ANALYSIS OF THORIUM/U-233 LATTICES AND CORES IN A BREEDER/BURNER HEAVY WATER REACTOR

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ABSTRACT – Due to the inevitable dwindling of uranium resources, advanced fuel cycles in the current generation of reactors stand to be of great benefit in the future. Heavy water moderated reactors have much potential to make use of thorium, a currently unexploited resource. Core fuelling configurations of a Heavy Water Reactor based on the self-sufficient thorium fuel cycle were simulated using the DRAGON and DONJON reactor physics codes. Three heterogeneously fuelled reactors and one homogeneously fuelled reactor were studied.

Introduction

Increasing population, urbanization, and energy demand, and the escalating role of nuclear power to meet that demand will significantly reduce the time period of relatively cheap and easily accessible uranium. Uranium is currently used worldwide at a rate of about 68,000 tonnes per year [1]. It is estimated that there are 5.3 million tonnes of easily accessible uranium ore and an additional 7.6 million tonnes that would be much more costly to extract. Further, there is an estimated 4 billion tonnes [1] of uranium in seawater, though no commercial process for its extraction currently exists. Thus it is advantageous to develop and implement advanced fuel cycles to extend and perhaps replace uranium resources.

Since the 1950s, thorium has been proposed as an alternative fertile nuclear fuel to complement or replace uranium [2]. This is owing to its relatively high abundance (three times more abundant than uranium [3]) and other advantageous physical and chemical properties. While thorium[1] is not fissile itself, its absorption of a neutron results in the production[2] of ^{233}Pa, which decays to fissile ^{233}U with a half-life of approximately 27 days. ^{233}U has a low capture-to-fission ratio (α) and thus a high reproduction factor (η) in a thermal neutron energy spectrum. Because thorium and ^{233}U are lower in atomic mass than the heavy element isotopes in conventional uranium-based fuels, there is reduced production of heavier minor actinides (such as plutonium, americium and curium) by successive stages of neutron capture and decay, compared with ^{238}U-based fuels.. This reduces the amount of long-lived minor actinides produced and the radiotoxicity of spent fuel. The absorption rate, and thus the thermal utilization factor (f) of thorium, are higher than ^{238}U [2]. When fabricated into fuel pellets, thorium dioxide (ThO_2) is more chemically stable, has higher thermal conductivity, lower thermal expansion, and higher melting point than UO_2 [2]. These characteristics lead to a lower fuel temperature, and better fuel performance, including lower gaseous release of volatile fission products.

[1] Specifically, ^{232}Th, which is the only naturally abundant isotope of thorium.
[2] The absorption of a neutron by ^{232}Th results in its transmutation to ^{233}Th, which β decays with a half-life of approximately 22 min to ^{233}Pa.

The use of thorium fuel in heavy water reactors has been investigated since the 1960s [4]. The majority of that work has been focused on once-through thorium (OTT) cycles with homogeneous fuels where fertile thorium is initially mixed with fissile fuel (topped) using plutonium and/or enriched uranium. This study will instead perform simulations of self-sufficient equilibrium thorium (SSET) cycles where thorium fuel contains an initial amount of ^{233}U which will be replenished during its time in the reactor. The goal is to simulate various fuelling configurations which result in a breakeven conversion ratio (*CR*~1), while maintaining criticality and reactor power. There is some overlap between this study and that of earlier studies of HWRs with homogeneous cores designed to achieve an SSET cycle with U-233/thorium fuels [5] [6].

1. Lattice physics analysis

Lattice physics calculations were performed using DRAGON [7], a lattice cell calculation code that solves the neutron transport equation [8]. DRAGON is developed and maintained by the Groupe D'Analyse Nucléaire at École Polytechnique de Montréal. This code was used to calculate macroscopic cross sections, isotopic densities and infinite cell multiplication constants over a series of constant-power burnup increments. A DRAGON model of the 37-element bundle using the IAEA WIMS-D4 nuclear data library was used for all calculations in this study.

1.1. Specific power

Specific power is an integral parameter in lattice calculations. Though the mass of heavy elements (HE) in a bundle can be easily calculated, specific power cannot be found without knowing the average bundle power of the cell being considered. The reference specific power of a 37-element bundle fuelled with natural uranium is approximately 32 W/g [7] This value is based on a flux-squared average of bundle powers. Instead, (as a first approximation) the numeric mean bundle power will be estimated based on the known reactor power of a CANDU 6 (2061.4 MW divided by 380 channels and 12 bundles is approximately 452 kW per bundle). During full core calculations, the core can be subdivided to obtain more accurate bundle powers for each region. The value can then be revised through an iterative process. For a 37-element bundle fuelled with thorium and 1.4 at% ^{233}U, the heavy element mass[3] is ~18.15 kg. This results in a specific power of ~24.91 W/g. To test the sensitivity of lattice calculations to specific power, a simulation of the aforementioned thorium bundle was irradiated for 1000 days using several different values of specific power. The results of these are shown in Figure 1 and Figure 2. It can be seen that changes on specific power do not significantly affect results at lower burnup values. However at higher burnups, an increase in specific power results in a significantly lower multiplication constant and a higher fissile nuclide concentration[4]. Although this seems initially counter-intuitive (as ^{233}U concentration decreases with higher specific power), it was found that the rise in FNC (fissile nuclide concentration) was due to a significant increase in ^{233}Pa concentration (which does not immediately contribute to criticality.)

