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THORIUM FUEL CYCLES IN THE CANDU SUPERCRITICAL WATER REACTOR

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CW-123700-CONF-012

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THORIUM FUEL CYCLES IN THE CANDU SUPERCRITICAL WATER REACTOR

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Abstract

The CANDU® supercritical water-cooled reactor (SCWR) is Canada's primary contribution to the Generation IV International Forum (GIF). The goals of GIF include the development of next-generation reactors with enhanced safety, resource sustainability, economic benefit and proliferation resistance. There is great potential for enhancing the sustainability of the nuclear fuel cycle by extending the availability of current resources through the use of thorium fuel cycles. Recent studies of thorium-based fuel cycles in contemporary CANDU reactors demonstrate the possibility for substantial reductions in natural uranium (NU) requirements of the fuel cycle via the recycling of U-233 bred from thorium [1-4]. As thorium itself is not fissile, neutrons must be provided by adding a fissile material, either within or outside of the thorium-based fuel. Various thorium fuel cycles can be categorized by the type and geometry of the added fissile material. The simplest of these fuel cycles are based on homogeneous thorium fuel designs, where the fissile material is mixed uniformly with the fertile thorium. These fuel cycles can be competitive in resource utilization with the best uranium-based fuel cycles, while building up an inventory of U-233 in the spent fuel for possible recycling in thermal reactors. When U-233 is recycled from the spent fuel, thorium-based fuel cycles can provide substantial improvements in the efficiency of energy production from existing fissile resources. In this paper, two homogeneous CANDU-SCWR thorium-based fuel cycles using reactor-grade plutonium as the fissile driver material have been examined. As the CANDU-SCWR reactor concept is still in the early development and design stages, various lattice and channel parameters can be varied to optimize the reactor for a specific fuel type. The impact of varying some of these parameters to optimize for thorium fuel has been studied. In this paper, thorium fuel cycle options are examined and compared with respect to initial Pu driver fuel requirements, U-233 recycling, and exit burnup.

1. Introduction

Canada is developing the CANDU supercritical water-cooled reactor (SCWR) as its primary contribution to the GIF [5]. Reactor development by the GIF is focussed on four goals: enhanced safety, resource sustainability, economic benefit and proliferation resistance [6]. Thorium-based fuel cycles are of interest in the GIF since these cycles have features that are well aligned with the mandates of GIF, as discussed in [7], and summarized below:

- Thorium dioxide (ThO_2) is superior to uranium dioxide (UO_2) with respect to its chemical stability, fission product release, thermal conductivity and coefficient of thermal expansion. Consequently, use of ThO_2-based fuels will enhance fuel safety (via reduced fuel failure) compared to UO_2-based fuels.

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- Thorium is three times more abundant than uranium and is composed primarily of fertile Th-232. Consequently, there is great potential for enhancing the sustainability of the nuclear fuel cycle by extending the availability of current resources through the use of thorium fuel cycles.
- Superior performance of ThO_2 at high burnup (e.g. because of reduced fission gas release) suggests the possibility of longer core residence times (compared to uranium-based fuels) which could lead to potential economic gains.
- The formation of U-232 via (n,2n) reactions with Th-232, Pa-233 and U-233 (and subsequent decay into strong gamma emitting daughter products) in Th–based fuels leads to intrinsic proliferation-resistance in Th-based fuel cycles.

Interest in thorium-based fuel cycles dates back to the mid 1950's (see, e.g., [8], [9]), but this interest was not sustained, due to the discovery of additional uranium deposits and improved availability of uranium [7]. The present revitalized interest in thorium-based fuel cycles is the result of a renewed focus on safety, sustainability, economics and proliferation resistance, as discussed above. In this paper, thorium fuel cycle options are examined and compared with respect to initial Pu driver fuel requirements, U-233 recycling and exit burnup.

2. Modelling and Analysis Methods

A 54-element bundle design was used for these studies, as described in [10] and shown in Figures 1 and 2 below. The bundle has three concentric fuel rings, with 12, 18 and 24 elements composed of mixtures of thorium oxide, plutonium oxide and, in some cases, U-233 oxide. All fuel pins within a bundle have the same isotopic composition and all bundles within a channel also have the same isotopic composition. The bundle also has a large centre element filled with zirconia. Detailed bundle specifications can be found in [10]. Two fuel channel designs were examined in this report, a re-entrant type fuel channel (REC) and a high-efficiency type channel (HEC). These channel designs are described in [11] and shown in Figure 1 and Figure 2, respectively. For both channels, the outermost component is a zirconium alloy pressure tube. In conventional CANDU reactor designs, the pressure tube is separated from moderator by a CO_2-filled gap surrounded by a calandria tube. However, for both the REC and HEC used in this paper, the pressure tube is in direct contact with the moderator – i.e. the calandria tubes are omitted. In the case of the REC, the fuel bundle is surround by a solid liner tube, separated from the pressure tube by an annular coolant layer. Coolant flow is re-entrant; i.e. the outer coolant layer flows past the outside of the liner tube and then re-enters the channel inside the liner tube where it flows through the fuel bundles. In the HEC, a porous zirconia insulator is located directly inside the pressure tube and isolates it from the high temperatures in the coolant. This insulator is supported on its inner surface by a perforated liner tube. Coolant, which flows past the bundles, is allowed to pass through the liner tube support and the porous insulator.

