MOVING TOWARDS SUSTAINABLE THORIUM FUEL CYCLES

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Abstract

The CANDU® reactor has an unsurpassed degree of fuel-cycle flexibility as a consequence of its fuel-channel design, excellent neutron economy, on-power refueling, and simple fuel bundle design. These features facilitate the introduction and full exploitation of thorium fuel cycles in CANDU reactors in an evolutionary fashion.

Thoria (ThO_2) based fuel offers both fuel performance and safety advantages over urania (UO_2) based fuel, due its higher thermal conductivity which results in lower fuel-operating temperatures at similar linear element powers. Thoria fuel has demonstrated lower fission gas release than UO_2 under similar operating powers during test irradiations. In addition, thoria has a higher melting point than urania and is far less reactive in hypothetical accident scenarios owing to the fact that it has only one oxidation state.

This paper examines one possible strategy for the introduction of thorium fuel cycles into CANDU reactors. In the short term, the initial fissile material would be provided in a heterogeneous bundle of low-enriched uranium and thorium. The medium term scenario uses homogeneous Pu/Th bundles in the CANDU reactor, further increasing the energy derived from the thorium. In the long term, the full energy potential from thorium would be realized through the recycle of the U-233 in the used fuel. With U-233 recycle in CANDU reactors, plutonium would then only be required to top up the fissile content to achieve the desired burnup.

1. Introduction

There has been increased interest world-wide in thorium as a fuel in nuclear power reactors. The CANDU reactor has unsurpassed fuel cycle flexibility, allowing it to use a wide range of fuels, including thorium. The high neutron economy allows for more neutrons in the core to breed the fissile U-233 from Th-232, with lower amounts of driver material than may otherwise be required. Online refueling and a small, simple fuel design allow current CANDU 6 reactors to use thorium fuel with minimal changes to the reactor design [1].

Thoria (ThO_2) based fuel offers both fuel performance and safety advantages over urania (UO_2) based fuel. The thermal conductivity of thoria is significantly higher than urania resulting in lower fuel-operating temperatures compared to urania fuel operating at the same power. Consequently, any thermally activated processes, such as fission gas release, should be lower for thoria fuel [2]. Thoria fuel has demonstrated lower fission gas release than UO_2 under similar operating temperatures during test irradiations. Beyond the advantage of lower operating temperature, thoria has a higher melting point than urania and is far less reactive in hypothetical accident scenarios owing to the fact that it has only one oxidation state. In this paper only thoria is considered as the fuel material.

This paper describes a phased approach [3] to introduce thorium fuel cycles in CANDU reactors, thereby contributing to energy and resource sustainability in the short, medium and long term. In the first phase, benefit is derived in the short term from thorium cycles using existing fuel and core designs using low enriched uranium (LEU) as the driver fuel.

In the medium term a homogeneous mixture of thorium and reactor-grade plutonium is used. As thorium does not have a fissile isotope, an initial fissile inventory needs to be provided until enough U-233 can be bred from the thorium to sustain the reaction. This fuel cycle can be implemented once full commercial reprocessing of used LWR fuel has been established, with surplus plutonium used to drive the thorium cycle in CANDU reactors. This homogeneous bundle design significantly increases the energy that is derived from thorium over the heterogeneous near-term cycle.

In the long term, the maximum benefit is derived from thorium through recycling the U-233 (and Th-232) in the used fuel of CANDU reactors. In this cycle the amount of U-233 at the beginning and end of cycles is roughly equivalent, with a small amount of plutonium required to achieve the desired exit burnup.

2. Benefits and Challenges of Thorium Fuel

As a result of its "fertile only" naturally occurring state, the attraction to thorium as an energy source necessitates irradiated fuel recycle. As such, thoria fuel is a dichotomy; what makes it good as fuel makes it bad for recycle. As mentioned previously, thoria is very stable, bordering on inert, and many of the uses of thoria outside the nuclear industry have been centred on this property. Thoria has the highest melting point of the oxides (3200°C) and cannot oxidize. This latter fact is an important difference between thoria and UO_2 fuels in defective fuel and accident scenarios. Urania will oxidize when exposed to coolant at high temperatures resulting in a disintegration of the fuel due to the spalling off of higher oxides of uranium from the UO_2 pellet surface. This "washout" in the case of defective fuel results in the spread of tramp uranium throughout the primary coolant circuit, increasing the dose to workers and complicating reactor maintenance. Historical tests on intentionally defected thoria fuel at Chalk River Laboratories have indicated minimal washout or interaction with the coolant.

