

The influence of thorium, uranium and plutonium fuel mixture composition on the reactivity coefficients of pressurised water reactors

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ABSTRACT

Temperature reactivity coefficients of a PWR (Pressurized Water Reactor) containing three fuel mixture compositions of uranium, thorium and plutonium in the form of UO_2 with Low Enriched Uranium, Th- UO_2 with Low Enriched Uranium and Pu-MOX with Low Enriched Uranium were analysed. The LEU UO_2 fuel is of 4.5% U-235 & 95.5% U-238 (Fuel model A). The LEU Th- UO_2 is of 5% U-235, 93% U-238 & 2% Th-232 (Fuel model B). And the LEU Pu-MOX is of 0.8% Pu(PWR), 4.5276% U-235 & 94.6724% U-238 (Fuel model C). Each fuel mixture composition was simulated in an infinite unit cell fuel pin model with reflective boundaries (a cylindrical fuel rod surrounded by a cuboid moderator) in Monte Carlo N Particle code (MCNP 6.2). A 17x17 lattice PWR core with a three-batch refuelling scheme of fresh, once burnt and twice burnt fuel is assumed and it was mimicked using the infinite unit cell fuel pin model.

The reactivity coefficients were analysed at the BOC (beginning of cycle) and at criticality ($k_{\text{eff}} = 1$). To achieve criticality, burnable absorbers gadolinia and soluble boron are added in the fuel models. The reactivity coefficients of the three fuel mixture compositions were sufficiently negative as required by regulatory bodies and were all within the set ranges in terms of magnitude as published in the source *Safety margins of operating reactors* by the IAEA, 2003. The DC (Doppler Coefficient) and MTC (moderator temperature coefficient) behave quite differently with increasing temperature. The DC becomes less negative with increasing fuel temperatures and the MTC grows strongly negative with increasing moderator temperatures. The largest effect on both the Doppler coefficient and Moderator temperature coefficient of the fuel model compositions is owing to the thermal and resonance absorption in the fuel isotopes. The total capture-fission ratio of all fuel models substantially increases with increasing fuel temperatures from CZP (293.6K) to 1200K. A decrease in thermal fissions and an increase in resonance absorptions is directly linked to a decrease in moderator density and it was found that the density is highly influential on the MTC. The moderator density substantially decreases with increasing moderator temperatures from 1.003g/cc at CZP(293.6K) to 0.6613g/cc at HFP (600K), then to 0.098g/cc at boiling (620K) even under a pressure of 15.5MPa(155bar). Boiling that is significant rarely occurs in a PWR and it is seen from the results that by the time it occurs, the reactor long reached sub-criticality ($k_{\text{eff}} < 1$). Overall, the LEU Pu-MOX and the LEU Th- UO_2 fuels produced more favourable temperature reactivity coefficients than the conventional LEU UO_2 fuel.

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Keywords: Reactivity Coefficients, Doppler coefficient (DC), moderator temperature coefficient (MTC), Critical Boron Concentration (CBC), UO₂ (Uranium dioxide), MOX (Mixed Oxide fuel), Thorium, fuel pin, PWR (Pressurized Water Reactor), MCNP (Monte Carlo N Particle Code).

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List of abbreviations, acronyms and symbols.

Acronyms, abbreviations and symbols	Definition
*DC	Buckling corrected Doppler Coefficient
Δ	Delta
a/o%	atom fraction enrichment percentage
B-10	boron 10
BA	Burnable absorber
BP	Burnable Poison
BWR	Boiling Water Reactor
CZP	Cold Zero Power (room temperature)
DC /FTC/	Doppler Coefficient, Fuel Temperature Coefficient
ENDF-B VII	Evaluated Nuclear Data Files
eV	electron Volt
FMESH	F MESH Tallies
Fuel model A	4.5% UO ₂ (Koeberg reference) / LEU UO ₂ fuel
Fuel model B	(5% ²³⁵ U, 93% ²³⁸ U & 2% ²³² Th)O ₂ /LEU Th-UO ₂ fuel
Fuel model C	(0.8% Pu(PWR), 4.5276 % ²³⁵ U & 94.6724% ²³⁸ U)O ₂ /LEU Pu-MOX fuel
Gd	Gadolinium
Gd₂O₃	Gadolinia
GWd/THM	Giga Watt days/ ton heavy metal
GWd/TU	Giga Watt days/ ton uranium
HFP	Hot Full Power (600k≈moderator, 900k≈fuel)
HZP	Hot Zero Power (600k ≈fuel, <600k moderator)
IAEA	International Atomic Energy Agency
IBA	integral Burnable absorber
IFBA	Integral Fuel Burnable absorber
K	Temperature Kelvin
KCODE	Criticality calculation code
keff	Finite neutron multiplication factor
kinf / K[∞]	Infinite neutron multiplication factor

