Removing Fuelling Transient Using Neutron Absorbers

S. Paquette[1], P.K. Chan, H.W. Bonin and A. Pant[2]
Chemistry and Chemical Engineering Department
Royal Military College of Canada, PO Box 17000, Kingston, Ontario, Canada
[1]Stephane.Paquette@rmc.ca
[2]Cameco Fuel Manufacturing, Port Hope, Ontario, Canada, L1A 3A1

Summary

Preliminary criticality and burnup calculation results indicate that by employing a small amount of neutron absorber the fuelling transient, currently occurring in a CANDU 37-element fuel bundle, can be significantly reduced. A parametric study using the Los Alamos National Laboratories'MCNP 5 code and Atomic Energy of Canada Limited's WIMS-AECL 3.1 is presented in this paper.

1. Introduction

The reactor size, geometry and the distribution of fluence control the flux and power distribution across a reactor core. From a fuel management point of view, it is well known that fuel with high fluence has low reactivity. The opposite is also true. The neutron flux (and power density) is generally higher in regions where fuel has low irradiation. The on-site fuelling engineer and reactor physicist use these principles to shape the radial flux and power distributions throughout the reactor core. On-power refuelling is the primary means of maintaining a CANDU reactor in a critical condition [1]. Replacing irradiated fuel with fresh fuel in a specific channel has immediate consequences on the local power density distribution and reactivity. These power ripples (also known as refuelling ripples) could potentially bring channel power very close to compliance values and require immediate zone reactivity control system actions.

Figure 1 shows the infinite-lattice multiplication constant, k_∞, as a function of fuel burnup for a standard CANDU reactor fuelled with natural uranium (UO_2). Note that k_∞ is a measure of the multiplicative properties of the lattice in the absence of leakage from the fuel channel, also referred to as the "ideal situation". As shown, a typical fuel burnup attained in CANDU reactors using the 37-element fuel bundle is 7500 MWd $Tonne^{-1}$ of U [2]. However, the "actual" burnup attained depends on the operating parameters of the core and its reactivity (taking into account the "actual" neutron leakage).

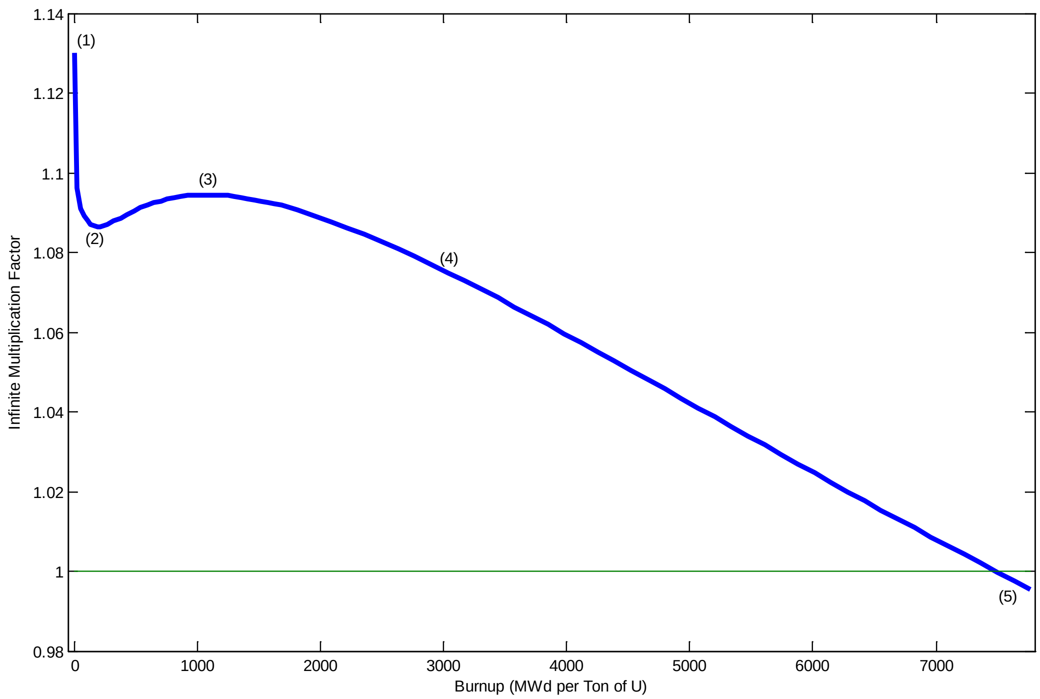

Figure 1: Infinite Multiplication Factor (k_∞) versus Burnup for a Typical CANDU Lattice

One cell in the previously discussed infinite lattice can be represented by a fuel channel. When such a channel is refuelled (1) with fresh fuel, its reactivity is high. This higher local reactivity tends to promote a power increase in the newly fuelled channel, as well as in its neighboring channels. The saturating fission products reaching saturation in the clean fuel (2) cause a large drop of reactivity and bundle power after a few days of operation. The production of plutonium from neutron absorption in U^{238} causes the reactivity (and power) to slowly increase again with irradiation over the next 40 to 50 Full Power Days (FPD). Following the plutonium peak (3), the plutonium production can no longer compensate for the depletion of U^{235} and the buildup of fission products. The excess reactivity and power density in the refuelled channel, as well as the power in its neighboring channels, slowly decrease (4). Eventually, the channel (or the cell) becomes sub-critical (5).