The thermal conductivity of pure thoria is almost double that of UO_2 at low temperatures, but the difference in thermal conductivities decreases with increasing temperature and they converge at approximately 1600°C. Still, the integrated thermal conductivity across the pellet radius is better for thoria than UO_2 and this difference is proportional to the element rating. The consequences of better thermal conductivity are lower centreline temperatures for thoria elements compared to urania elements under the same operating conditions. This is beneficial to fuel performance as a primary factor in gas release is the operating temperature of the fuel. The minimization of gas release is desirable due to its negative impact on stress-corrosion cracking, or pellet-clad interaction, and the possibility of internal gas overpressure.

A multitude of irradiation tests have been carried out at Chalk River Laboratories [2]. Unfortunately, a large number of the tests conducted with pellets of thoria containing Highly Enriched Uranium (Th-HEU) were carried out on fuel that had an undesirable microstructure. While the fuel pellets had high density, the microstructures were "granular" in nature (very high density regions surrounded by porous regions or voids). The affect of the granular microstructure was two-fold; the regions of low-density or void acted like an insulator degrading the effective thermal conductivity and thus increasing the fuel operating temperature, and those same regions then acted as a conduit for the gas to escape the pellet and be released into the gap. As a result several irradiation experiments conducted with this type of fuel resulted in higher gas release than expected for thoria fuel. The gas release was similar to what would be expected from UO_2 fuel operated under similar conditions. Recent irradiation tests of Th-HEU and pure thoria fuel with an appropriate microstructure and previous tests of Th-plutonium fuel [4] have verified the improved gas release performance expected from thoria-based fuels.

While once-through thorium fuel cycles offer some benefit in the reduction of uranium resources, to maximize the benefits of thorium the U-233 must be extracted from the irradiated fuel and recycled. Thoria is very difficult to dissolve even in strong acids that easily dissolve UO_2. While aqueous processes similar to the PUREX process used for UO_2 exist and have been applied to irradiated thoria fuel, they are not yet optimised and face challenges that are still being addressed today[5][6]. This barrier to an industrially viable recycling process is one of the major impediments to global acceptance and implementation of thorium-based fuel cycles.

3. The Short Term Heterogeneous Bundle Option

Since thorium has no fissile isotope, the fissile component that is initially added to the fuel or to the core defines a large range of thorium fuel cycle options [7]. The easiest way to initiate the thorium fuel cycle in the CANDU reactor is by adding the fissile component as LEU in separate elements in a mixed LEU/Th fuel bundle [8]. This does not depend on reprocessing to recover plutonium and avoids the expense and complication of fabrication and handling of highly-radiotoxic fuels. In the mixed LEU/Th CANDU bundle design, LEU fuel is in the outer elements of the fuel bundle, with ThO_2 in the center and the inner ring of elements (Figure 1). This placement of fertile and fissile material locates the ThO_2 in the low thermal flux region of the bundle, where it is shielded by the outer LEU fuel elements. This improves the conversion ratio by reducing neutron capture in Pa-233 (which would result in production of U-234), favouring its decay to U-233. The mixed LEU/Th bundle configuration also reduces coolant void reactivity (CVR), since the ThO_2 in the central elements initially acts as a neutron absorber.