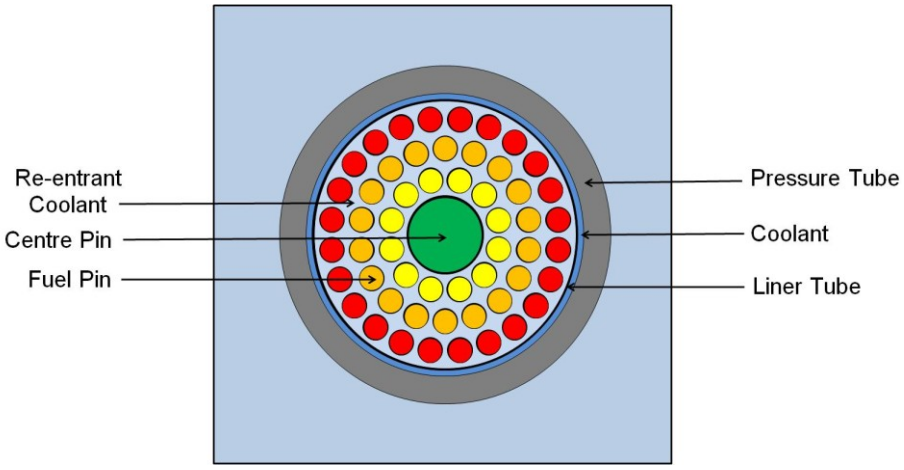

Figure 1 SCWR 54-Element Bundle Design and Re-entrant channel (REC) lattice cell

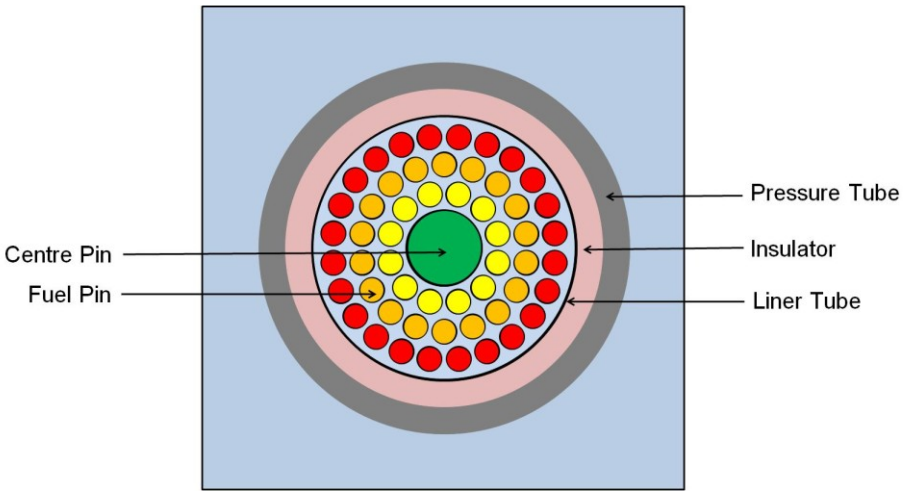

Figure 2 SCWR 54-Element Bundle Design and High Efficiency channel (HEC) lattice cell

Lattice physics calculations were performed using WIMS-AECL 3.1, which is a two-dimensional multi-group deterministic lattice physics code that solves the integral neutron transport equation using collision probabilities [12]. For this study, WIMS-AECL was used in conjunction with an 89-group nuclear data library based on ENDF/B-VII [13]. For the results presented here, WIMS-AECL was used to evaluate lattice reactivity as a function of fuel burnup. Fuel channel models in WIMS-AECL consisted of two-dimensional slices through the channels, perpendicular to the channel axis, as illustrated in Figure 1. Calculations were performed at five axial positions along the channel corresponding to axial distances (from the channel inlet) of 0.5 m, 1.5 m, 2.5 m, 3.5 m, and 4.5 m. The coolant densities and temperatures at these axial distances are taken from [14] and are also provided in **Table 1**. All other parameters are as specified in [10].

Table 1
Coolant density and temperature versus distance from inlet

Axial Distance (m)	Coolant Density (kg/m^3)	Coolant Temperature (ºC)
0.50	592.54	359.2
1.50	382.46	383.2
2.50	160.92	402.1
3.50	89.49	500.9
4.50	69.63	608.3

The impact of initial concentrations of recovered Pu and U-233 were assessed based on exit burnup, burnup-weighted average coolant void reactivity (CVR), and the ratio of final combined concentration of U-233 and Pa-233 to the initial U-233 concentration, denoted here as the "conversion ratio" or CR, given by,

$$CR = \frac{[\text{Pa-233}]_f + [\text{U-233}]_f}{[\text{U-233}]_i} , \quad (2)$$

where the subscripts, f and i refer to the final and initial concentrations of isotopes. Note that when removed from the core, the Pa-233 will decay to U-233 (with a 27 day half life), and so the Pa-233 is counted as U-233 for the CR calculation.