1.1 Objectives

A neutronic analysis on improving operating margins of a CANDU 37-element fuel bundle shown in Figure 2, is described in the following sections. Each of the 37 fuel rods consists of a thin zircaloy tube, the fuel sheath, filled with high density natural UO_2 fuel pellets. A 6 μm thick graphite-based coating, known as CANLUB, is located between the pellets and the sheath. This CANLUB graphite coating was developed to mitigate stress-corrosion cracking of the sheath during, or following, power ramps [3].

Figure 2: CANDU 37-Element Fuel Bundle [4]

In this parametric study, Gadolinium Oxide (Gd_2O_3), a burnable neutron absorber, is uniformly added in the CANLUB for the purpose of mitigating the fission products saturation and xenon free effects (from (1) to (2) in Figure 1) in fresh bundles loaded into the reactor core during refuelling. Gadolinium is commonly used for this application as its depletion rate matches closely the xenon build-up rate. It acts as a temporary high absorption cross section surrogate for fission product that would burn-out as fission products build-up. The main objective is to determine the amount of Gd_2O_3 required to suppress reactor core reactivity at low burnup.

2. Analysis Tools

The CANDU nuclear reactor has been modeled using the MCNP5 Monte Carlo code [5] and the Winfrith Improved Multigroup Scheme-Atomic Energy of Canada Limited (WIMS-AECL Version 3.1) lattice-depletion code [6].

MCNP 5 is a general-purpose, continuous-energy, generalized-geometry, time-dependent, coupled neutron/photon/electron Monte Carlo transport code with the capability to calculate k_{eff} eigenvalues for fissile systems. The code simulates individual neutrons in a 3D fissile environment based on transport data and records their average behaviours. It follows the nuclear particles from the source throughout their entire life. The outcome at each step of their life is determined based on probabilistic events (based on random probability generators). MCNP5 provides the effective multiplication factor of a system in the form of a statistical answer to the transport equation. A 3 Dimension Graphical User Interface continually provides the user with an updated view of the model. It also ensures that gaps in the geometry and overlapping of cells are avoided.

The computer code WIMS-AECL, Version 3.1, is a two-dimensional multi-group neutron transport code capable of applying leakage corrections and of performing fuel depletion routines for multi-cell lattices. The code is well suited for flux distribution calculations and determines the reactivity of a nuclear reactor by solving the integral form of the neutron transport equation. This is why WIMS-AECL is called a “deterministic” code, whereas MCNP5 is referred to as a probabilistic code since it uses a random probability generator [7].

The effects of finite reactor dimensions are taken into account by the leakage and buckling calculations of WIMS [8]. In order to obtain the effective multiplication factor, k_{eff}, the user must input a set of input geometric buckling based on the reactor core radius and height (radial and axial buckling values).

3. Natural Gadolinium

Gadolinium was selected because of its appropriate neutronic characteristics for thermal nuclear reactors such as CANDU. Natural gadolinium is comprised of seven naturally occurring isotopes. As demonstrated later in Section 5.1, only 2 isotopes, Gd-155 and Gd-157, are useful absorbers. The others have small thermal cross sections and are primarily responsible for undesirable absorber residuals into the CANLUB. Properties of natural gadolinium and the isotopic composition of gadolinium oxide (Gd_2O_3) are displayed in Tables 1 and 2, respectively.

Table 1: Properties of Natural Gd [9].

Isotope	Molar Mass (g mol^{-1})	Half-life (y)	Abundance (atom %)	Thermal σ_a (barns)
Gd-152	151.9197879	1.08×10^{14}	0.20	1056
Gd-154	153.9208623	Stable	2.18	84.99
Gd-155	154.9226188	Stable	14.80	60 889
Gd-156	155.9221196	Stable	20.47	2.188
Gd-157	156.9239567	Stable	15.65	254 078
Gd-158	157.9241005	Stable	24.84	2.496
Gd-160	159.9270506	Stable	21.86	0.7981
Natural Gd	157.2521		100	48 780

Notes: 1. Thermal refers to thermal neutrons at 2200 ms^{-1} or 0.025 eV; and
2. Density of Natural Gadolinium is 7.89 g cm^{-3}, based on periodic table.

Table 2: Isotopic Composition of Gd_2O_3.