[3] HE mass = volume of 37 elements × HE density of fuel = $[37\pi r^2 h] \times [(0.014\rho_{UO2} + 0.986\rho_{ThO2}) \times$ fuel's HE wt%]
mass = $[37\pi(0.6\text{ cm})^2(0.493\text{ cm})] \times [(0.014(10.6\text{ g/cm}^3) + 0.986(10\text{ g/cm}^3)) \times 0.879] = 18.15$ kg

[4] It must be noted that in this case the fissile nuclide concentration refers to the numerical concentrations of ^{233}U, ^{233}Pa and ^{235}U atoms in the fuel. As previously mentioned, ^{233}Pa decays to ^{233}U with a half life of about 27 days. ^{235}U is produced in small quantities (which become more significant at higher burnups) by neutron capture on ^{233}Pa and ^{233}U.

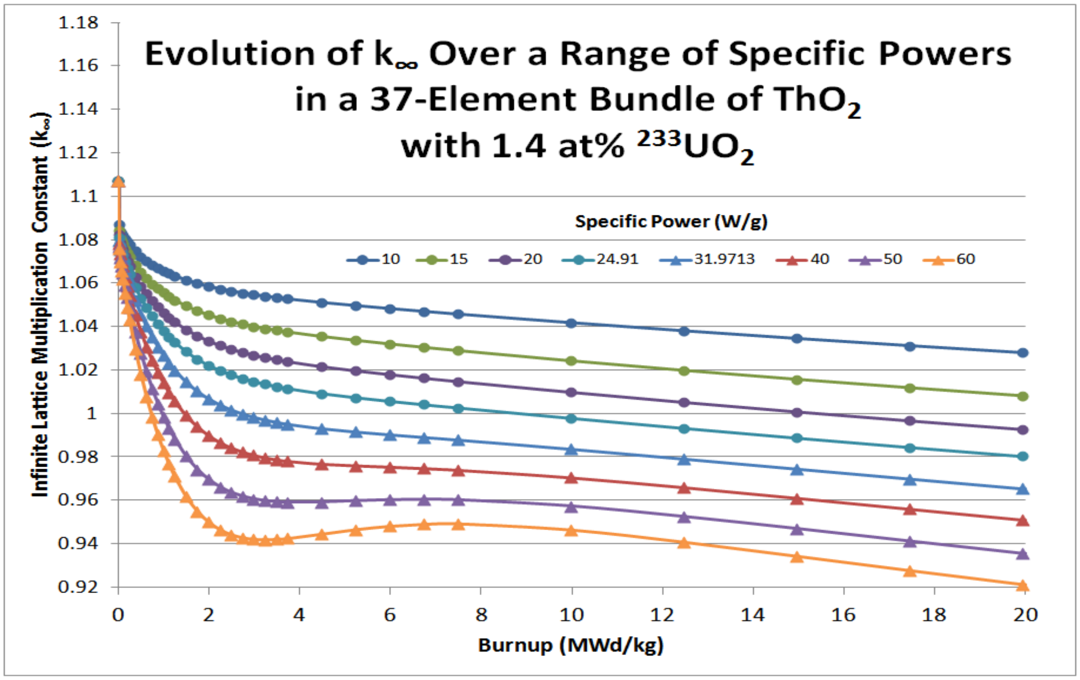

Figure 1 – Effect of varying specific power on the evolution of the infinite lattice multiplication constant