It was assumed that the exit burnup was reached when the channel averaged integrated k-infinity (averaged over five channel positions) reached a value of 1.040, where core leakage and absorption were assumed to have a worth of approximately 40 mk (40 mk corresponds to dk/k = 0.040, 4% or 4000 pcm). A more detailed description of the evaluation of exit burnup is provided in [15]. Values for the CVR and the CR were similarly averaged over all five axial positions.

Note that the values for exit burnup calculated by WIMS-AECL are for continuous refuelling. The exit burnup, B_n, for a cycle based on n batches is related to the exit burnup for a continuous-refuelling-based cycle, B_d, by [16],

$$B_n = \frac{n}{n+1} B_d . \quad (2)$$

The fuelling cycle currently under consideration for the CANDU-SCWR is based on three batches. Consequently, the exit burnup evaluated by WIMS-AECL for continuous refuelling must be corrected by a factor of 0.75. The exit burnups presented in this report correspond to 3-batch refuelling.

Two categories of fuel cycle were examined here. In the first, the once-through-thorium (OTT) fuel cycle, fuel was composed of a mixture of Pu driver fuel, obtained from recycled LWR fuel, and Th-232. The isotopic composition of the Pu driver fuel is the same as that used in [1] and is provided in **Table 2**. The second fuel cycle examined was based on U-233 recycling (UR). For this cycle, fuel was composed of a mixture of the same Pu driver fuel as in the OTT cycle, Th-232 and U-233. For both cycles, there are three objectives: obtaining a batch refuelling exit burnup of approximately 45 MWd/kg, obtaining a small, negative value for CVR (approximately -5 mk), and minimizing the [Pu] required as driver fuel. The lattice pitch (LP) and concentration

of Pu (for the OTT) or Pu and U-233 (for the recycle) were varied in order to obtain these objectives. For the UR cycle, the CR was also optimized to near unity by varying LP and the concentrations of Pu and U-233. Because of the similarities between the REC and HEC, scoping studies were performed only for the REC. The optimized lattice pitch obtained for the REC case was then corrected to approximately the same moderator-to-fuel ratio for the HEC case.

Table 2
Isotopic composition of Pu driver fuel derived from recycled LWR fuel

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

3. Results for Once Through Thorium Cycles

3.1 [Pu] and Lattice Pitch Optimization

Values for exit burnup and CVR were examined, using the REC channel, as a function of lattice pitch and Pu enrichment for once through thorium cycles (OTT). Cases were examined with lattice pitches ranging from 22 cm to 27 cm and [Pu] ranging from 10% to 14% by weight. Channel averaged results are shown in Figure 3 and are based on batch refuelling (i.e. WIMS-AECL exit burnup results are corrected by a factor of 0.75 based on a 3 batch cycle). Examination of Figure 3 shows that exit burnup increases with both lattice pitch and [Pu]. Moreover, the CVR becomes more positive with increases in both LP and [Pu]. These trends present a challenge since it is desirable to increase exit burnup while minimizing the CVR.

As can be seen from the results in Figure 3, changes to the LP have a larger influence on CVR (approximately +10 mk change in CVR for +1 cm change in LP) than [Pu] (approximately +2 mk change in CVR for +1% change in [Pu]), while changes to [Pu] have a much more profound influence on exit burnup (approximately +10 MWd/kg for +1% [Pu]) than changes to LP (approximately +3 MWd/kg for +1 cm change in LP). These results suggest that the best way to maximize exit burnup while minimizing CVR is to decrease the LP while increasing [Pu]. The combination of an LP of 24 cm and [Pu] of 13% leads to near optimal values for exit burnup and CVR, 43 MWd/kg and -4.5 mk, respectively. For the HEC channel, adjustment to approximately the same fuel-to-moderator ratio requires an increase in LP to 25 cm. WIMS calculations for the HEC channel with an LP of 25 cm and [Pu] of 13 weight % give a CVR of -6.9 mk, but an exit burnup of only 40 MWd/kg. Increasing [Pu] to 14 weight % gives near optimal values for exit burnup and CVR, 45 MWd/kg and -5.9 mk, respectively.