Isotope	Mass Fraction (%)
Gd-152	0.17
Gd-154	1.85
Gd-155	12.65
Gd-156	17.61
Gd-157	13.55
Gd-158	21.64
Gd-160	19.29
O	13.24

Notes: 1. Molar Mass of Oxygen is 15.9949146 g mol^{-1};
2. Density of Gd_2O_3 is 7.41 g cm^{-3}, based on periodic table;
3. Molar Mass of Gd_2O_3 is 362.4890 g mol^{-1}.

4. Gd_2O_3 into CANLUB

In this study, the CANLUB coating is doped with various amounts of Gd_2O_3. The intent is to observe the effects of Gd_2O_3 on a typical CANDU nuclear reactor core reactivity and burnup over time. It is assumed that the internal radius of each fuel element is initially coated with a uniform 6 μm thick layer of graphite (CANLUB) over its entire length. Considering a density of 1.7 g cm^{-3} [10] for nuclear grade graphite, the total mass of CANLUB is estimated at 7.3 g per 37-element fuel bundle. Amounts of Gd_2O_3 added to CANLUB is in the 100-300 mg range, which indeed, represents only very small mass fractions (1.3% to 4%).

5. Simulation Results and Discussions

The first step consists of modeling a 480 fuel channels CANDU nuclear reactor (Bruce A configuration) with MCNP and WIMS, as detailed in Figures 3 and 4. A full 3D core is modeled with MCNP and a lattice cell (a fuel channel) is modeled with WIMS. Geometrical buckling values (radial and axial) reflecting the CANDU reactor core radius and height are provided to WIMS, allowing for the code to account for neutron leakage and evaluate k_{eff} : at this point, the initial k_{eff} value can be estimated with both codes and compared.

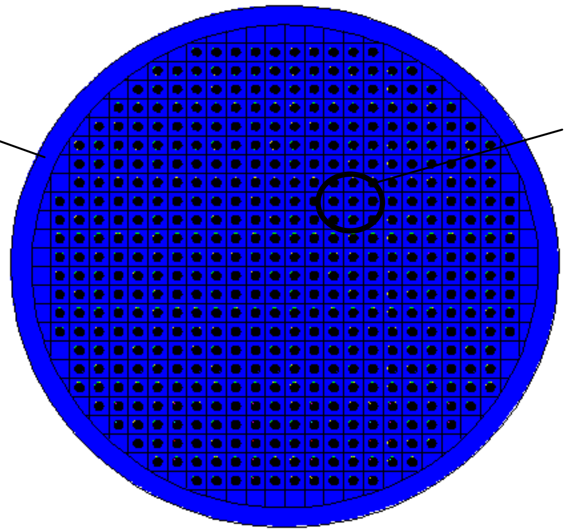

Figure 3: Cross-Section View of a 480 Channels CANDU Nuclear Reactor Model

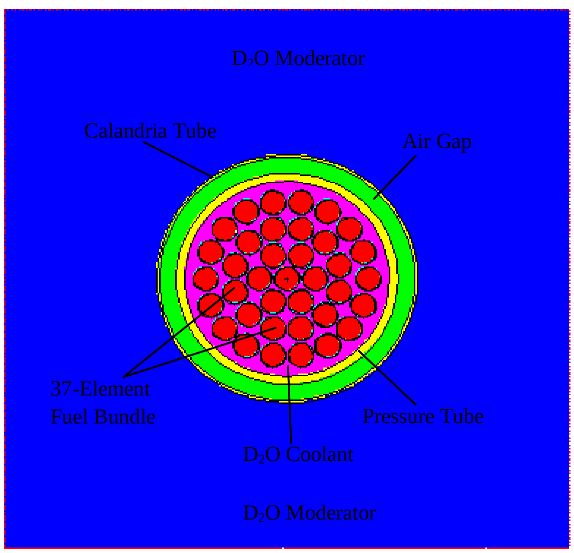

Figure 4: Cross-Section View of a CANDU Nuclear Reactor Fuel Channel (Cell) Model

Criticality calculations (k_{eff}) are performed with both codes for various amounts of Gd_2O_3 added to CANLUB. The results are shown in Table 3. Note that the MCNP results include an uncertainty equivalent to 3 standard deviations (level of confidence of 99%).

Table 3: Criticality Calculation Results (k_{eff})

Gd_2O_3 added per fuel bundle (mg)	MCNP (±0.003)	WIMS (±0.001)
0	1.107	1.105
9	1.105	1.103
30	1.098	1.096
100	1.077	1.077
140	1.068	1.066
150	1.066	1.064
160	1.061	1.061
170	1.056	1.057
180	1.054	1.055
190	1.053	1.053
200	1.051	1.050
308	1.025	1.024

Burnup calculations are then carried out with WIMS for each scenario. The intent is to observe the effects that Gd_2O_3 may have on reactor core reactivity while maintaining a channel output of 6.5 MW_{th}, the designed thermal power. The reactor is freshly fuelled and burns until it becomes critical (k_{eff} equals 1). The burnup evaluations are shown in Figures 5 and 6.

