235* and *OTT_2.per_2U235* (considered only for verification purposes), corresponding to the "initial fissile content" of 0, 1.7 and 2 wt% respectively.

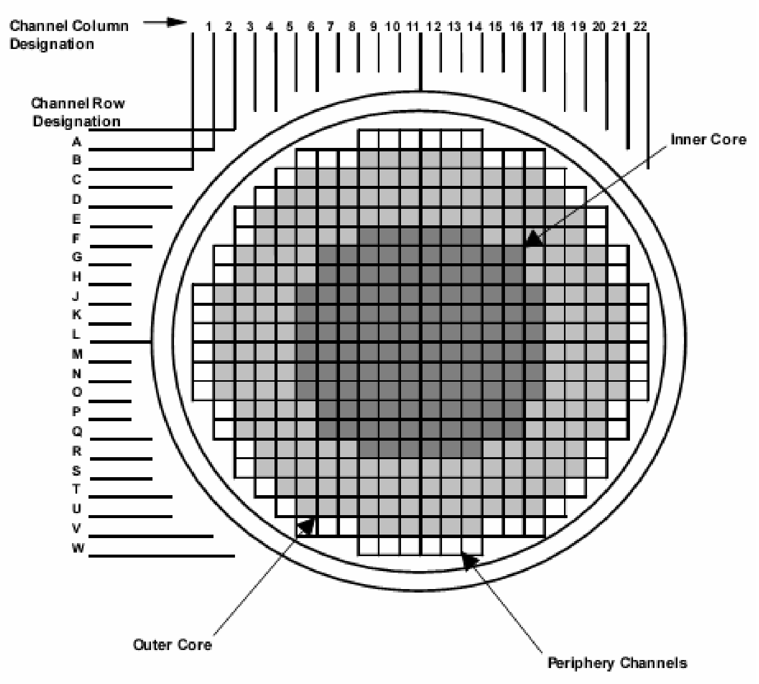

Figure 2: CANDU reactor core model for OTT option 2, [4].

RESULTS AND DISCUSSION

In the paper we only performed lattice cell calculations for the mentioned fuel bundles and also for two standard bundles, for comparison reasons. The supplementary cases are: 37NAT (standard 37 element CANDU bundle with natural UO_2) and 43NAT (43 element CANFLEX bundle with natural UO_2). The lattice codes WIMS ([5]) and DRAGON ([6]) were used, together with 69-group nuclear data library provided by IAEA-supported project WLUP ([7]).

An important assumption of our study is that the considered fuel bundles are to be used in CANDU-6 core "as is", without any supplementary reactivity- or safety-related features. Therefore, the physical properties of the fuel channel are assumed to be the same as for a standard CANDU core.

Figure 3 shows a brief comparison between the two codes when dealing with Uranium- and Thorium-based fuels. Both codes were broadly used (and validated) in calculating Uranium-based cells, but further validation and nuclear data improvements are still needed in order to ensure the suitability of results regardless the transport method calculation used for describing Thorium-based cells. The figure shows that the bundles containing Thorium can achieve higher burnup (up to 50 MWd/kg HE) than the "standard" UO_2 ones. Similar results were presented by AECL in [4].