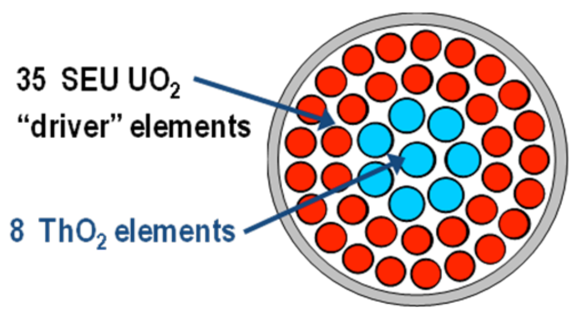

Figure 1. LEU/Th CANDU fuel bundle

The 43-element CANFLEX bundle is favoured for this application [9]: it has more elements in which to produce power than does the standard 37-element CANDU fuel, and the inner 8 elements are larger than the outer 35 elements (both factors reduce the linear element ratings relative to the 37-element bundle); the central 8 elements containing thorium comprise about 20% of the fuel mass; the bundle employs critical heat flux-enhancing buttons that improve the thermalhydraulic margins. This use of an existing fuel design is a low-risk approach for initiating the thorium fuel cycle. The mechanical performance of the bundle has been fully qualified through three separate demonstration irradiations of the CANFLEX fuel bundle in power reactors: with natural uranium fuel in the CANDU 6 reactors at the Point Lepreau [10] and Wolsong 1 [11] nuclear power plants and in a unit of the Bruce B reactors in the Low Void Reactivity Fuel (LVRF) configuration [12]. The enrichment of the LEU elements can be varied to give the desired burnup, which can be gradually increased with experience.

Benefit is derived from the fissioning in-situ of the U-233 produced through neutron capture in Th-232. The U-233 level gradually increases with burnup, reaching a level of around 1.4%. To extract the maximum benefit from the U-233 produced without recycle, a bundle-average burnup of at least 20 MWd/kg is desirable. In this cycle, the uranium utilization is about 20% better than for natural uranium in the CANDU reactor. Benefit is derived from the utilization of thorium today at no additional cost (either in terms of fuel cycle costs or uranium utilization). The U-233 in the used fuel can be stored, and then recovered and recycled in the future, to provide the maximum benefit.

4. The Medium Term Homogeneous Pu-Driven Thorium Fuel Cycle

The second phase of implementation of thorium fuel cycles in CANDU reactors makes use of recycled plutonium from light water reactors[13]. Used CANDU fuel is also a potential source of plutonium. However, due to the prevalence of LWRs and the existence of operating facilities that recycle used LWR fuel, that source is used for this study. This plutonium is used as a fissile driver material, in order to sustain the chain reaction until enough U-233 has been produced by the thorium to power the reactions. This fuel cycle will create a store of U-233 in the spent fuel which would be used in the long-term thorium fuel cycle.

Another option for a fissile driver material is low-enriched uranium, on the order of 20% U-235[13]. Low-enriched uranium could be used where Pu reserves are insufficient, but uranium enrichment facilities exist. The use of LEU does create a complication in that the uranium that is in the spent fuel is not isotopically pure as it is in the case of a Pu-driven fuel. This will make the use of the U-233 later more complicated, but not insurmountable.

Two options for Pu/Th fuel are shown here, a low burnup case obtaining 20 MWd/kg initial heavy element (IHE) and a higher burnup case to 40 MWd/kg IHE. There is significantly better thorium utilization with the higher burnup.

4.1 Fuel Design

A different fuel bundle design was used for the low and high burnup cases. The bundle designs were changed to optimize for the amount of energy derived from thorium while maintaining a maximum linear element rating (LER) of 60 kW/m for the outer elements.

Each bundle design has a centre element consisting of a tube of zirconia-filled hafnium. This configuration serves to introduce a neutron absorber to the center of the bundle in a way that is simple and easy when creating computer models. The composition of the center poison can be changed later when a particular bundle design is chosen for fuel cycle development.

For the high burnup case, the fuel was graded, with more fissile content in the outer rings. This grading helps minimize the amount of poison needed in the central absorbing element, which in turn leads to better thorium utilization. However, this grading also leads to larger radial form factors. In order to decrease the linear element ratings the size of the fuel pins was reduced and the number of fuel pins increased. This change in geometry results in around 10% less fuel in the bundle. The low burnup cases have 42 fuel elements, with 7, 14, and 21 elements in the inner, intermediate and outer rings, respectively. The high burnup cases have a larger centre pin, and 12, 18, and 24 fuel elements in the inner, intermediate, and outer rings, respectively, see Figure 2. The bundle composition is given in Table 1.