It is observed that a reactor loaded with fresh fuel bundles free of Gd_2O_3 has an initial excess of reactivity in the vicinity of 95 mk (k_{eff} of 1.105). The reactor experiences, as expected, a large drop in reactivity (and power) over the first 2 days of operation mainly due to fission products saturation, resulting in a reactivity value in the vicinity of 60 mk (k_{eff} of 1.065). Following this large drop (-35mk), the reactivity increases slowly for approximately 50 FPD, at which point the plutonium concentration reaches a maximum. The plutonium peak occurs near a burnup of 1000 MWd $Tonne^{-1}$ of U [11]. The excess of reactivity at this point is estimated at 67 mk (k_{eff} of 1.072), resulting in a small increase of 7 mk. Following the plutonium peak, the reactor reactivity slowly decreases (monotonically) until criticality is reached. Based on WIMS results, an average thermal neutron flux of 2.11×10^{14} n cm^{-2} s^{-1} is required to maintain a channel power of 6.5 MW_{th}. As the amount of Gd_2O_3 added to a fuel bundle increases, the initial reactor excess of reactivity decreases. Furthermore, one can also observe that the reactivity drop over the first 2 days of operation significantly decreases as the amount of Gd_2O_3 added to a fuel bundle increases.

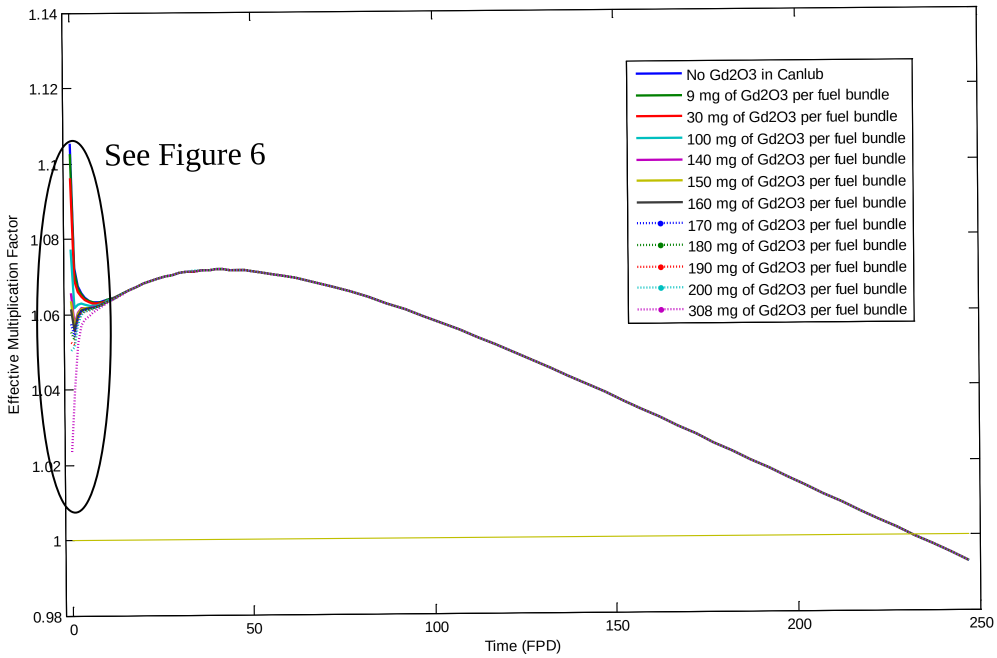

Figure 5: k_{eff} vs Time for a CANDU Reactor

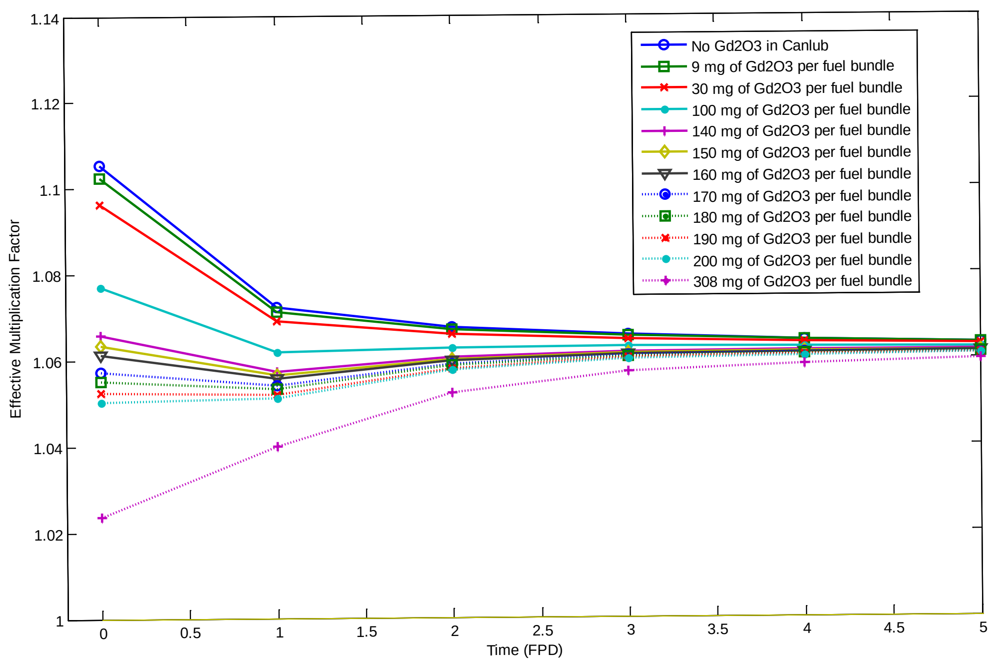

Figure 6: k_{eff} vs Time for a CANDU Reactor

It is shown that with 190 mg of Gd_2O_3 per fuel bundle, the initial reactor reactivity is in the vicinity of 50 mk (k_{eff} of 1.054). This reactivity value remains constant for the first day of operation and slightly increases, by approximately 2 mk, over the second day. Then, it gradually increases over the next 40-50 FPD until the plutonium peak is reached. Considering the main objective of the present work, this is quite an achievement when compared to a reactor fuelled with bundles free of Gd_2O_3.