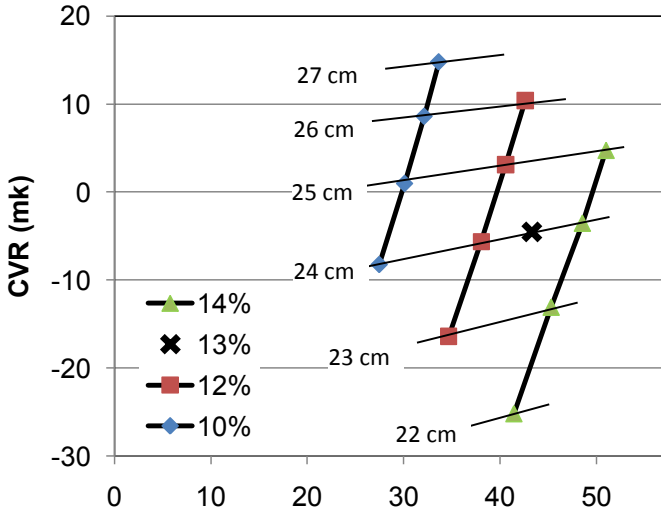

Figure 3 CVR versus Exit Burnup, Lattice Pitch, and initial [Pu] for REC

Since there is a significant variation in the coolant temperature and density along the channel axis, there is the possibility that there could be concomitant variations in both burnup and CVR. The axial variation in exit burnup and CVR are shown for the near optimal parameters for the REC and HEC channels in Figure 4 and Figure 5, respectively. In both cases, the exit burnup is essentially constant as a function of axial position. However, this behaviour is an artefact arising from the simplified model of the core in which equal power levels are applied at all axial locations along the channel. Factors such as core leakage will lead to axial variation in power profiles, which in turn will lead to axial variations in burnup. Examination of CVR versus axial position shows that there are axial variations (approximately ± 4 mk) in CVR for both the REC and HEC. A non-uniform axial power profile would also likely influence the observed axial variation in CVR, and this will be investigated in subsequent studies.

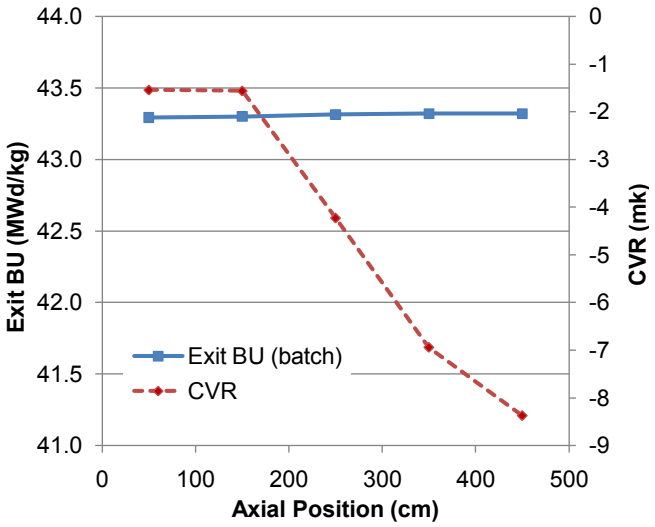

Figure 4 Exit Burnup and CVR versus Axial Position (REC)

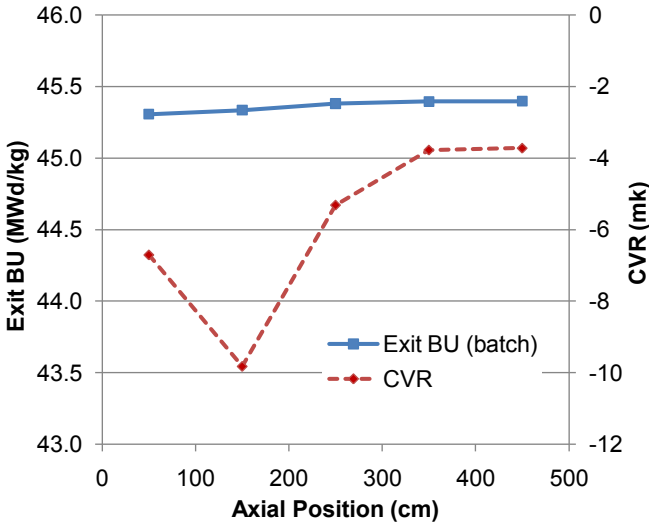

Figure 5 Exit Burnup and CVR versus Axial Position (HEC)

3.2 Conversion of Th-232 to U-233

As well as providing a sustainable alternative to natural uranium (NU) or U-235 enriched uranium-based fuel cycles, the OTT cycle also produces U-233 which is required to start the UR cycle. It is therefore important to evaluate U-233 production during the OTT cycle. The channel average exit [Pa-233 + U-233] for the REC and HEC are given in **Table 3** below.

The exit [U-233 + Pa-233] is shown for the REC and HEC as a function of axial position in Figure 6. The exit [U-233 + Pa-233] increases from inlet to outlet by approximately 10% for the REC and 20% for HEC. These results suggest that it may be possible to further optimize for U-233 conversion via axial variation in the initial [Pu].

It is also of interest to examine the fissions from the Pu driver fuel versus those from Th-232-derived isotopes. Although the latter fissions will come primarily from U-233, additional fissions will also occur in Th-232 and U-235, which is also produced from transmutation products of Th-232. At the beginning of the cycle, reactivity will clearly be supported purely from the Pu driver fuel. As the cycle proceeds and Pu is expended, the Th-232-derived isotopes will be responsible for an increasing fraction of the total fissions, and consequently, the power produced. The percent fissions from Pu driver fuel and the Th-232-derived isotopes are shown for the REC in Figure 7. Results for the HEC (not shown) are very similar to those of the REC. REC and HEC calculations show that by the end of the OTT cycle, approximately 30% of fissions taking place occur in the Th-232-derived isotopes. The burnup-weighted average percent fissions in Th-232-derived isotopes for the REC and HEC are provided in **Table 3**.