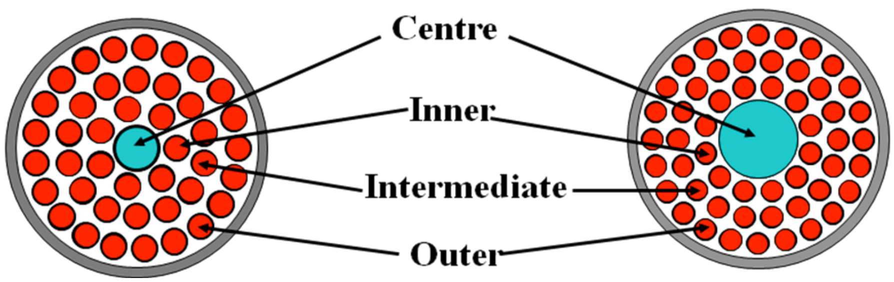

Figure 2. Fuel bundle design for the low burnup cases (left) and the high burnup cases (right).

Table 1. Bundle composition for the Pu-driven homogeneous thorium fuel cycle

Burnup	Total Number of Fuel Elements	Bundle Average Pu wt%
Low	42	3.5
High	54	4.9

4.2 Calculation

This paper addresses physics involved in lattice cell calculations and does not consider details of full-core implementation of thorium fuel cycles in CANDU 6 reactors. All models for this report were lattice cell calculations performed using WIMS-AECL v. 3.1.2.1 [14] with and ENDF/B-VI-based library[15].The isotopic composition of the reactor-grade driver plutonium is given in Table I.

Table 2. Input Isotopic Composition for the Plutonium Driver Fuel

Nuclide	% by Weight
Pu-238	2.5
Pu-239	54.2
Pu-240	23.8
Pu-241	12.6
Pu-242	6.8

The power normalization used a constant flux, chosen such that the power over the total burnup averaged to approximately 32 W/g. Leakage and absorption by unmodelled reactor components were assumed to be worth 30 mk in total, which is typical of a CANDU 6 reactor with the adjustor rods removed. A total bundle power of 800 kW was assumed for the calculation of linear element ratings. These models were developed to maximize the amount of energy derived from the input thorium

To calculate the amount of energy that came from the thorium in the fuel, the reaction data from the WIMS output was extracted. The fission reaction for nuclides derived from thorium, U-233, U-235 and Th-232, was compared to that from the driver fuel, Pu-239 and Pu-241. This reaction data, along with the power and the length of the timestep was used to calculate the amount of energy that came from thorium versus the driver fuel. It was assumed that the same amount of energy results from each fission, independent of which nuclide was the source of the fission.

To calculate the fuel temperature coefficients (FTC), WIMS models were created such that the temperature of the fuel was increased and decreased by 50°C at each burnup step. The change in reactivity across this 100°C range was used to calculate the burnup-weighted average FTC.

4.3 Results

Values obtained for the burnup, fuel temperature coefficient, maximum linear element rating, and percentage of energy derived from thorium are shown in Table 3. All of the cases run have negative fuel temperature coefficients that are comparable to those for a CANDU 6 reactor with natural uranium fuel, and a maximum linear element rating less than 60 kW/m. Figure 3 and Figure 4 show the distribution of fissions derived from the thorium fuel vs. fissions from plutonium, and the distribution of Pa-233 and U-233 in the bundle for the low burnup case. These graphs for the high burnup Pu-driven thorium case are in Figure 5 and Figure 6. For these once-through fuel cycles, the driver fuel is the only fissile material present at the beginning of the irradiation and, therefore produces all of the power. As the irradiation proceeds, U-233 is bred in and, produces an increasing fraction of the power as the irradiation proceeds..

At the beginning of the irradiation the fissions are coming primarily from the driver fuel. For the once-through low burnup Pu-driven case (Figure 3), Pu remains the dominant source for the whole irradiation. For the high burnup Pu-driven case the majority of the fissions switches during the irradiation so that by the end U-233 becomes the dominant source. This cross-over point, after which most of the power is derived from the thorium fuel, occurs at 33 MWd/kg (Figure 5). The U-233 grows in the outer rings quickest; this is where the flux is highest (Figure 4 and Figure 6), and more neutrons are available to capture onto Th-232 and lead to the creation of U-233.