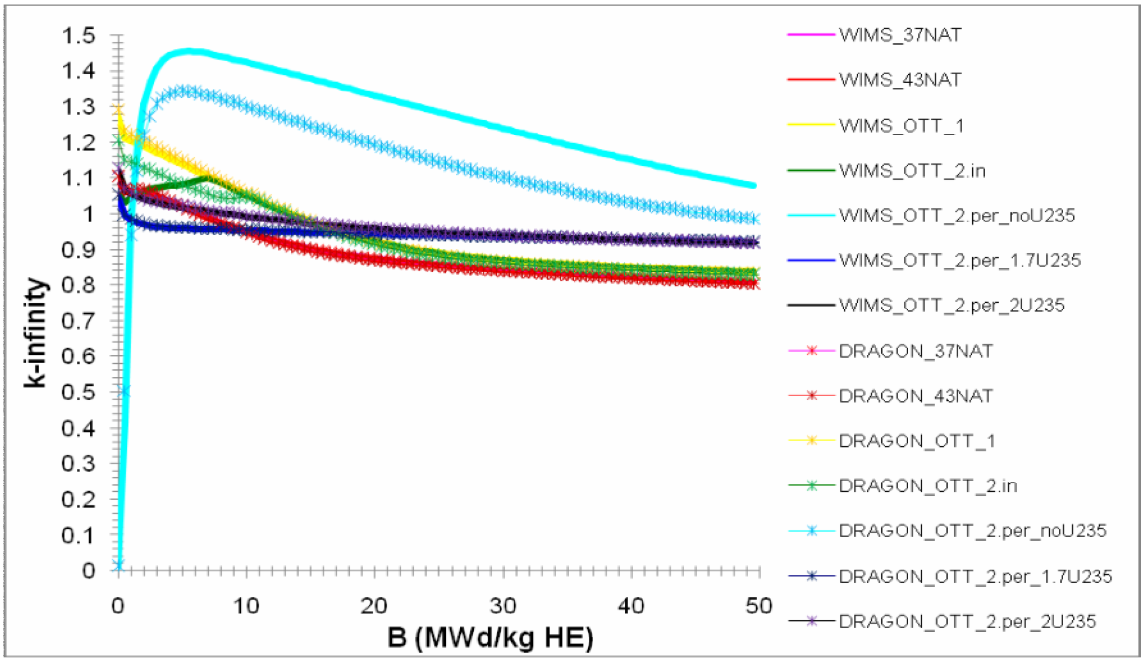

Figure 3a: Lattice k-infinity vs. fuel burnup (WIMS & DRAGON calculations).

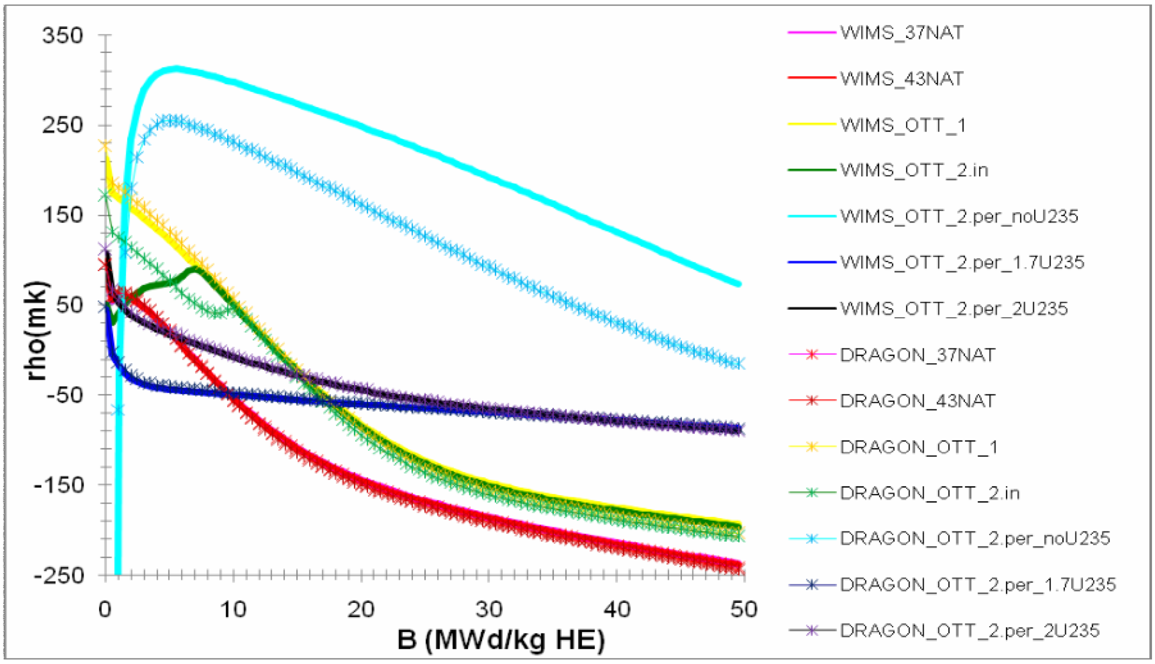

Figure 3b: Lattice reactivity vs. fuel burnup (WIMS & DRAGON calculations).