It can be predicted that the gadolinium isotopes referred to as useful absorbers, Gd-155 and Gd-157, burn out within the first 3-4 FPD following a refuelling. Beyond the fourth day of operation, all curves from Figures 5 and 6 converge to a single one, corresponding to the curve generated by a reactor fuelled with fuel bundles free of Gd_2O_3. According to the burnup evaluations, the location of the plutonium peak and the useful life of a fuel bundle (burnup) remain unchanged, regardless of the amount of Gd_2O_3 initially added into the CANLUB of a fuel bundle.

5.1 Gadolinium Isotopes Depletion

The nuclear reactions with the natural gadolinium isotopes occurring in a thermal nuclear reactor such as CANDU are demonstrated in Figure 7.

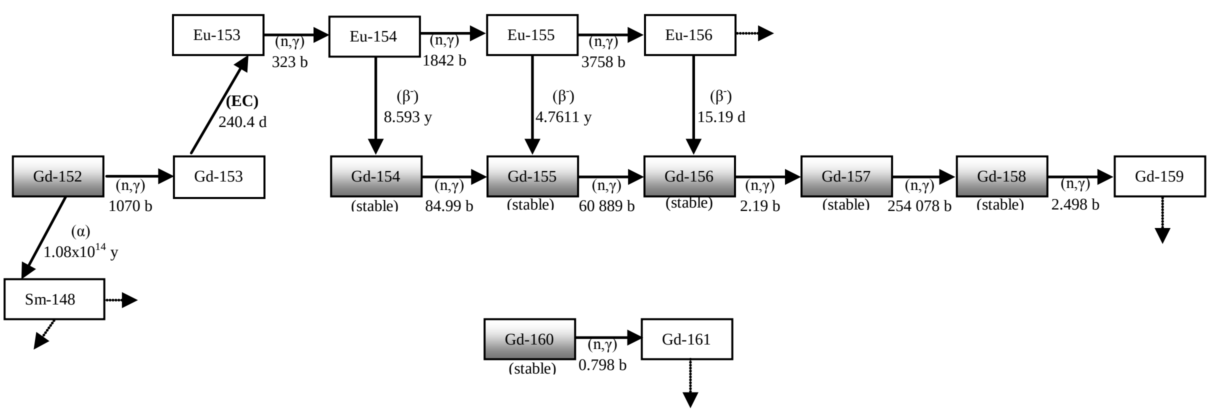

Figure 7: Nuclear Reactions with Natural Gadolinium in a Thermal Reactor

Of all the naturally occurring isotopes, shaded in grey, only Gd-152, with a molar fraction of 0.2 %, is radioactive. Due to its half-life of 1.08×10^{14} years, only traces of Sm-148 will be produced through alpha decay. Most of the Gd-152 atoms present, with a thermal absorption cross section of 1070 barns, will capture a neutron. With a half-life of 240.4 days, the Gd-153 atoms decay by electron capture to Eu-153. From there, Eu-154, Eu-155 and Eu-156 isotopes are produced through successive radiative captures. It can be demonstrated that the amounts of Sm-148 generated, as well as Gd-153, Eu-153, Eu-154, Eu-155 and Eu-156 rely entirely on the initial concentration of Gd-152 present in CANLUB. Despite their significant thermal absorption cross sections, their concentrations are too small to affect the channel reactivity significantly. It is also observed that Eu-154, Eu-155 and Eu-156 beta decay to Gd-154, Gd-155 and Gd-156. It can also be demonstrated that their contribution in terms of Gd atom production is very little, taking into account the small molar fraction of Gd-152 initially present and their relative positions in the chain.