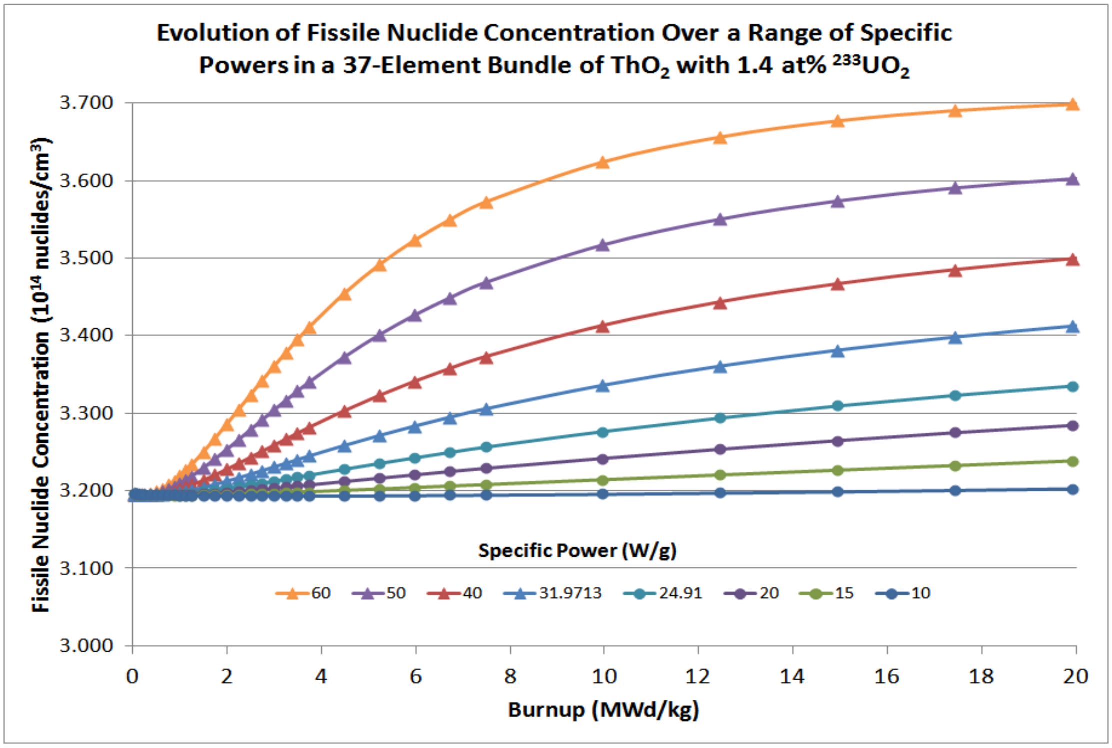

Figure 2 – Effect of varying specific power on the evolution of the fissile nuclide concentration

1.2. Initial fissile nuclide concentration

In order to find the optimal initial composition of the fuel, a second study was performed, holding the specific power constant at 24.91 W/g while varying the initial FNC. It has been previously stated that around 1.5 at% ^{233}U is the approximate concentration that results in a breakeven cycle [5] [9]. Therefore, several concentrations between 1.3 at% and 1.6 at% were calculated using DRAGON and the results are shown in Figure 3 and Figure 4.

At high values of burnup, the concentration of fissile nuclides converges to a nearly asymptotic value, at slightly less than 1.5 at% (Figure 3). As the fuels approach this asymptotic composition, their infinite lattice multiplication constant (k_∞) also converges (Figure 4). The convergenceto a relatively high FNC but a low k_∞ is due to the accumulation of fission products.

The difficulty of achieving a self sufficient fuel cycle can be observed from these graphs. The 1.6 at%, 1.55 at% and 1.5 at% fuels, while capable of reaching higher burnups, are not sustainable, as the fissile nuclide concentration depletes. Reactors fuelled with these concentrations would need an ongoing, outside source of ^{233}U. The 1.3 at%, 1.35 at% and 1.4 at% achieve net production of fissile nuclides almost immediately. However, they become subcritical[5] after a very short burnup ($\lesssim$ 2 MWd/kg). The 1.45 at% fuel can reach self sufficiency at approximately 6 MWd/kg , but it cannot obtain higher burnups and maintain criticality.

It therefore seems that homogeneous fuelling of the reactor is not viable or sustainable long term, as high FNC fuels do not breed sufficiently, while low FNC fuels cannot stay in the reactor long enough due to excess negative reactivity. Low or medium FNC fuels may be able to achieve a critical self-sustaining reactor, but would require very low discharge burnups and high refuelling rates. Instead, heterogeneously fuelled reactors containing multiple initial ^{233}U concentrations shall be explored. This work builds on past experience with thorium-fuelled light water breeder reactors (LWBR) which used heterogeneous cores [4] [5] [10].