The presence of Gadolinium in the *OTT_2.in* bundle central element assures a smaller positive reactivity than that of *OTT_1*, but this effect vanishes by Gd burnout at about 8 MWd/kg HE.

As for the bundles fissile content, it decreases with burnup for most of the studied fuel types, excepting for the "peripheral" bundles that also contain enriched Uranium (*OTT_2.per_1.7U235* and *OTT_2.per_2U235*). The most important fissile isotope is U^{233} whose concentration is more than 10 times greater than U^{235} and Pu^{239} concentrations. The fissile (U^{233}, U^{235} and Pu^{239}) content evolution with burnup is shown in Figures 4 - 7.

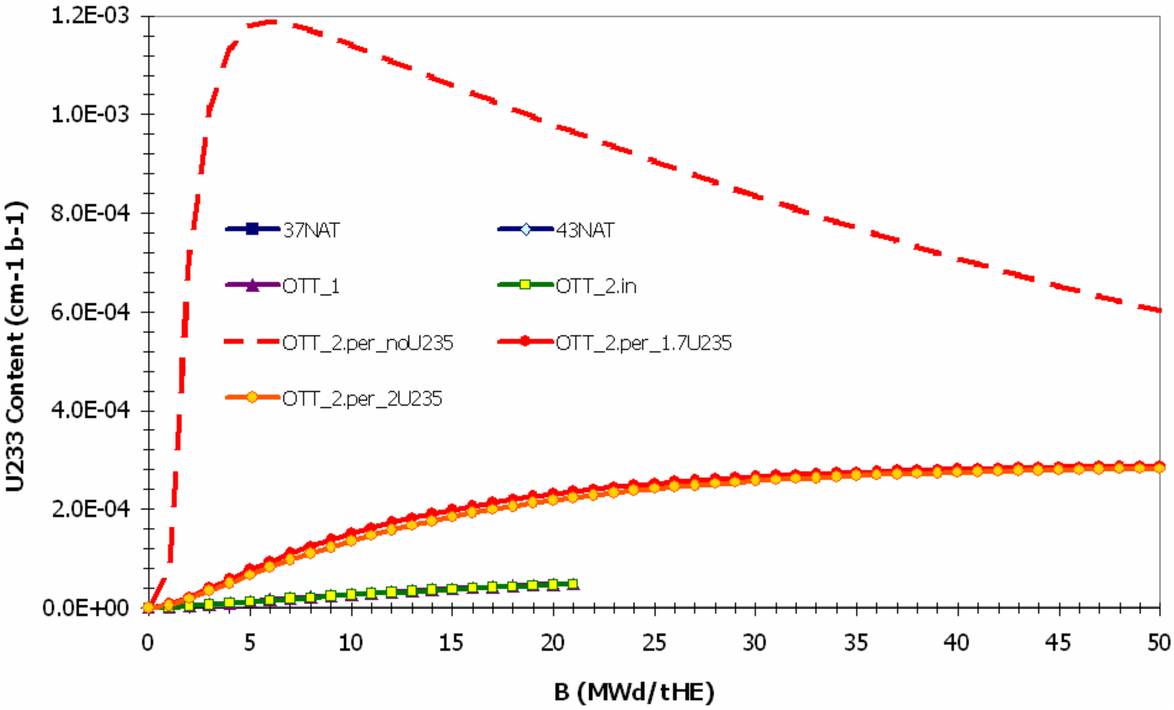

Figure 4: U^{233} content vs. fuel burnup.