Table 3
Channel average exit burnup, CVR, exit isotope concentrations and burnup-weighted average % fissions from Th-232-derived isotopes

	wt % Pu	Exit BU (MWd/kg)	CVR (mk)	Exit [Pa-233+U-233]	% Fissions from Th-232, U-233, and U-235
REC OTT	13%	45	-5.9	1.4%	14%
HEC OTT	14%	43	-4.5	1.4%	13%

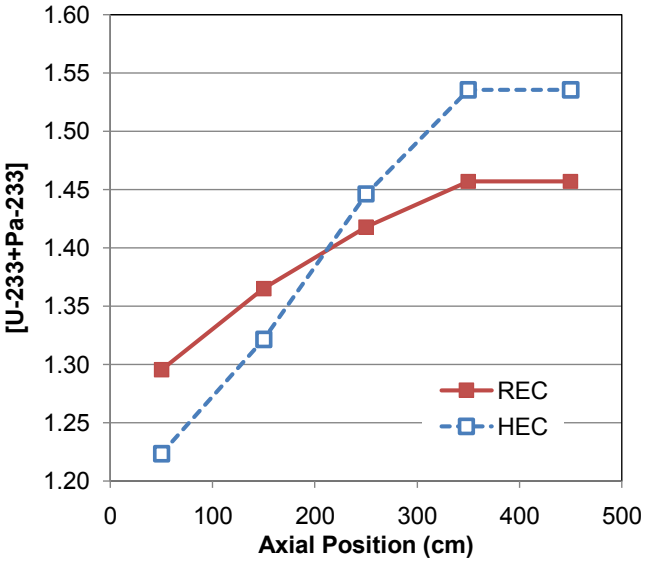

Figure 6 Final [U-233+Pa-233] versus axial position

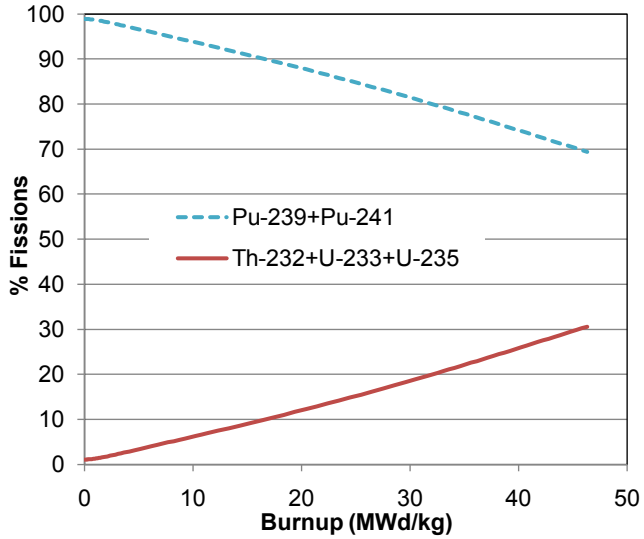

Figure 7 Percent Fissions from Pu-derived and ^{232}Th-derived isotopes (REC)

4. Results for U-233 Recycling Based Cycles

4.1 [U-233], [Pu], and Lattice Pitch Optimization

For UR cycle studies, the fuel was composed of a mixture of Pu driver fuel, U-233 and Th-232, and the exit burnup, CVR and CR were examined over a range of LP, [Pu] and [U-233] using the REC. Cases were examined with lattice pitches ranging from 23 cm to 25 cm, [Pu] ranging from 7% to 9% by weight and [U-233] ranging between 0.5% and 3.0% by weight. As was observed in the OTT cycle, exit burnup increases with both lattice pitch and [Pu] or [U-233]. Likewise, CVR becomes more positive as both lattice pitch and [Pu] or [U-233] increase. The selected LP, [Pu] and [U-233] for the REC and corresponding exit burnup and CVR are summarized in **Table 4**, below. Corresponding values for the HEC are also shown. Assuming that the reactors and fuel bundle design used for the OTT cycle are the same as for the UR cycle, approximately 1.5 OTT reactors are required to produce enough U-233 to begin the UR cycle in one reactor. This result holds for both the REC and HEC designs.

Table 4
Channel average exit burnup, CVR, exit isotope concentrations and burnup-weighted average % fissions from Th-232-derived isotopes

	wt % U-233	wt % Pu	Exit BU (MWd/kg)	CVR (mk)	CR	Exit [Pa-233+U-233]	% Fissions from Th-232, U-233, and U-235
REC recycle	2.12%	8%	45	-5.9	1.01	2.1%	37%
HEC recycle	2.12%	8%	43	-4.5	1.01	2.2%	37%

Note that the values for CR for the chosen input parameters are only slightly greater than 1. Because of potential reprocessing losses, it may be required to increase the CR. This can be achieved by decreasing the initial U-233 concentration, as shown in Figure 8. This would result in a concomitant decrease in exit burnup, unless the initial [Pu] were increased to compensate, as shown in Figure 9.