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kinf(crit)	Lower operational limit Infinite neutron multiplication factor
LEU	Low Enriched Uranium
LWR	Light Water Reactor
Makxsf code	Make cross-section files code
MCNP 6.1 β/ 6.2	Monte Carlo N Particle
MOX	Mixed oxide
MPa	Mega Pascals
MTC	Moderator Temperature Coefficient
MWth	Mega Watt thermal
η	neutron yield factor
(n, alpha) / (n,α)	(neutron, alpha) absorption reaction
NPP	Nuclear Power Plant
O₂	Oxide
pcm	percent milli
pcm/k	percent milli/ kelvin
ppm	parts per million
Pu	Plutonium
Pu(PWR)	Reactor grade plutonium
Pu-MOX	Plutonium Mixed Oxide fuel
PWR	Pressurized Water Reactor
s(α,β)/ Sab	Alpha, beta thermal scattering
Sm	Samarium
Th	Thorium
U	Uranium
UOX/UO₂	Uranium Oxide/ Uranium dioxide
VRC	Void Reactivity Coefficient
WoBA	Without Burnable absorbers
wt%	weight percentage
x	void fraction
Xe	Xenon
xsdir	Cross-section directory
ZrB₂	Zirconium boride
β	Delayed neutron fraction or beta decay

**The influence of thorium, uranium and plutonium fuel mixture composition on the
reactivity coefficients of pressurised water reactors**

Acronyms, abbreviations and symbols	Definition
ρ	reactivity
σ_f	fission cross-section
σ_y	capture cross-section

1 INTRODUCTION

1.1 Background

When the temperature inside the core changes, various parameters in the fuel, moderator and coolant are subsequently altered and this directly affects the reactivity. Fundamentally the amount of heat generated from a fuel pin is determined by the number of fission occurrences in that particular fuel pin. As the neutron population varies over time, this has direct impact on the number of fissions and influences the reactor power and therefore the temperature of the Fuel and the surroundings. Neutronic properties may significantly change with changes in temperature and burn-up, and as a result alter the reactivity of the entire system. This change in reactivity with temperature is represented by the Temperature Reactivity Coefficients (TRCs).

In a Pressurized Water Reactor (PWR) there are two main reactivity coefficients associated with the changes in temperature, the fuel temperature coefficient/Doppler coefficient (DC) and the moderator temperature coefficient (MTC) - both are referred to as the TRCs. These provide a feedback that could either bring stability or instability into a reactor when the power and consequently the temperature changes. The DC is particularly important as it gives prompt feedback with temperature changes while the MTC has a slightly delayed response. The TRCs can be measured and their magnitude and sign (positive or - negative) basically determines the type of feedback to expect. The greater the magnitude of the TRCs of a particular fuel composition, the greater the sensitivity of the fuel composition to temperature changes (Edenius, 1976). The typical total TRCs values (sum of DC and MTC) of a PWR fuelled with LEU are in the range -5pcm/°C to -30 pcm/°C (Nyalunga, 2016).

But not all reactivity coefficients are directly linked to changes in core temperature. A reactivity coefficient such as the BRC (boron reactivity coefficient) is associated with changes in soluble boron concentration in the moderator with burn-up. Another, the VRC (void reactivity coefficient) which is mostly of concern in BWRs (Boiling water reactors) and liquid metal cooled reactors is associated with void formation (vapour bubbles) and drastic coolant/moderator density changes. It is important to note that in a PWR, the VRC is related to the MTC simply because changes in the void fraction means there's a change in the coolant/moderator density according to (Lamarsh & Baratta, 2001). As mentioned the sign (positive or negative) of the reactivity coefficients determines what feedback to expect and the magnitude of the reactivity coefficients determines

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the sensitivity to changes in that reactivity coefficients dependant parameter – the larger the magnitude of the reactivity coefficient the greater the sensitivity.

Understanding neutron nuclear reactions is the basis of how to build and furthermore how to optimise nuclear power systems particularly the nuclear fuel. With existing nuclear power reactors particularly, the PWR's, a few adjustments can be made to the Fuel composition to enhance the plants overall performance. The right blend of fertile and fissile material including burnable absorbers could lead to optimal fuel designs for PWR's with no need for any geometric alterations to the reactor or to the fuel itself.