The remaining of the natural gadolinium isotopes initially present in CANLUB are stable. Their depletion rate depends mainly on their thermal absorption cross section. It can predicted based on Figures 8 and 9 that Gd-155 and Gd-157, the useful absorbers, completely disappear within the first 3-4 FPD and largely contribute to the production Gd-156 and Gd-158. As previously mentioned, one can see a correlation between Figures 5, 6, 8 and 9. It is quite obvious that Gd-155 and Gd-157, with their important thermal absorption cross sections, are responsible for the suppression of the initial channel power ripple (power spike) following a refuelling. In other words, they reduce the channel reactivity following a refuelling by approximately 50%, minimizing channel (and zone) power fluctuations and control system actions without compromising the useful life (burnup) of the fuel bundles.

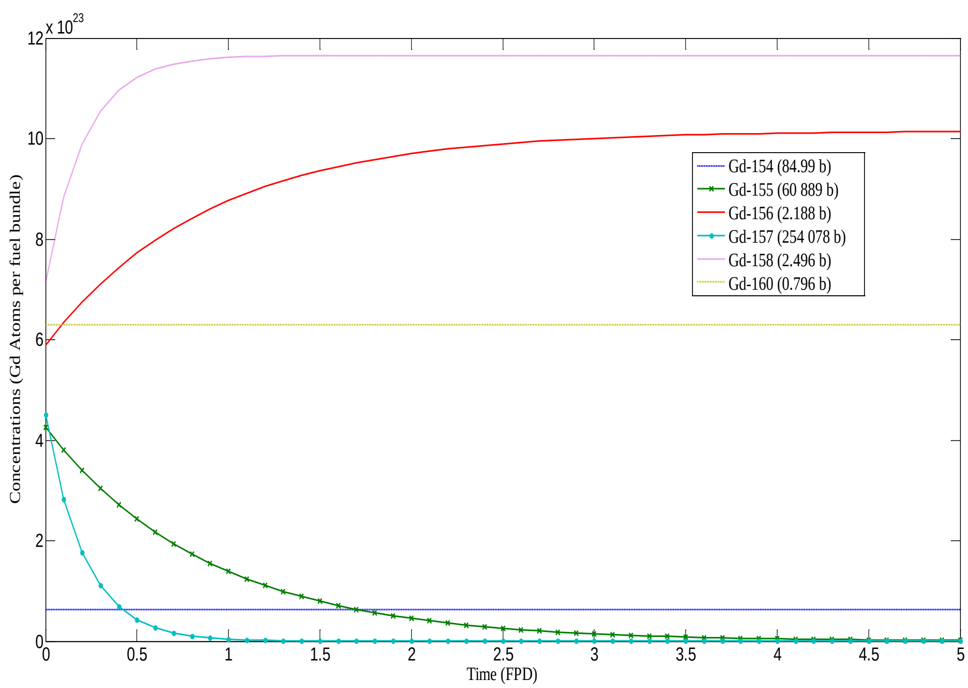

Figure 8: Depletion of Gd Atoms in a Thermal Reactor

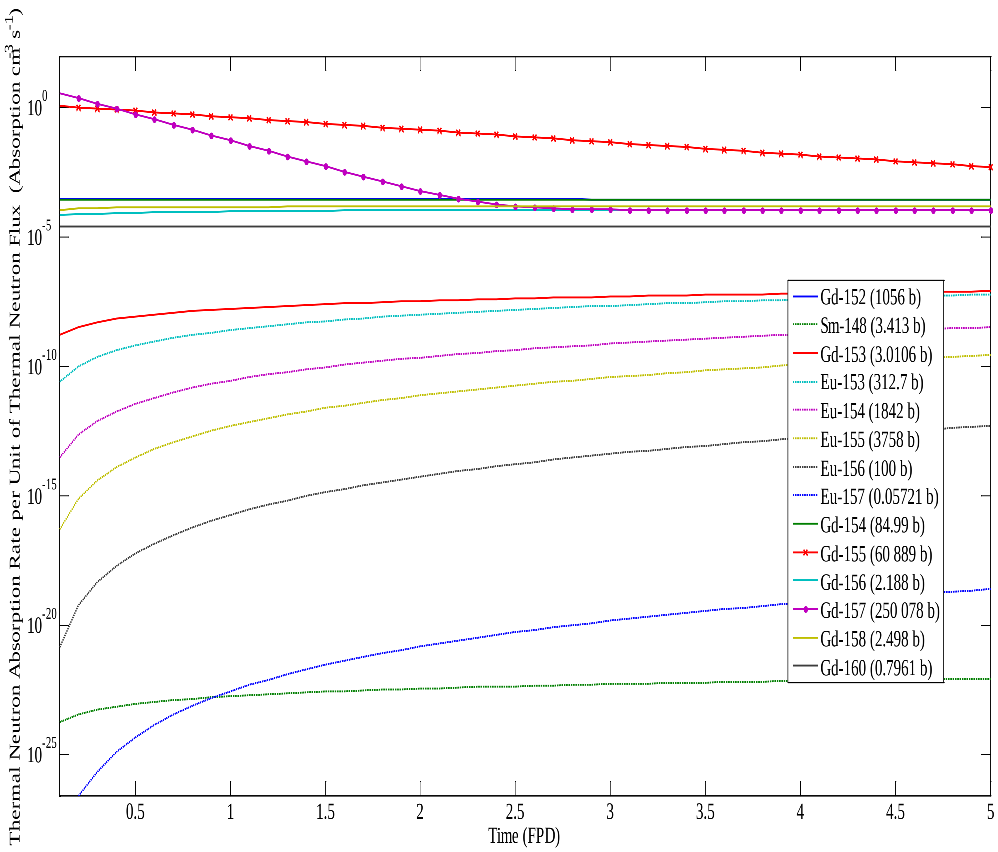

Figure 9: Macroscopic Cross Sections of Absorber Isotopes in a Thermal Reactor

The concentration of the other isotopes, referred to as the undesirable residual absorbers, including Gd-156 and Gd-158 following the burn out of Gd-155 and Gd-157, will remain fairly constant over the useful life of the fuel bundle, as seen on Figures 8 and 9, without affecting the reactor reactivity significantly.

6. Closing Remarks