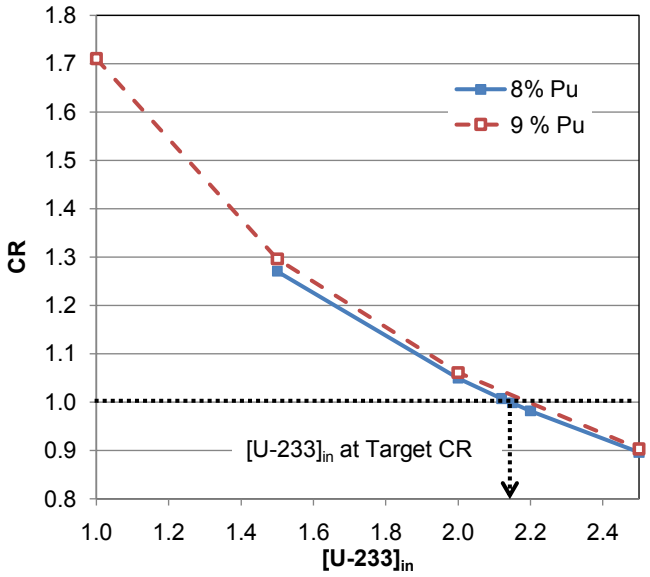

Figure 8 CR versus input [U-233] for 24 cm LP

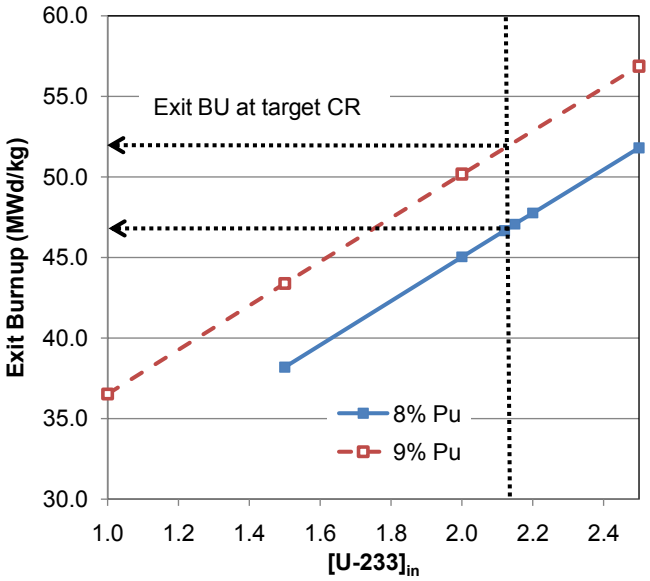

Figure 9 Exit burnup (MWd/kg) versus input [U-233] for 24 cm LP

The axial variation in exit burnup and CVR are shown for the selected parameters for the REC and HEC channels in Figure 10 and Figure 11, respectively. In both cases, the results are similar to those found for the OTT cases; the exit burnup is essentially constant (as a result of the same

modelling artefact as described in Section 3.1) as a function of axial position, but there are variations (approximately ± 4 mk) in CVR.

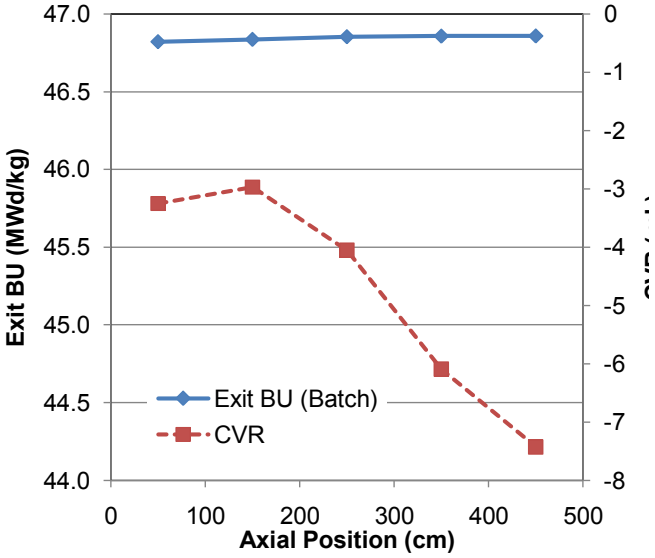

Figure 10 Exit Burnup and CVR versus Axial Position (REC)