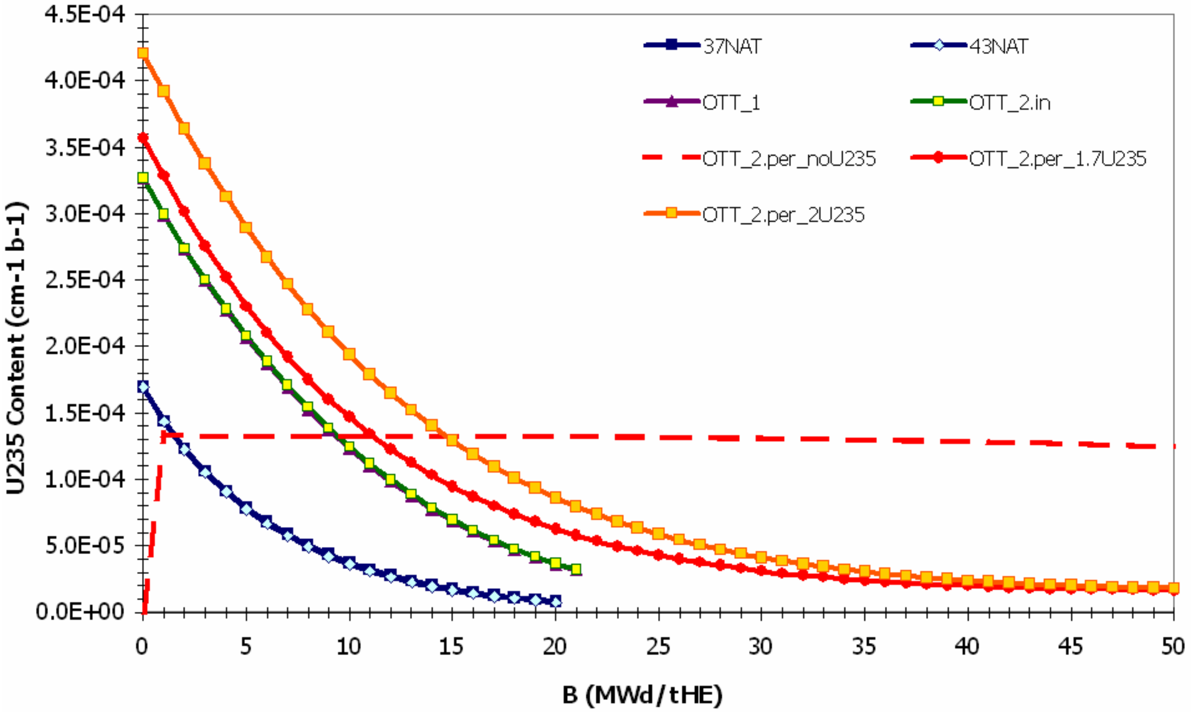

Figure 5: U^{235} content vs. fuel burnup.