The neutronic analysis results presented in the previous sections reveal that 190 mg of Gd_2O_3 per fuel bundle meet the objectives of the present work. As presented in Figure 10, 190 mg of Gd_2O_3 per fuel bundle clearly mitigate the fission products saturation and xenon free effects in fresh bundles loaded into the reactor core. From a safety aspect, it is anticipated that the ratio of the peak transient channel power relative to the steady state channel power will be reduced. Furthermore, the required actions from the zone reactivity control systems will also be less demanding.

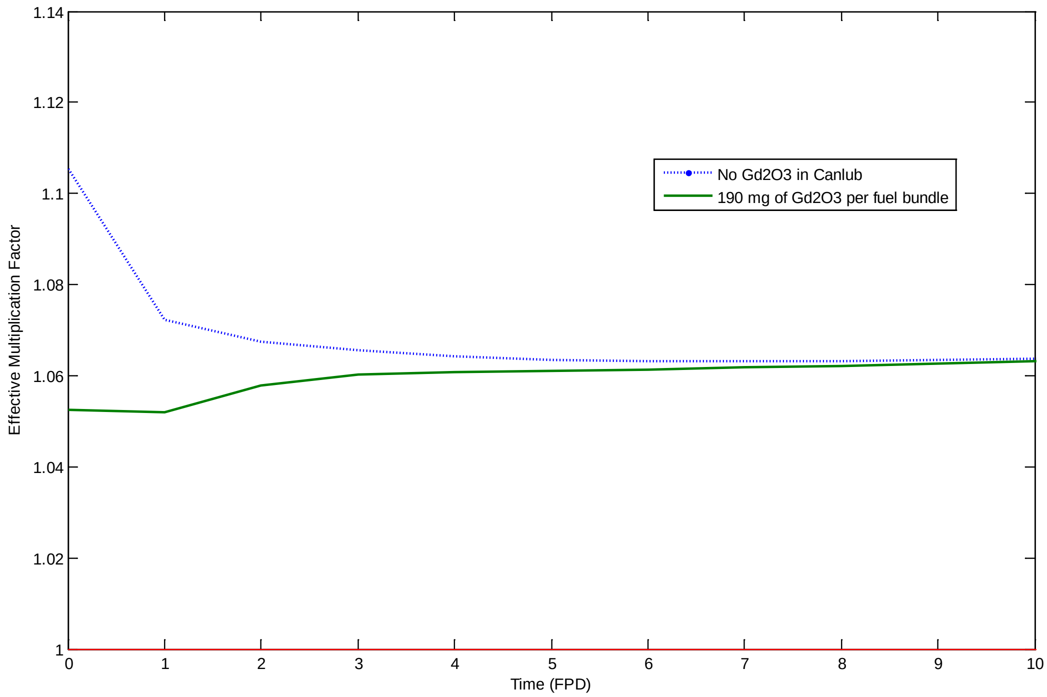

Figure 10: k_{eff} vs Time for a CANDU Reactor with 190 mg of Gd_2O_3 per Fuel Bundle

From a fuel management perspective, the refuelling scheme and/or Standard Operating Procedures (SOPs) could be modified. By reducing the amount of reactivity introduced by fresh fuel bundles into a specific channel, the amplitude of the refuelling ripples, dampened by the irradiated fuel of the surrounding channels, will not be as important as the ones currently occurring in operating CANDU reactor cores. In other words, inserting several fresh fuel bundles with Gd absorber in a given channel, will not cause a power ramp as high for the fresh bundles in the adjacent channels, thus permitting them to meet the power ramp criterion more easily. In the end, eliminating or reducing the amount of heavily irradiated fuel bundles in the reactor core translates into an increase in neutron economy although this may reduce the average discharged burnup of the fuel bundles.