Table 3. Results for the Once-Through Homogeneous Thorium Fuel Cycle Cases

Burnup (MWd/kg IHE)	Fuel Temperature Coefficient (μk/°C)	Maximum Linear Element Rating (kW/m)	% Energy Derived from Thorium
19.4	-3.8	56	18.9
45.0	-5.0	61	29.2

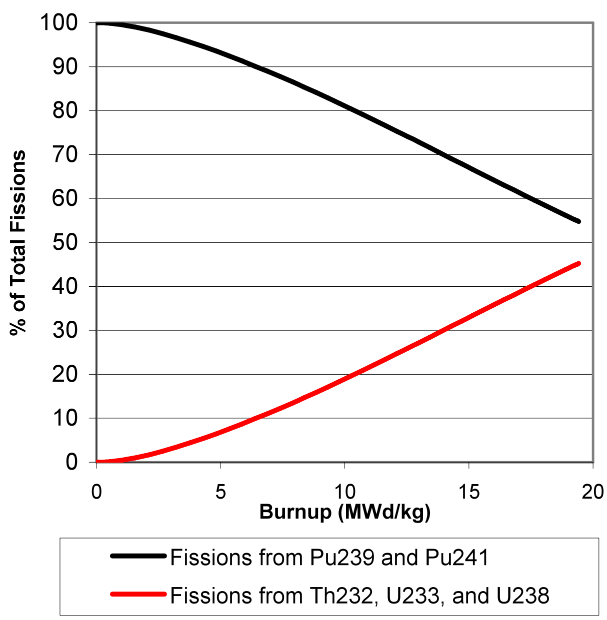

Figure 3. Distribution of fissions for the low burnup once-through Pu driven thorium case.

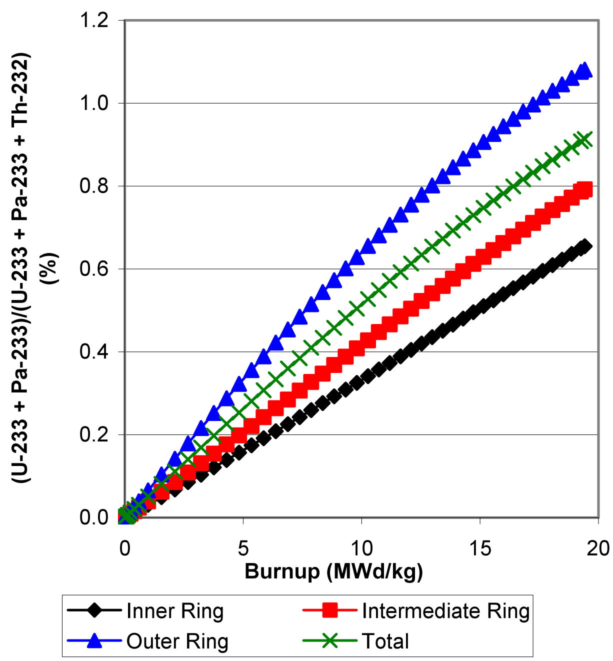

Figure 4. Distribution of U-233 and Pa-233 in the bundle for the low burnup once-through Pu driven thorium case.

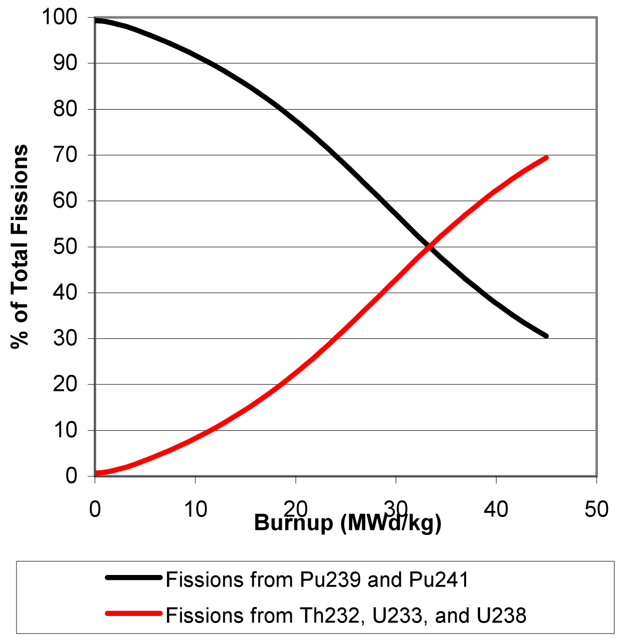

Figure 5. Distribution of fissions for the high burnup once-through Pu driven thorium case.