[5] $k_{\text{eff}} < 1.00$ or $k_\infty < 1.03$

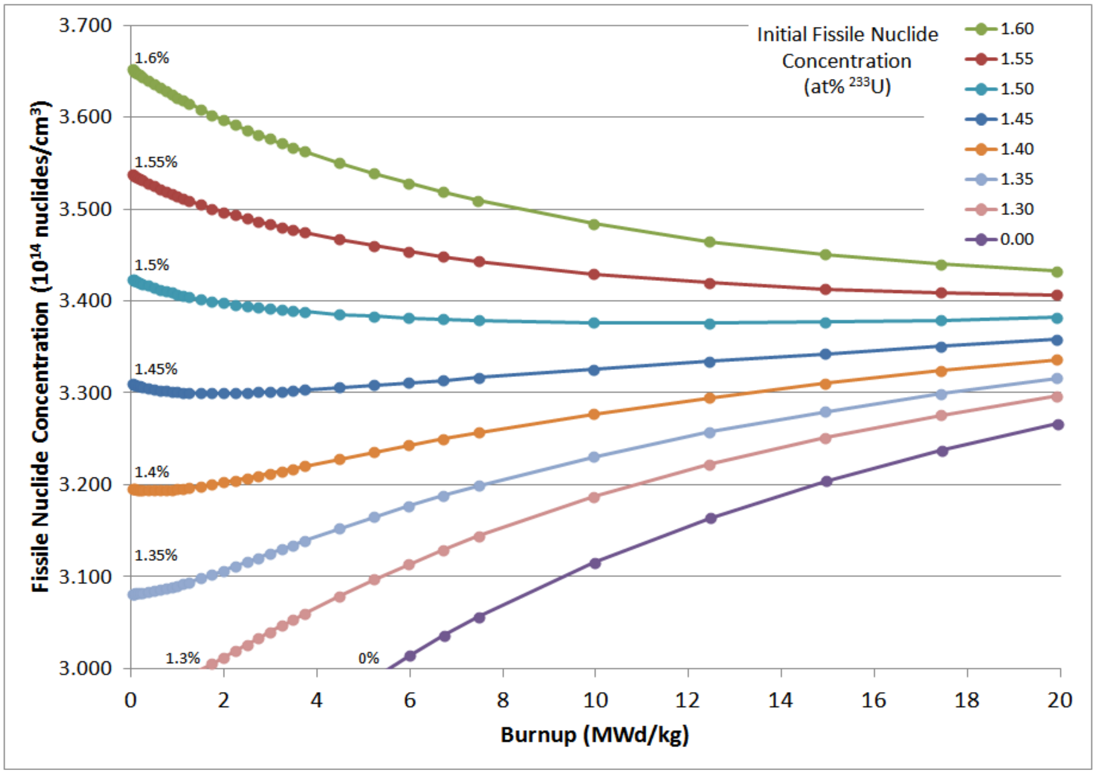

Figure 3 – Evolution of Fissile Nuclide Concentration with Burnup

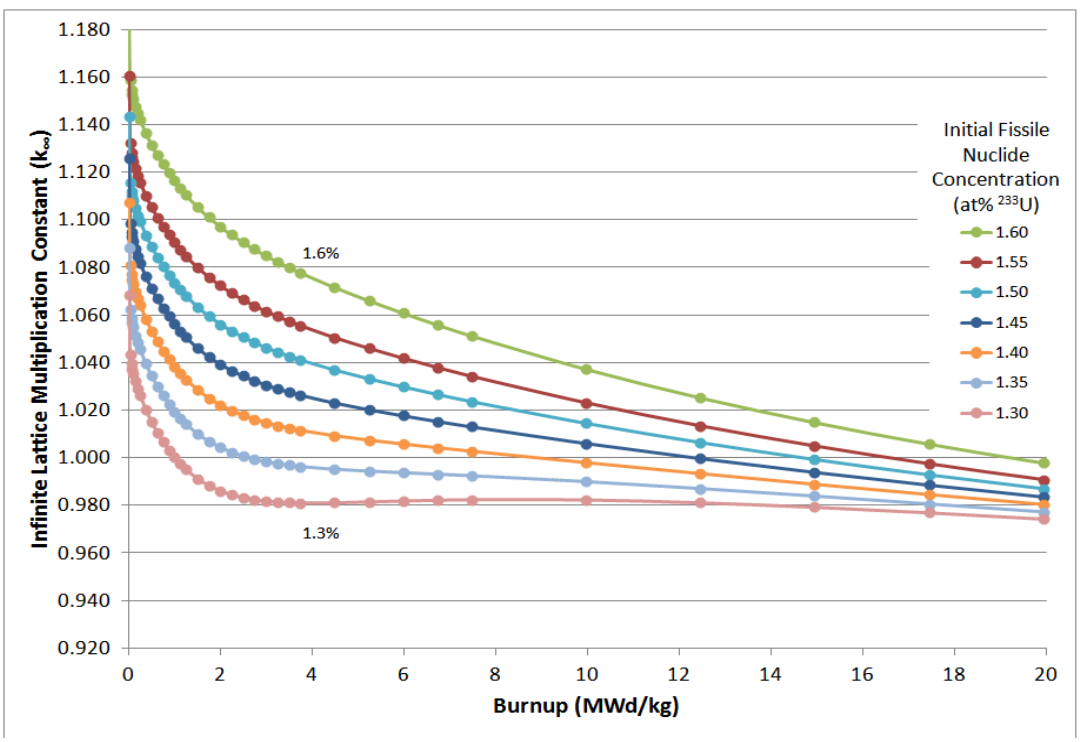

Figure 4 – Evolution of Infinite Lattice Multiplication Constant with Burnup

2. Full core analysis

DONJON [11] is a finite reactor analysis code (also developed and maintained by the Groupe D'Analyse Nucléaire at École Polytechnique de Montréal) that solves the neutron diffusion equation [8]. A DONJON model of the CANDU 6 core along with the lattice cell cross sections calculated in DRAGON were used to calculate the overall multiplication constant of the reactor, the neutron flux shape and the bundle and channel powers.

2.1 Reactivity control devices

The full-core model contained all devices and components for reactor control and flux detection. It is desirable, if possible, to eliminate the need for parasitic losses in order to obtain the best achievable neutron economy. Therefore, the adjuster rods were initially fully withdrawn from the reactor, and were inserted only as required for power profile flattening or negative reactivity. The liquid zone controllers were kept initially empty since they are used for fine tuning and control. If a reactor is supercritical with the liquid zone controls at 100%, then the ability to control the reactor is diminished.