Mainly the reactivity in a core depends on the enrichment, Fuel composition, poisons and temperature amongst other things (O. Safarzadeh, 2015). In the study titled "Increasing the Fuel cycle length of a PWR by means of a homogenous uranium, thorium and plutonium fuel design" a few fuel pins were designed for the Koeberg nuclear power plant. The fuel pins were designed with aims to optimize burn-up (BU) and increase the Fuel cycle length of the power utility by a considerably good margin. The fuel pins designed contained various compositions of fissile and fertile material namely LEU, MOX and thorium and the burnable absorber boron all incorporated into homogenous fuel model compositions.

A few of the designed Fuel compositions met the objectives and increased the fuel cycle length. However, like in most fuel designs a detailed study into the behaviour of the proposed Fuel compositions with changes in temperature must be conducted to determine the feasibility of the implementation of the design in an actual reactor. In other words, a study into how the U, Pu and Th fuel compositions influence the reactivity coefficients of a PWR must be conducted as this is crucial for criticality and safety during reactor operation.

1.2 Problem statement

In the quest for sustainable development in the nuclear energy industry, looking for alternative fuels to sustain nuclear resources for the next generations, to minimize the production of radio-toxic nuclear material and to minimize the risk of proliferation is the current and ongoing research in nuclear fuel design. Research into alternative fuels which include the fabrication of MOX Fuels of reactor grade plutonium Pu(PWR) and thorium-based fuels to be incorporated into existing PWR's is underway - MOX fuel has been used since the 1960's.

Different fuel designers come up with different fuel mixture compositions which may include either MOX with fresh UO_2 or thorium with fresh UO_2 or a combination of both alternatives depending

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on which aspect of traditional UO_2 fuel is desired to be improved. If fuels with such compositions are to be implemented in existing PWR's, it is important that key safety parameters of a reactor aren't compromised, and inherent safety, as provided by the temperature reactivity coefficients is retained.

The problem is that the magnitude and sign of the individual reactivity coefficients of the uranium, plutonium and thorium fuel compositions designed in the study titled: *Increasing the Fuel Cycle length of a PWR by means of a homogenous uranium, thorium and plutonium fuel design* by (Erlank, 2017) are unknown. For that reason, an in-depth investigation into reactivity coefficients of the various fuel compositions is to be carried out considering that the reactivity and power excursion analysis of a reactor mostly depends on the temperature reactivity coefficients. Furthermore, it is important to gauge the degree of inherent safety behaviour a reactor has by knowing the sensitivity of the reactivity coefficients of various fuel compositions. The two reactivity coefficients associated with a PWR are the Doppler reactivity coefficient (DC) and the moderator temperature coefficient (MTC). The VRC comes into play in a PWR due to nucleate boiling and severely in loss of coolant accidental situations (LOCA).

Regulatory bodies strictly require that temperature reactivity coefficients of a reactor adhere to certain safety standards. Firstly, both the DC and MTC must be negative because a negative reactivity coefficient ensures the desired reactivity feedback mechanism necessary for stable reactor operation in-case of a power transient without relying on external reactivity control for safety. Without this intrinsic safety feature in a reactor the regulatory body is authorized to deny the utility access to operate (Lamarsh & Baratta, 2001). Secondly, it is required that the magnitude of the temperature reactivity coefficients fall within a recommended range to give a feed-back that retains stable reactor operation. The recommended range for the DC is (-4.9; - 2.4) and the recommended MTC is in ranges (-70; 0) (IAEA, 2003).

Again, studies show that more favourable reactivity coefficients in PWRs are achievable with Th-based fuels and Pu-MOX fuels than with conventional UOX fuels. Not many studies investigate these findings in a simple pin cell model and see how the results match with full core models.

1.3 General aims and objectives of the study

Aim: Implement a safety analysis study of the homogenous uranium, thorium & plutonium fuel compositions by investigating the degree of inherent safety in terms of the DC and MTC and compare results with 4,5% LEU Koeberg fuel pin composition.

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General objectives of the study:

- Find a suitable and reliable method of analysing the reactivity coefficients of an infinite fuel pin model with reflective boundaries.
- Design a simple model that conceptually aims to mimic a full PWR core containing fresh, once burnt and twice burnt fuel and use it to analyse the two important TRCs in a PWR.
- Investigate and understand the factors that contribute to a negative feed-back in a PWR by looking at factors that influence the DC and MTC.
- Analyse the behaviour of the DC of U, Th and Pu fuels in terms of isotopic reaction rates.
- Analyse the behaviour of the MTC of U, Th and Pu fuelled in terms of isotopic reactions rates in the thermal, epithermal and fast neutron spectrum.
- Convert an infinite model to a finite model by theoretical means.
- Understand the use of burnable absorbers to reduce reactivity.
- Find the critical boron concentration of the uranium, plutonium and thorium Fuel model compositions.
- Understand the use of the KCODE, Burnup/depletion, makxsf code (make cross-section file to generate new nuclide data) and FMESH reaction rate Tallies in MCNP.