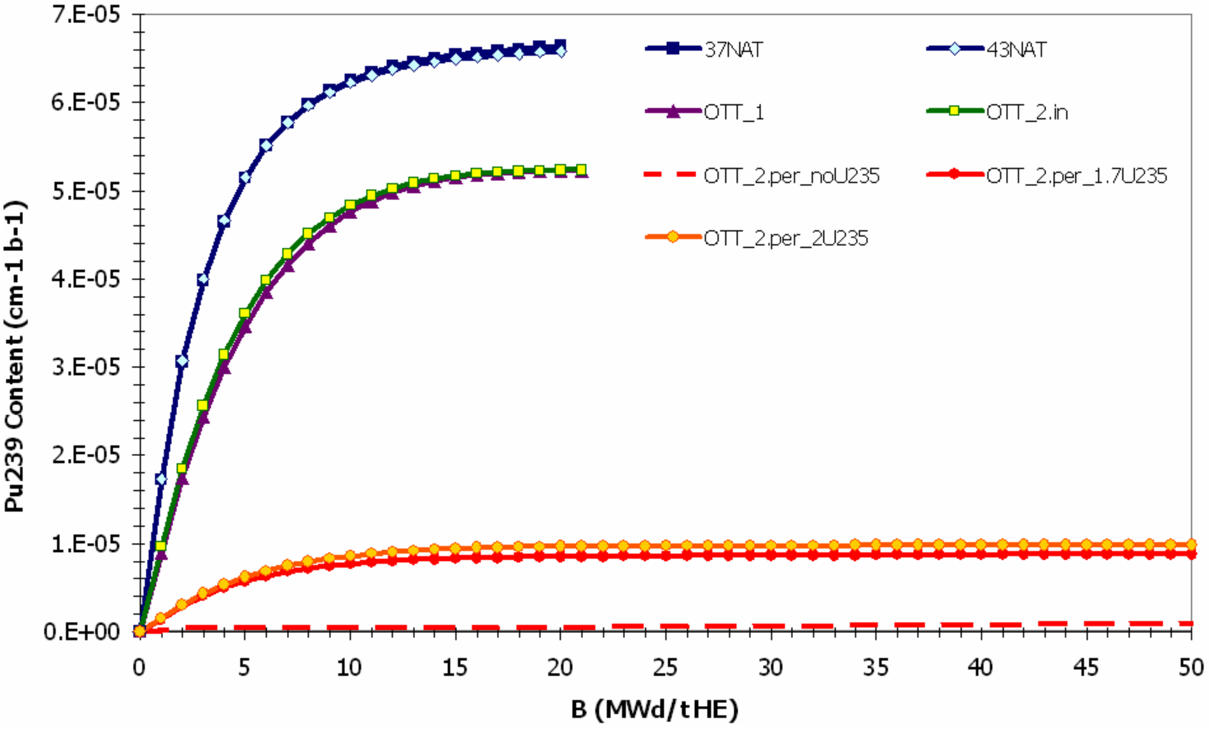

Figure 6: Pu^{239} content vs. fuel burnup.

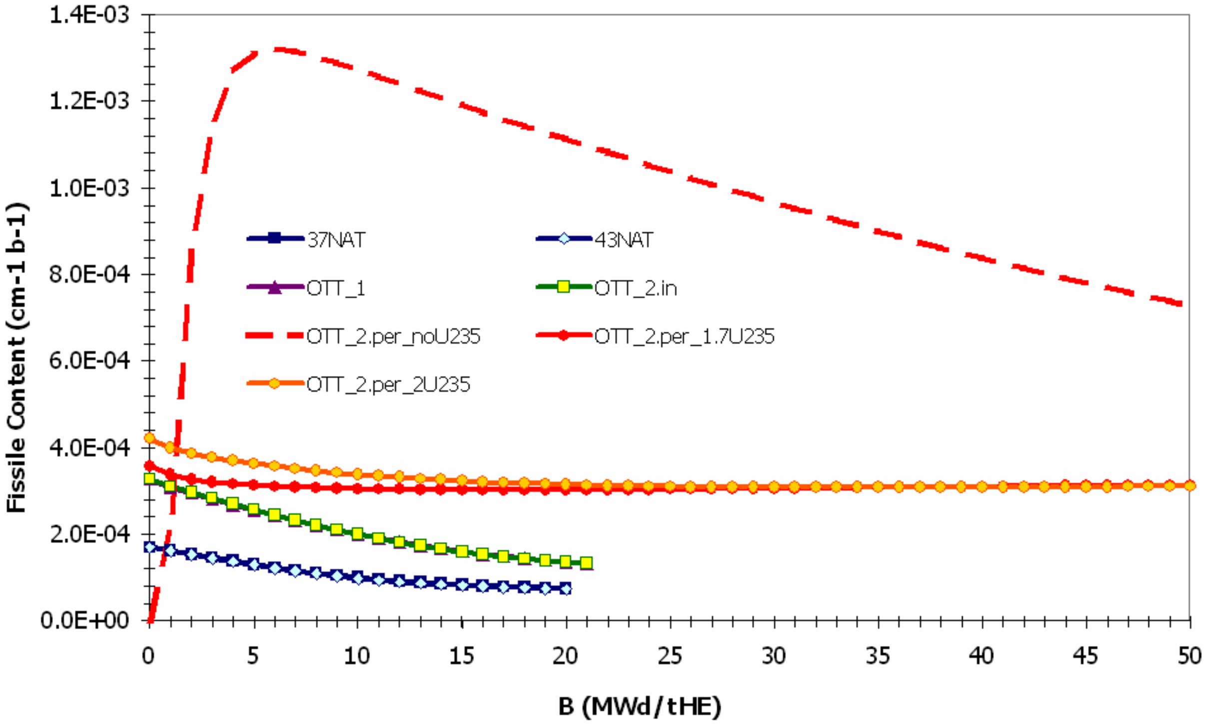

Figure 7: Fissile (U^{233}+ U^{235}+Pu^{239}) content vs. fuel burnup.