2.2 Criteria

A number of parameters were used to judge the viability of each simulation. The reactor power was maintained at 2061.4 MW_{th}. The target effective multiplication constant was 1.002-1.003. This 2-3 mk cushion was meant to account for errors in calculations. Bundle and channel powers were kept below their respective license limits (935 kW and 7.3 MW, respectively [12]). Axial and radial flattening of the power profiles was attempted. The normal 8-bundle shift refueling scheme was used for all simulations. Lastly, a self-sufficient fuel cycle (with conversion ratio[6] $\geq$ 1.0) was the ultimate goal of the study.

2.3 Preliminary core configurations

Four core configurations were studied (see Figure 5). Initial FNCs of 1.4 at% and 1.6 at% were used for the blanket and seed fuels, respectively.

The first configuration uses an inner seed and outer blanket (ISOB) approach. The inner core of the reactor is fuelled with high fissile content "driver" or "seed" bundles and the peripheral ring of channels are fuelled with lower fissile content "breeding" or "blanket" bundles. The negative reactivity blanket fuel can be used due to the excess neutrons produced in the supercritical seed region. This blanket will also ideally capture a significant fraction of neutron leakage from the inner seed region.

The second configuration is the reverse of the first and uses an inner blanket and outer seed (IBOS) arrangement. Since higher power results in more fissile nuclide production (as shown in Section 1.1), the blanket fuel is placed in the high power inner core. Similarly, lower power

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[6] The conversion ratio is defined as the production rate of fissile nuclides divided by the consumption of fissile nuclides. A reactor with CR > 1.0 produces more fissile nuclides than it consumes and it is said to be breeding.

results in fewer absorptions on ^{233}Pa (and thus fewer parasitic captures), so seed fuel is placed in lower power channels. This arrangement may also have a flattening effect on the power profile.

The third configuration arranges breeding and burning channels in a checkerboard seed and blanket (XSB) pattern. The high reactivity of a fresh seed fuel will counteract the low reactivity of an almost-discharged blanket bundle. The opposite effect (but slightly lower in magnitude) may also occur (depending on the initial FNC of blanket fuel) on the other side of the reactor (where the slightly high reactivity of fresh blanket fuel will counteract the slightly low reactivity of almost spent seed fuel.) This arrangement may also help to alleviate the initial spike in reactivity caused by refueling.

The final configuration studied is homogeneously fuelled (HF) with a self sufficient initial ^{233}U content of 1.45 at% and expected to use a relatively low discharge burnup.