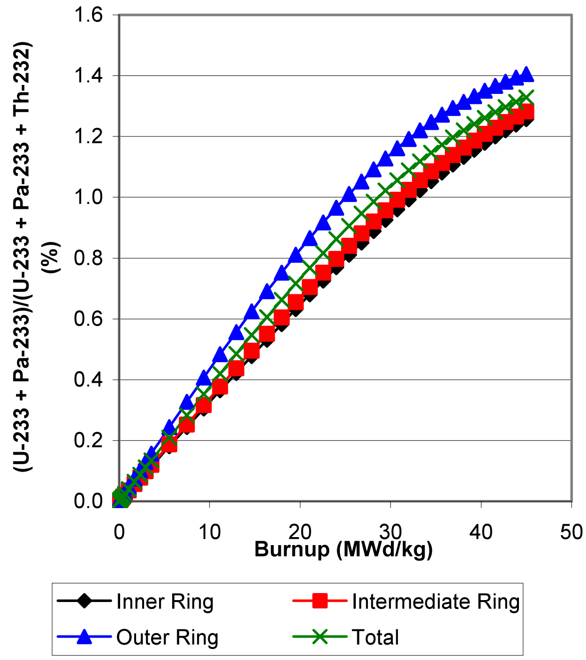

Figure 6. Distribution of U-233 and Pa-233 in the bundle for the high burnup once-through Pu driven thorium case.

5. The Long Term Thorium Fuel Cycle Using Recycled U-233

On a long-term horizon, potential shortages of natural uranium resources will increase the appeal of closed fuel cycles. Closing the fuel cycle enables the full benefit of thorium to be realized, but is more complicated, and likely more expensive. The implementation of this fuel cycle requires an available supply of U-233 to use in the fresh fuel, and it requires mature technology to recycle the used fuel to extract the U-233 from the spent fuel.

The reactor implementation of the closed cycle is similar to the once-through cycle discussed in Section 4. Homogeneous mixtures of U-233, plutonium and thorium are used. A small amount of Pu is required in addition to the U-233 in order to achieve the desired burnup. Lower burnups require less U-233, but in turn will have a shorter residence time in the reactor, less energy derived per bundle, and hence will require a larger reprocessing capacity. The selection of the burnup and reprocessing capacity would be determined by the value of plutonium, U-233, and the expense of reprocessing. In this study two exit burnups are examined: a low burnup of 20 MWd/kg IHE, and a higher burnup of 45 MWd/kg IHE. It is noted that going to even lower burnups, on the order of those of current generation CANDU reactors using natural uranium fuel, could enable a fuel cycle that is entirely self-sufficient in U-233, that is, it would not require any top-up of plutonium.

5.1 Fuel Design

The fuel design for these bundles is similar to that for the once-through case, Section 4.1 with similar geometry and isotopic composition of plutonium, see Figure 2 and Table 2. The bundle composition is different, as the fresh fuel now contains Pu, U-233 and thorium. The placement of U-233 and Pu is graded across the bundle, that is, it is different in each ring. The composition is given in Table 4. The placement of the U-233 in the bundles in the recycle cases is not in a configuration designed to maximize the breeding of U-233. The requirement for these models was to have roughly the same amount of U-233 input and output (or slightly more on output to allow for losses during reprocessing). The same calculation methods were also used, including the same code, libraries, and input parameters, as discussed in Section 1.1.

Table 4. Bundle compositions of the thorium with recycled U-233 cases

Burnup	Total Number of Fuel Elements	Bundle Average Pu wt%	Bundle Average U-233 wt%
Low	42	0.8	1.4
High	54	2.1	1.4

5.2 Results

Values obtained for the burnup, fuel temperature coefficient, maximum linear element rating, and percentage of energy derived from thorium are shown in Table 5. All of the cases run have negative fuel temperature coefficients that are comparable to those for a CANDU 6 reactor with natural uranium fuel, and a maximum linear element rating less than 60 kW/m. Figure 7 shows the distribution of fissions derived from the thorium fuel vs. fissions from plutonium, and Figure 8 gives the distribution of Pa-233 and U-233 in the bundle for the low burnup case. These graphs for the high burnup case are in Figure 9 and Figure 10.