7. Future Work

In the second phase of this project, neutronic analysis using various combinations of neutron absorbers will be carried out. The intent is to minimize power peaking and flatten axial power distribution at high burnup without compromising the useful life of a fuel bundle.

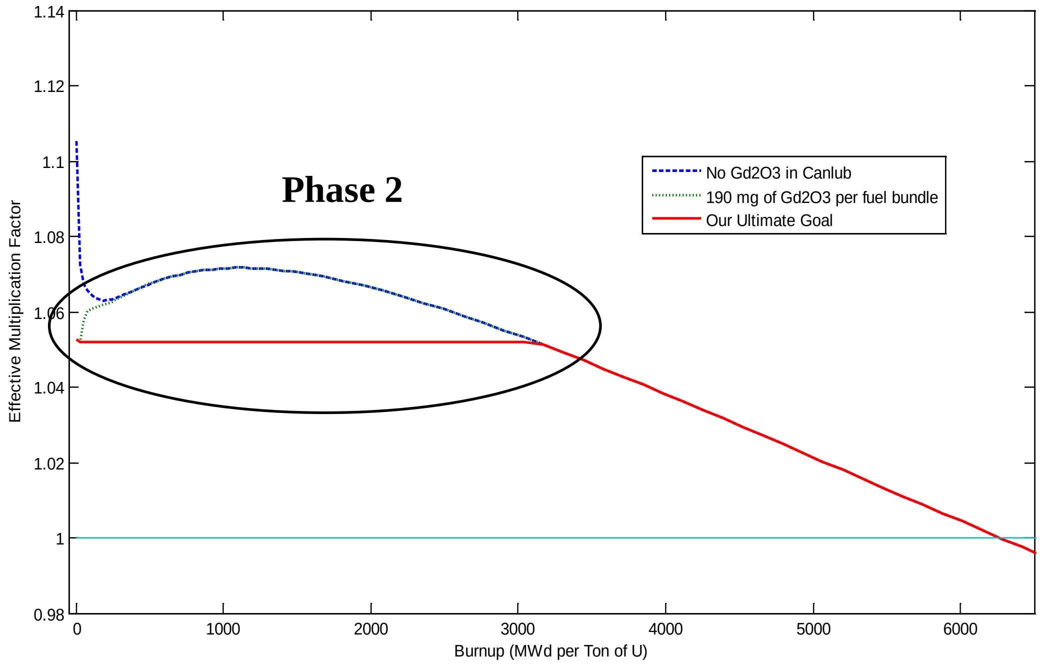

Figure 11: Ultimate Goal of the Present Project

Finally, power calculations will be performed with RFSP using the final fuel bundle design (doped with the final combination of neutron absorbers). The intent will be to investigate the power ripples and xenon free effects (fuelling transients) for various fuelling rates and bundle shifts (i.e. 2, 4, or 8 bundle shift). Potential benefits could be translated into a relaxation in the channel parameters compliance uncertainties, as well as a gain in Neutrons Over-Power (NOP) trip margin and mitigation for reactor derating.

8. References

[1] B. Rouben, "CANDU Fuel Management Course", Atomic Energy of Canada Limited, 2002, page 37.

[2] B. Rouben, "CANDU Fuel Management Course", Atomic Energy of Canada Limited, 2002, page 10.

[3] P.K. Chan, K.J. Kaddatz, "How Does CANLUB Work?", Atomic Energy of Canada Limited Report, 1996.

[4] Website: www.CanadianManufacturing.com.

[5] Brown, Forrest B., "MCNP – A General Monte Carlo N-Particle Transport Code, Version 5", Volume 1: Overview and Theory, Los Alamos National Laboratory, 24 April 2003.

[6] G. Jonkmans, "WIMS-AECL User's Manual, Version 3.1", Reactor and Radiation Physics Branch, Chalk River Laboratories, August 2006.

[7] S. Paquette, "The Super Near Boiling Reactor – Conceptual Design of a Small Nuclear Reactor to Supply District Heating and Electrical Energy to Canadian Forces Bases Located in the Arctic or Northern Remote Communities", Master's Degree Thesis, Royal Military College of Canada, April 2010.

[8] G. Jonkmans, "WIMS-AECL User's Manual, Version 3.1", Reactor and Radiation Physics Branch, Chalk River Laboratories, August 2006, page 36.

[9] Table of Nuclides: http//atom.kaeri.re.kr/.

[10] S. Glasstone, A. Sesonske, "Nuclear Reactor Engineering, Third Edition", Van Nostrand Reinhold Company, New York, N.Y., 1981, page 788.

[11] G. Jonkmans, "WIMS-AECL User's Manual, Version 3.1", Reactor and Radiation Physics Branch, Chalk River Laboratories, August 2006, page 31.