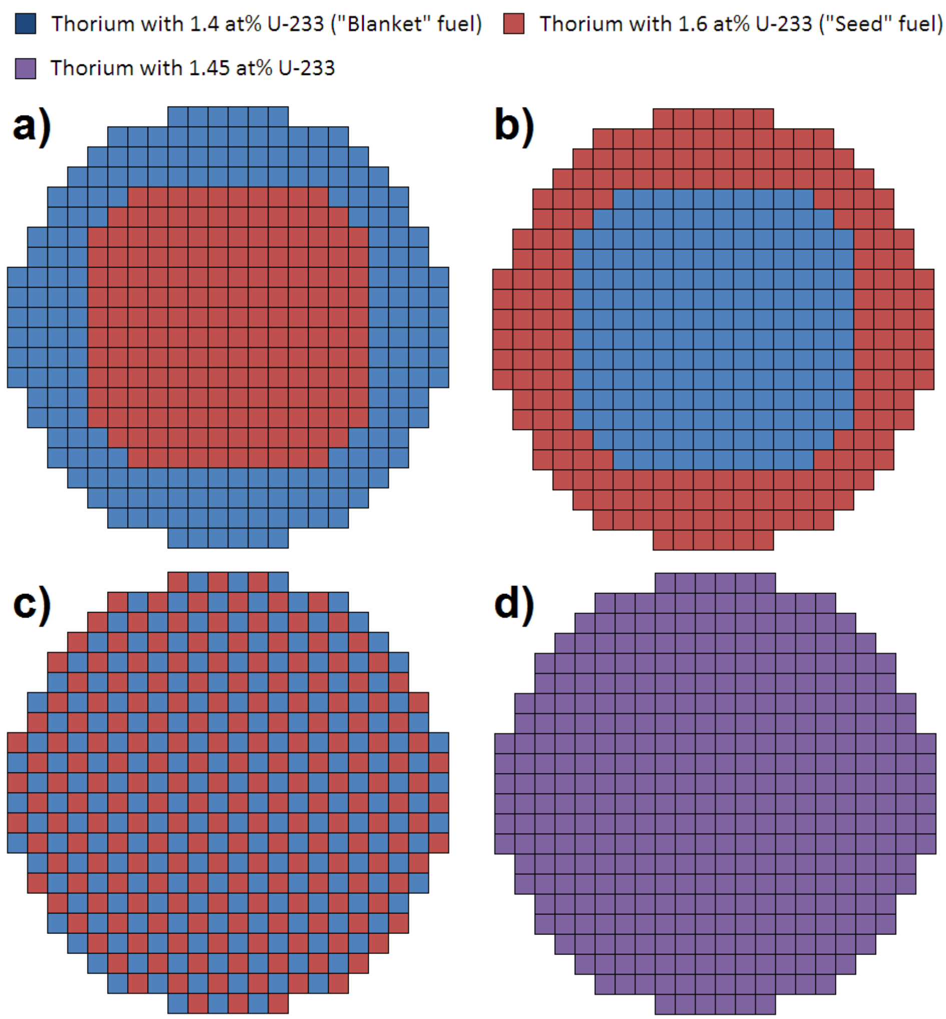

Figure 5 – Four general core configurations studied: a) Inner Seed/Outer Blanket (ISOB) b) Inner Blanket/Outer Seed (IBOS) c) Checkerboard Seed/Blanket (XSB) d) Homogeneously Fuelled (HF)

3. Results

The results of the simulations of each configuration are shown in Table 1. The values shown are the result of several permutations of parameters for each configuration that yielded results closest to the criteria specified in Section 2.2. The channel power distributions for each configuration are shown in Figure 6 and the channel power profiles for Row L are shown in Figure 7.

A single conversion ratio is difficult to calculate for heterogeneously fuelled reactors. Instead, the "Fissile Inventory Ratio" (*FIR*, see Equation 1) was calculated for each fuel region of the core.

(1)

The fuel-region FIRs were weighted by the mass refueling rate of their specific region to obtain a core-average FIR, given in Table 1.

The ISOB configuration was not particularly successful, owing mostly to power peaking in the seed channels. The channel and bundle powers at the center of the reactor could not be kept below the stated limits without significantly increasing the discharge burnup of those channels and inserting a majority of the adjusters, which significantly reduced the reactivity. To re-obtain criticality, the discharge burnup of the blanket channels had to be reduced drastically (to about 3 MWd/kg). As expected, the consequence was very high refueling rates (~2 channel visits per day in the blanket alone). Furthermore, this resulted in an FIR below 1.0, as the blanket bundles did not spend enough time in the reactor to breed sufficiently.

The IBOS configuration, however, showed very promising results. Fairly high discharge burnups were achieved for both seed and blanket fuel (20 MWd/kg and 12.39 MWd/kg, respectively). The FIR and k_{eff} were both above 1.0. The channel and bundle powers were maintained below the limits. Only the central adjusters were necessary and the radial form factor of Row L was the highest of the four configurations. The refueling rate of this configuration (0.96 channel visits per day) was considerably lower than the others and far below the standard natural uranium fuelled HWR (1.9 channel visits per day). This presents an additional benefit of decreasing the daily load of fuelling machines, which are expensive to purchase and maintain.

The checkerboard arrangement also yielded positive results. Criticality was easily achieved with fairly high discharge burnup values and maximum bundle and channel powers were quite low. Though very close, a FIR higher than 1.0 could not be reached, possibly due to the 1:1 ratio of seed and blanket channels.

As was speculated in Section 1.2, it was difficult to produce a homogeneously fuelled reactor that is simultaneously both breeding and critical. Although one which did both was eventually achieved, its average discharge burnup was quite low (~5.94 MWd/kg) and thus resulted in a high refueling rate (~2.52 channel visits per day).

Table 1 – Results of the Simulations for each Configuration Studied

Parameter	ISOB	IBOS	XSB	HF
Description	Inner Seed/Outer Blanket	Inner Blanket /Outer Seed	Checkerboard Seed/Blanket	Homogeneously Fuelled
Seed fuel initial ^{233}U content (at%)	1.60	1.60	1.60	1.45
Seed fuel average discharge burnup (MWd/kg)	27.57	20.00	17.42	5.94
Number of Seed Channels	184	196	190	380
Blanket fuel initial ^{233}U content (at%)	1.40	1.40	1.40	
Blanket fuel average discharge burnup (MWd/kg)	3.00	12.39	7.81	
Number of Blanket Channels	196	184	190	
k_{eff}	0.998876	1.002594	1.004308	1.002040
Fissile Inventory Ratio	0.99633	1.00498	0.99497	1.00106
Maximum Channel Power (MW)	7129.2	7158.5	6972.3	6902.0
└ Location	Channel L05	Channel L04	Channel Q12	Channel H11
Maximum Bundle Power (kW)	888.5	919.0	877.0	804.9
└ Location	Channel L05 Bundle #7	Channel L04 Bundle #7	Channel M04 Bundle #6	Channel E12 Bundle #7
Refueling Rate (channel visits/day)	2.29	0.96	1.32	2.52
Row L Radial Form Factor[7] (Average / Max)	0.89	0.91	0.86	0.88
Adjusters	Adjusters 2, 3, 4, 5, 6, 9, 10, 11, 12, 13, 16, 17, 18, 19, 20 inserted	Adjusters 4, 11, 18 inserted	Adjusters 2, 3, 5, 6, 9, 10, 12, 13, 16, 17, 19, 20 inserted	Adjusters 2, 3, 5, 6, 9, 10, 12, 13, 16, 17, 19, 20 inserted