For these cases with U-233 recycle, the initial division of power depends primarily on the distribution of fissile material in the fresh fuel. As the irradiation progresses, the amount of plutonium decreases, while the total amount of U-233 does not, resulting in an increasing fraction of the power being generated by U-233 fissions.

Table 5. Results for the thorium with recycled U-233 cases

Burnup (MWd/kg IHE)	Fuel Temperature Coefficient (μk/°C)	Maximum Linear Element Rating (kW/m)	% Energy Derived from Thorium
19.7	-7.5	49	78.1
44.0	-7.3	59	65.7

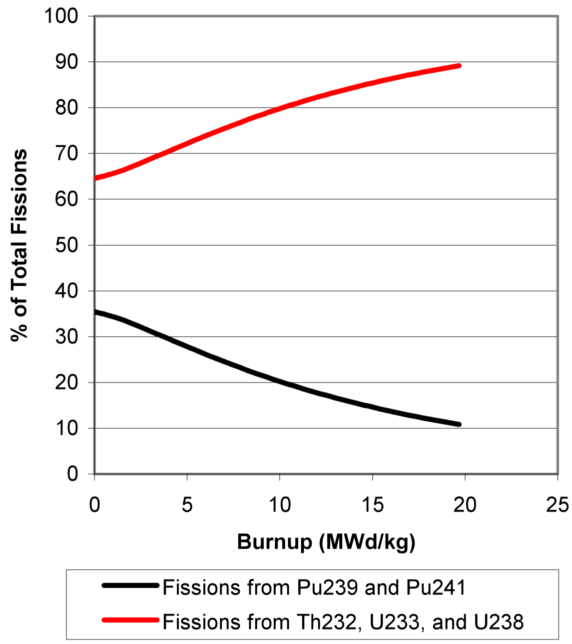

Figure 7. Distribution of fissions for the low burnup Pu driven thorium with U-233 recycle case.

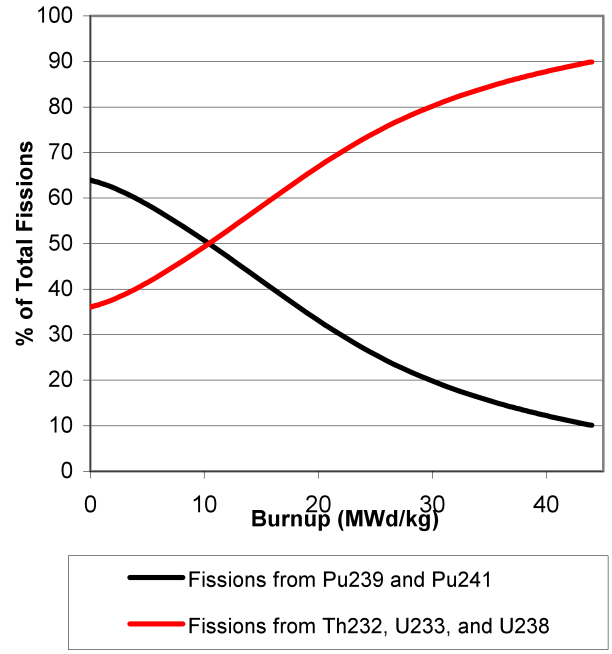

Figure 9. Distribution of fissions for the high burnup Pu driven thorium with U-233 recycle case.