Regarding the radiotoxicity associated with Pu and minor actinides, the bundles containing Thorium seem to be safer, since burning them produces much lesser quantity of Plutonium and long-lived minor actinides (Np, Am and Cm) than Uranium-based fuels (Figures 8 - 11).

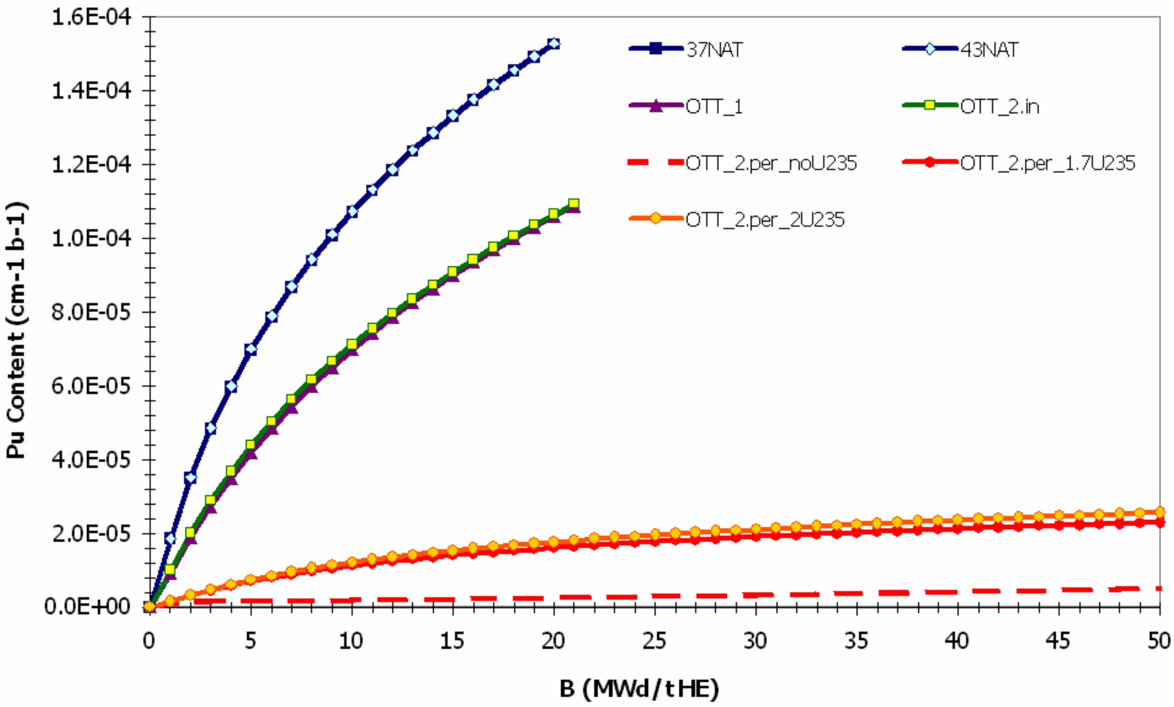

Figure 8: Plutonium content vs. fuel burnup.