[7] The form factor was calculated by dividing the average and maximum channel powers in row L

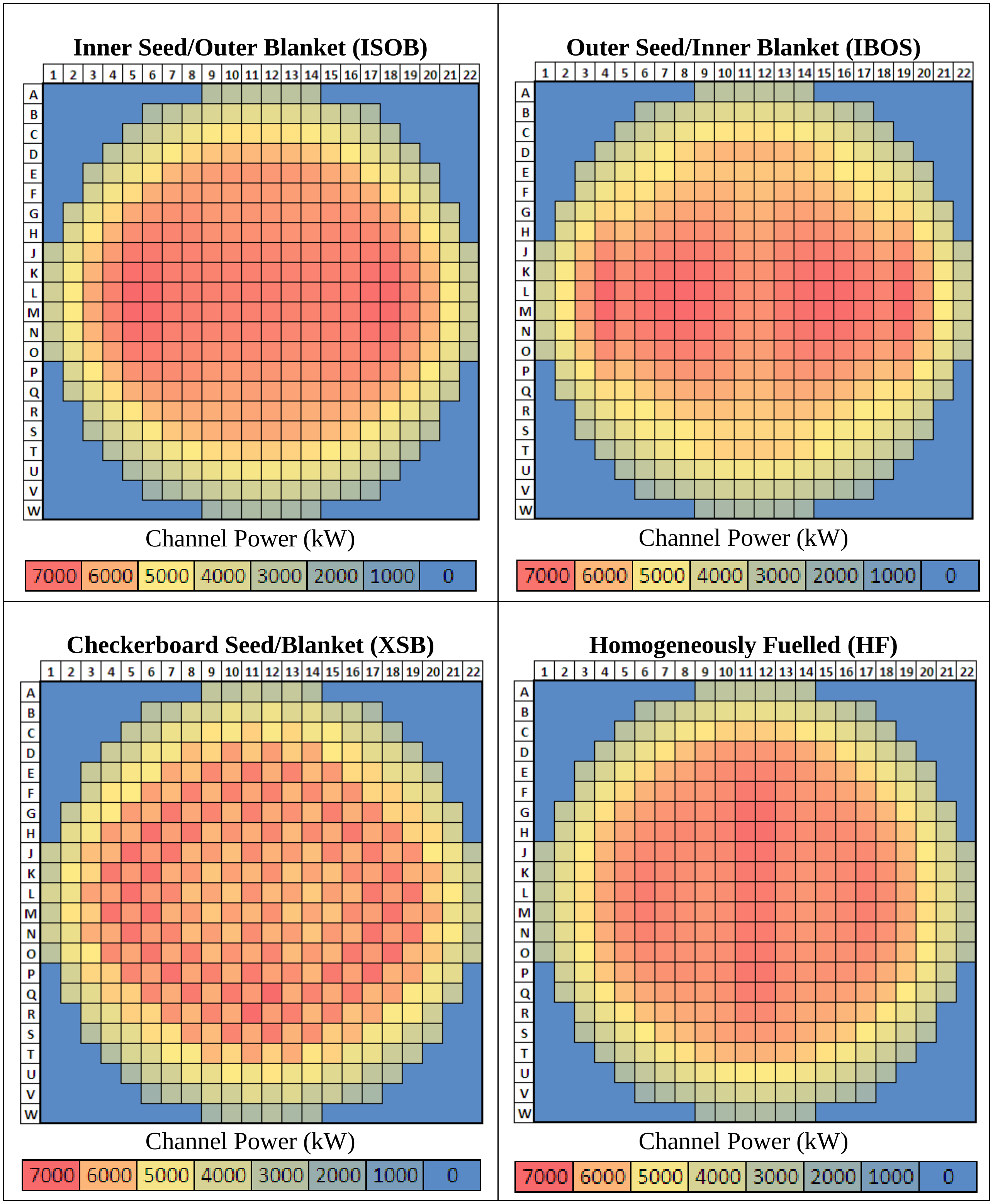

Figure 6 – Channel Power Distribution in the Studied Core Configurations

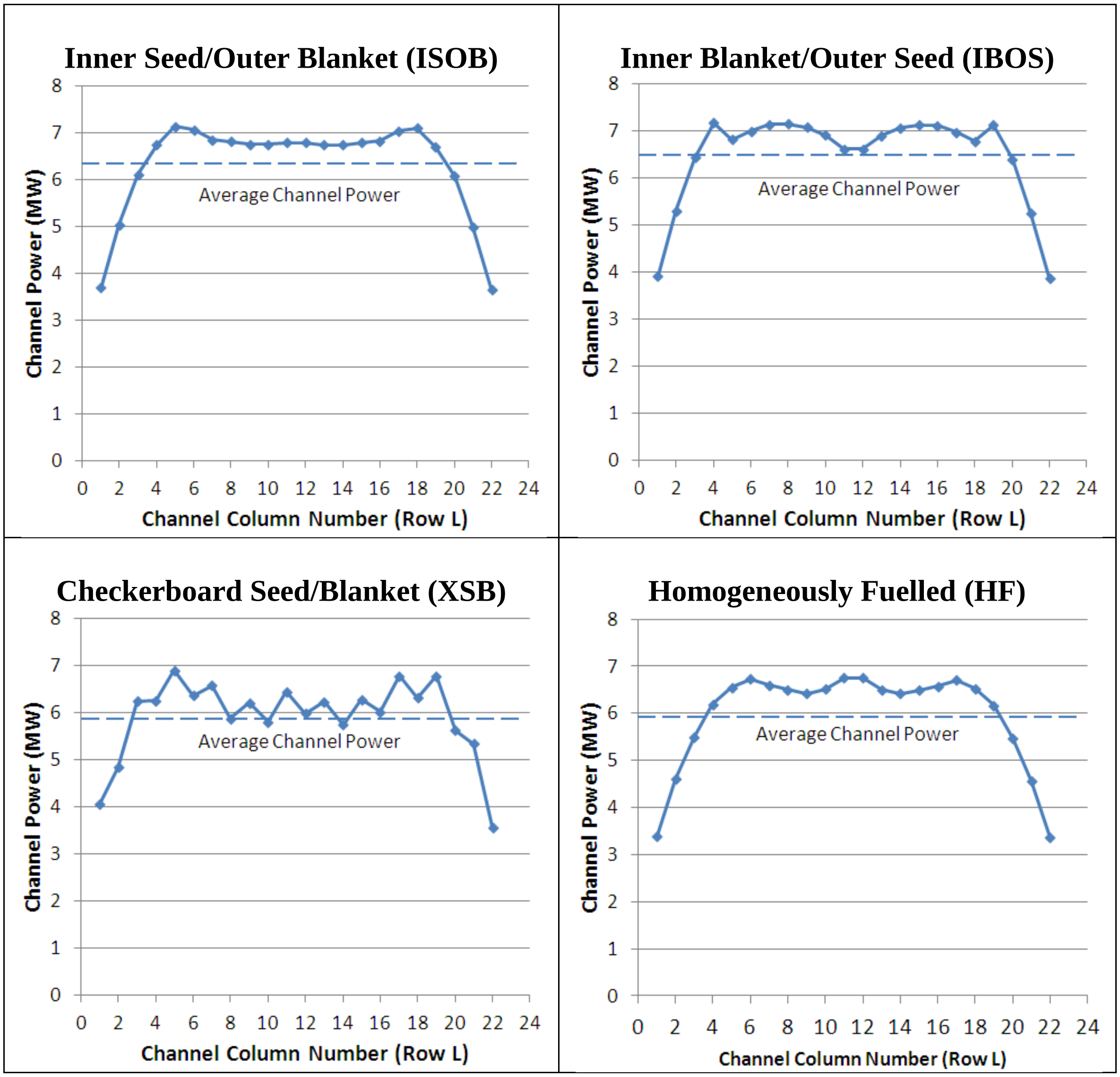

Figure 7 – Channel Power Profile in Row L of the Studied Core Configurations

4. Conclusions

Four configurations of breeder/burner HWR cores fuelled with mixed oxides of thorium and ^{233}U were simulated using the DONJON and DRAGON reactor physics codes. Three of these were heterogeneously fuelled cores with the seed fuel composed of thorium with 1.6 at% ^{233}U and the blanket of thorium with 1.4 at%. Of these, the inner seed/outer blanket arrangement could not simultaneously achieve criticality and a fissile inventory ratio greater than 1.0. The inner blanket/outer seed and checkerboard configurations performed well against the specified criteria, though the latter did not achieve net breeding. The fourth configuration was fuelled homogeneously with thorium containing 1.45 at% ^{233}U. While it could both reach criticality and self-sufficiency in ^{233}U, it required a far too rapid refueling rate, beyond the capability of current fuelling machines.

5. Future work

This study is part of a larger ongoing research effort. Further optimization of each configuration may yield better results. Additional work on safety margins, coolant void reactivity, refuelling ripple and delayed neutron effects is to be performed. Concepts such as enrichment of ^{90}Zr in the fuel sheath, pressure tubes and calandria tubes as well as advanced fuel bundle designs may help to overcome some of the challenges experieced above (particularly with the ISOB configuration.) The results of this report pertain only to a steady-state system (i.e. once a self-sustaining fuel cycle has been established.) Calculations for starting such a cycle (possibly with plutonium or enriched uranium) shall be carried out in the future. Although DONJON and DRAGON have been used extensively in industry and academia, the results of this particular project require verification. It is prudent to benchmark against other reactor physics codes.

6. References

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7. Acknowledgements

The authors would like to thank Ms. Bronwyn Hyland, Dr. Blair Bromley and Dr. Jeremy Pencer of AECL for their invaluable assistance and support. They would also like to thank Dr. Benjamin Rouben, Professor John Froats and Dr. Glen Harvel for their advice and mentorship. The present work also benefited greatly from and would not be possible without the work of the Groupe d'Analyse Nucléaire of L'École Polytechnique de Montréal.