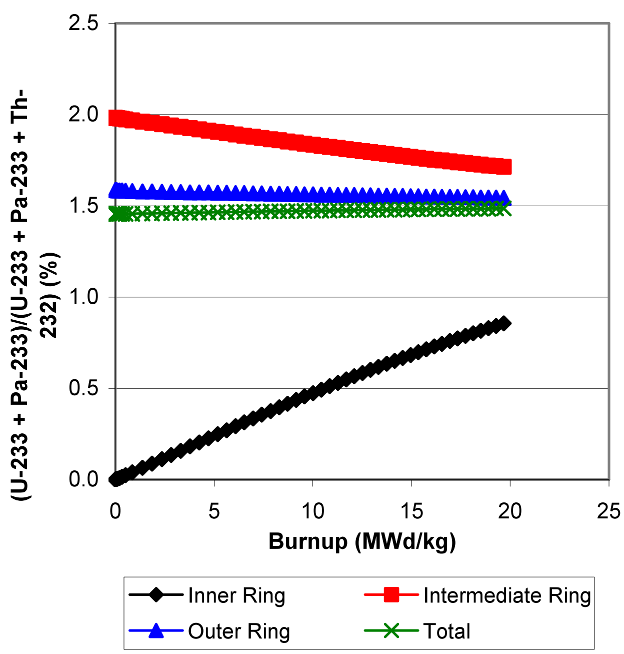

Figure 8. Distribution of U-233 and Pa-233 for the low burnup Pu driven thorium with U-233 recycle case.

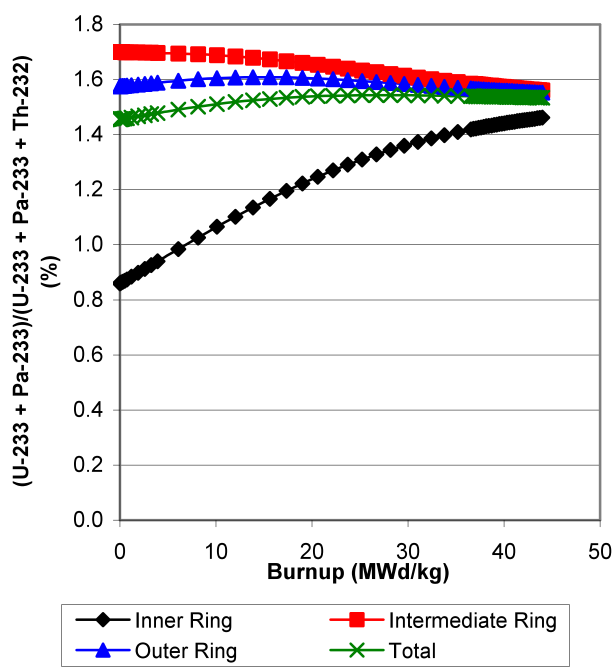

Figure 10. Distribution of U-233 and Pa-233 for the high burnup Pu driven thorium with U-233 recycle case.

The low burnup Pu-driven case with U-233 recycle has the majority of fissions coming from U-233 for the entire irradiation (Figure 6). This is because only a small amount of additional Pu is needed to achieve the exit burnup, so from the beginning of the irradiation U-233 is the dominant fissile isotope. This shows that there is more benefit to using thorium in cycles with recycle of U-233 and/or with higher burnup.

For the recycle cases the U-233 is initially placed in the outer and intermediate rings (Figures 7 and 9). The U-233 decreases from the intermediate ring, stays approximately constant in the outer ring, and grows into the inner ring. These fuel designs were chosen so that total amount of U-233 in the fuel bundle stays approximately constant throughout the irradiation, i.e. the rate of U-233 absorption is equal to the U-233 production.

6. Conclusion

Thorium offers considerable advantages over uranium as a fertile nuclear fuel material: higher thermal conductivity, higher stability, and higher melting point offer fuel performance and safety improvements due to lower operating temperatures, no non-stoicheometric states, and lower fission gas release.

The CANDU reactor presents an opportunity to ensure long-term resource sustainability through a staged implementation of the thorium cycle. In the fuel cycle vision presented in this paper, each phase reflects the projected economic, resource and infrastructure situation at that time. The near-term strategy reflects the current abundance, availability and cost of natural uranium, and uses LEU to convert Th-232 into U-233 at no additional cost today (either in terms of fuel cycle costs or uranium utilization). In this phase, benefit is derived from the in-situ burning of U-233, while safeguarding it in the used fuel for future recycling. The medium term would utilize the plutonium from reprocessing used LWR fuel as Pu/Th fuel in the CANDU reactor, further extending the energy obtained from thorium. Long-term resource sustainability can be ensured by closing the thorium cycle by recycling of U-233/Th in the CANDU reactor.

7. References

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