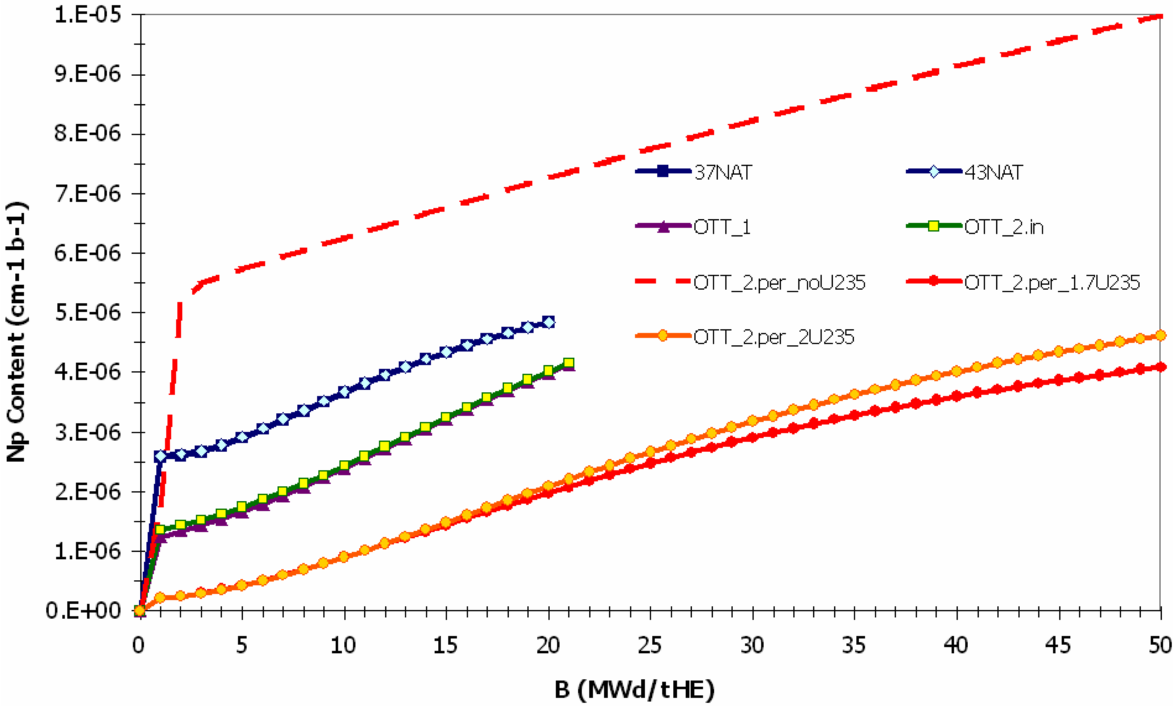

Figure 9: Neptunium content vs. fuel burnup.

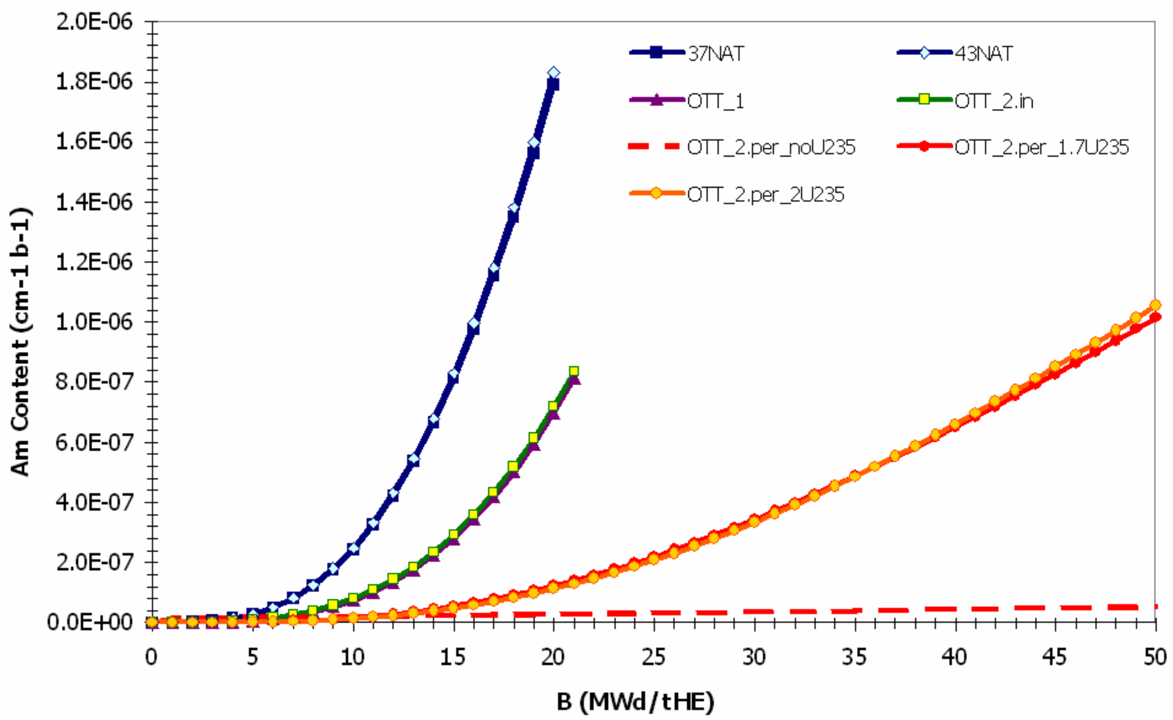

Figure 10: Americium content vs. fuel burnup.