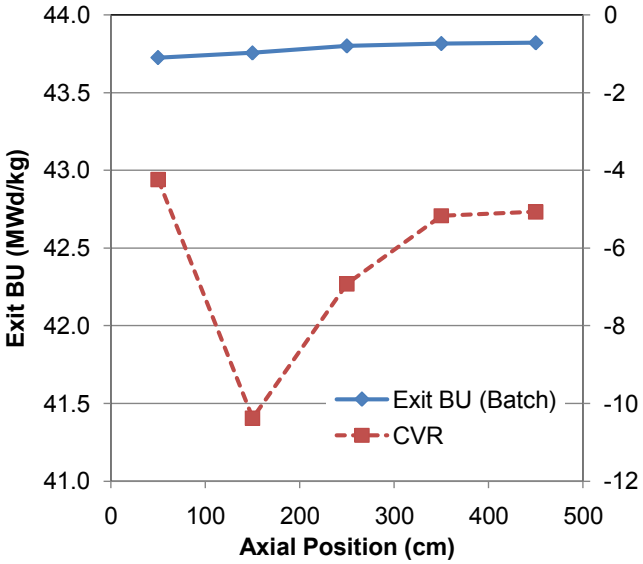

Figure 11 Exit Burnup and CVR versus Axial Position (HEC)

4.2 Conversion of Th-232 to U-233

One of the objectives of the UR cycle is to be self sufficient in U-233, i.e. produce enough U-233 at exit burnup to begin the next fuel cycle in the same reactor with the same initial fuel

composition; that is, CR = 1. The channel average exit [Pa-233 + U-233] for the REC and HEC are given in **Table 4** above.

The conversion ratios for the REC and HEC are shown as a function of axial position in Figure 12. It is interesting to note that for both the REC and HEC, the conversion ratio is significant lower (by approximately 10%) at the channel inlet than the channel outlet. This suggests that some advantage (with respect to CR) may be gained through axial variation in the initial ratio of [U-233] to [Pu].

As was the case with the OTT cycle, it is also of interest to determine the fraction of fissions from the Pu driver fuel compared to those from Th-232-derived isotopes. The percent fissions from Pu driver fuel and the Th-232-derived isotopes are shown for the REC in Figure 13. Results for the HEC (not shown) are similar to those for the REC. In both cases approximately 25% of fissions taking place at the beginning of the cycle occur in the Th-232-derived isotopes. This value increases to approximately 50% by the end of the cycle. The burnup-weighted average percent fissions in Th-232-derived isotopes are approximately 37% in both the REC and HEC.

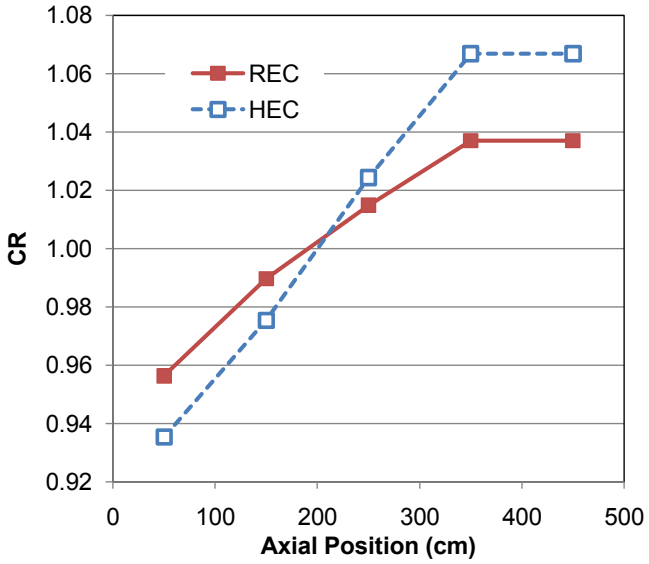

Figure 12 Conversion ratio versus axial position for the REC and HEC

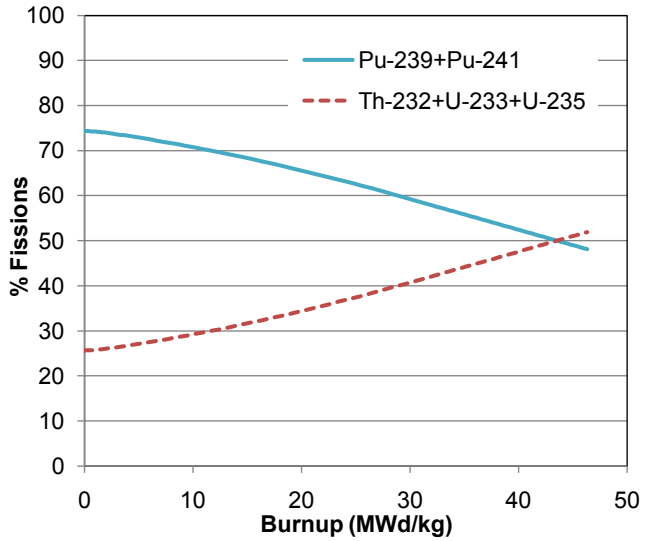

Figure 13 Percent Fissions from Pu-derived and Th-232-derived isotopes (REC)

5. Summary

In this paper, WIMS-AECL was used to examine two classes of homogenous CANDU-SCWR thorium-based fuel cycles, a once through thorium-based cycle, and a U-233-recycling-based cycle. Lattice pitch and fissile fuel enrichments were varied in order to obtain a target exit burnup of approximately 45 MWd/kg and negative CVR, while minimizing the requirement for Pu driver fuel. A single 54-element bundle design was examined, using two fuel channel designs, the REC and HEC.