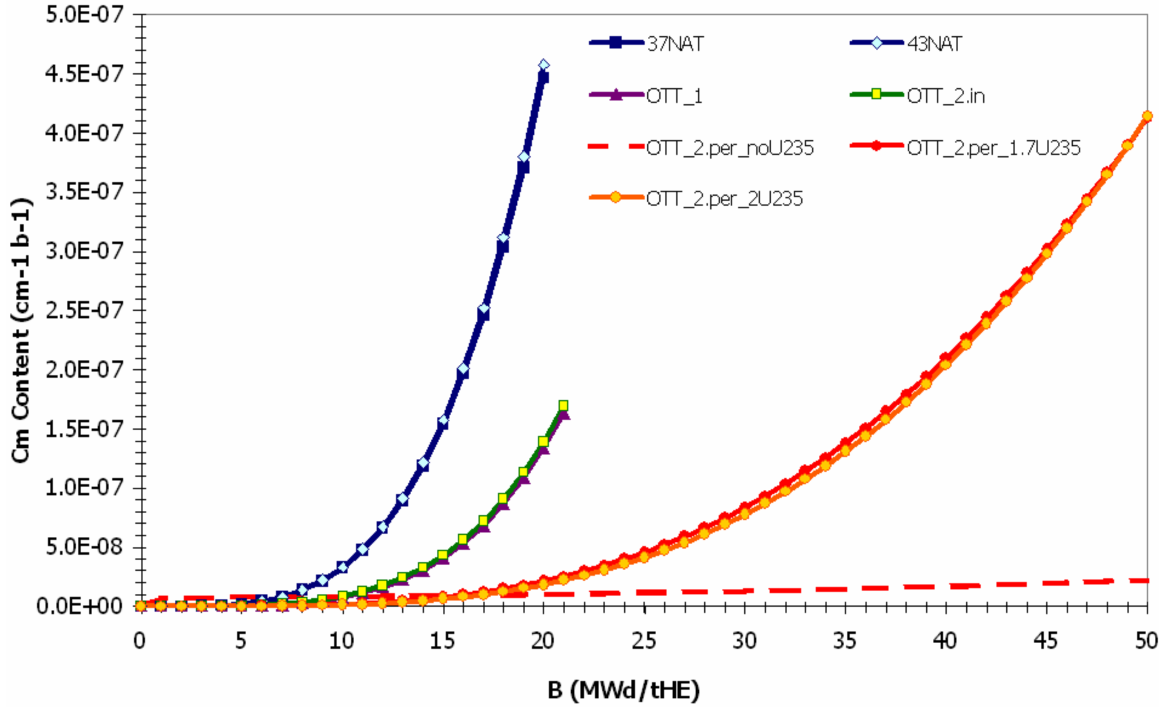

Figure 11: Curium content vs. fuel burnup.

CONCLUSIONS

The OTT cycle mixed-fuel bundle approach in CANDU reactors was proposed by AECL ([4]) to take advantage of the fuelling scheme flexibility of this type of reactors.

The lattice cell calculations performed both with WIMS and DRAGON codes for the proposed fuel types are in good agreement with the results presented by AECL, i.e. the Thorium-based bundles can achieve higher burnup, proving to be more cost-effective than the natural Uranium-based fuel bundles.

Moreover, this study confirmed that the mixed-fuel operating scheme produces less Plutonium and minor actinides than using natural UO_2, thus reducing the spent fuel radiotoxicity. However, in the back end of Th^{232}–U^{233} fuel cycle, there are other radionuclides such as Pa^{231}, Th^{229} and U^{230}, which may have long term radiological impact.

More investigations are still needed, i.e. reactor core calculations using the proposed fuelling scheme, to prove the viability of a CANDU core fuelled with Thorium-based CANFLEX bundles, in terms of flux & power distributions, reactivity control effectiveness, trip operation etc.

A reliable set of updated and documented nuclear data for Thorium is needed for further analyses.

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