Optimal values for LP and input isotope concentrations were obtained for the OTT and UR cycles, for both the REC and HEC. Although there are differences in the two channel designs, the optimal values obtained for the REC and HEC were similar for both fuel cycles. Axial variation in final [U-233] (for the OTT cycle), or CR (UR cycle) suggest that further improvements may be possible via axial variation in Pu content or Pu and U-233 content, respectively. Further studies will be performed to assess the potential use of axially and radially graded enrichment to further optimize exit burnup and CVR. In addition, the effects of core leakage and axially varying power profiles will be investigated.

6. References

[1] B. Hyland, et al., "Homogeneous Thorium Fuel Cycles in CANDU Reactors", Proceedings of the Global 2009 Conference, Paris, France, 2009 Sept. 6-11.

[2] P.G. Boczar, P.S.W. Chan, G.R. Dyck, R.J. Ellis, R.T. Jones, J.D. Sullivan and P. Taylor, "Thorium Fuel-Cycle Studies for CANDU Reactors", Thorium Fuel Utilization: Options and Trends, IAEA-TECDOC-1319, November 2002.

[3] P.G. Boczar, G.R. Dyck, P.S.W. Chan and D.B. Buss, "Recent Advances in Thorium Fuel Cycles for CANDU Reactors", Thorium Fuel Utilization: Options and Trends, IAEA-TECDOC-1319, November 2002.

[4] M. Ovanes, P. S. W Chan, and J. M Hopwood, "Thorium and Other Fuel Cycle Flexibility of ACR 1000", Proceedings of the Global 2009 Conference, Paris, France, 2009 Sept. 6-10.

[5] R. Duffey, L.K.H. Leung, and I. Pioro, "Design Principles and Features of Supercritical Water-cooled Reactors to Meet Design Goals of Generation-IV Nuclear Reactor Concepts", IAEA Technical Meeting on Heat Transfer, Thermal-Hydraulics and System Design for Supercritical Water Cooled Reactors, University of Pisa, Pisa, Italy, July 5-9, 2010.

[6] U.S. DOE Nuclear Energy Research Advisory Committee and the Generation IV International Forum, "A Technology Roadmap for Generation IV Nuclear Energy Systems", GIF-002-00, December 2002.

[7] Nuclear Fuel Cycle and Materials Section, IAEA, "Thorium fuel cycle - Potential benefits and challenges", IAEA-TECDOC-1450, May 2005.

[8] W.B. Lewis, "The Canadian Research Reactors and Their Uses", *British Journal of Applied Physics*, Vol. 7, Iss. S5, 1956, pp S96-S100.

[9] J.R. Dietrich, "The Physics of Advanced Reactors", *British Journal of Applied Physics*, Vol. 7, Iss. S5, 1956, pp S9-S23.

[10] P.G. Boczar, W. Shen, J. Pencer, B. Hyland, P.S.W. Chan and R.G. Dworshak, "Reactor Physics Studies for a Pressure Tube Supercritical Water Reactor (PT-SCWR)", The 2nd Canada-China Joint Workshop on Supercritical Water-Cooled Reactors (CCSC-2010), Toronto, Ontario, Canada, 2010 April 25-28.

[11] C.K. Chow and H. F. Khartabil, "Conceptual Fuel Channel Designs for CANDU-SCWR", *Nuclear Engineering and Technology*, Vol. 40, Iss. 2, 2007, pp 139-146.

[12] D.V. Altiparmakov, "New Capabilities of the Lattice Code WIMS-AECL", PHYSOR 2008, PHYSOR 2008 - International Conference on the Physics of Reactors, Nuclear Power: A Sustainable Resource, Interlaken, Switzerland, 2008 September 14 –19.

[13] D. Altiparmakov, "ENDF/B-VII.0 Versus ENDF/B-VI.8 in CANDU Calculations", PHYSOR 2010 – Advances in Reactor Physics to Power the Nuclear Renaissance, Pittsburgh, Pennsylvania, USA, May 9-14, 2010.

[14] R.B. Duffey and I.Pioro, "Advanced High Temperature Concepts for Pressure-Tube Reactors, Including Co-Generation and Sustainability", Proceedings HTR2006: 3rd International Topical Meeting on High Temperature Reactor Technology, Johannesburg, South Africa, 2006 October 1-4.

[15] J. Pencer, D. Guzonas, G.W.R. Edwards, and B. Hyland, "Impact of Materials on CANDU-SCWR Lattice Physics", The 5th International Symposium on SCWR (ISSCWR-5), Vancouver, British Columbia, Canada, 2011 March 13-16.

[16] M.J. Driscoll, T.J. Downar, and E.E. Pilat, "The Linear Reactivity Model for Nuclear Fuel Management", American Nuclear Society, La Grange Park, Illinois, 1990.