2 LITERATURE REVIEW

2.1 Homogeneous uranium, thorium and plutonium fuels

The literature in this section is based on a study by (Erlank, 2017), in which various fuel mixture compositions of uranium, thorium and plutonium were designed.

A few fuel pins have been designed for the Koeberg nuclear power plant (NPP) with aims to optimize burn-up (BU) and increase the fuel cycle length of the power utility. The fuel pins designed contained various compositions of fissile and fertile material in the form UOX and MOX. To this plutonium, thorium and boron, as a burnable absorber were added. All fuels were homogenous mixtures. The main idea behind the design of a fuel pin with such a mixture of isotopes (particularly those including thorium) was to achieve a lower starting reactivity at beginning of life (BOL), in order to reduce the need for boric acid in the water, and to include a fertile isotope that would breed fissile fuel over time and would therefore increase the reactivity towards the end of life, in order to increase the fuel cycle length, in order to increase the load factor of the reactor.

As for incorporating MOX fuel, the main idea was to use existing stockpiles of plutonium found in spent fuel to reduce the use of the limited resources of natural uranium, to reduce the burden on geological repositories and most importantly to minimize the risk of nuclear weapon proliferation.

Out of the tens of different isotopic compositions simulated, five met the objective to run for periods longer than 16 months. The feasibility criteria of the chosen fuel model compositions included reducing the infinite multiplication factor at BOC, reduced depletion rates of fissile atoms, increased breeding rates and therefore an increased fuel cycle length (Erlank, 2017).

2.1.1 The model

An infinite fuel pin model was created in MCNP 6.1 Beta to simulate the isotopic compositions of the various fuel designs. A cuboid control volume containing a cylindrical fuel pin surrounded by a moderator was used to model the isotopes. The geometry specifications of the fuel pin are those of the Koeberg NPP fuel pin. In an infinite fuel pin model reflective boundary conditions are used, and conditions assume that no neutrons leak from the periphery of the fuel core.

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2.1.2 Fuel model Concepts simulated

In all the UO₂ fuel model concepts simulated the atomic percentage enrichment (a/o %) was increased to 5 % (5 % U-235) compared to the 4.5 a/o % U-235 enrichment in the standard Koeberg NPP fuel pin.

All MOX fuels simulated included less than 8% Pu(PWR) and 92% DU. By law the maximum Pu content in MOX is limited to 10 % ($\text{Pu} \leq 10\%$). MOX 8-10 % reactor grade Pu and 90-92% depleted U. The Pu(PWR) recovered from spent fuel is comprised of the following isotopes of Pu, 2.763% Pu-238, 53.755% Pu-239, 23.615% Pu-240, 13.108% Pu-241, and 6.758% Pu-242 (Erlank, 2017).

2.1.3 Results

The fuel pins containing various isotopic compositions were simulated on the basis of meeting the objectives of the study which was to increase burn-up and increase the fuel cycle length. For the various fuel pins simulated the acceptability criteria were as follows:

- The initial infinite multiplication factor at BOC of the designed fuel must be equal or less than the initial infinite multiplication factor of the standard 4.5 % enriched UO₂ Koeberg fuel pin. ($k_{\infty} \leq 1.435$)
- K_{∞} of the fuel must stay well above 1.205 for a period longer than 16 months. Below 1.205 fission is no longer sustainable as k_{eff} then goes below one and that concludes the fuel cycle.

It should be noted that the aim was to obtain an understanding of the influence of the different isotopes on the neutron economy. Therefore, many synthetic compositions, i.e. compositions that cannot be realistically recreated in real life, were tested out. Examples include mixtures with Pu-241, but without the preceding Pu-239 and Pu-240. The fuel pins with the following isotopic compositions stood out from the rest in terms of increasing the fuel cycle length by more than 16 months:

- 5% U-235, 93% U-238 & 2% Th-232.
- 5% U-235, 94.9395% U-238 & 0.0605 % B-10.
- 10% MOX & 90% UOX (0.8% Pu(PWR), 4.5276% U-235 & 94.6724% U-238).
- 5% Pu-241, 47.5% Pu-238 & 47.5% Th-232.
- 5% Pu-241, 47.5% Pu-238 47.5% U-238.

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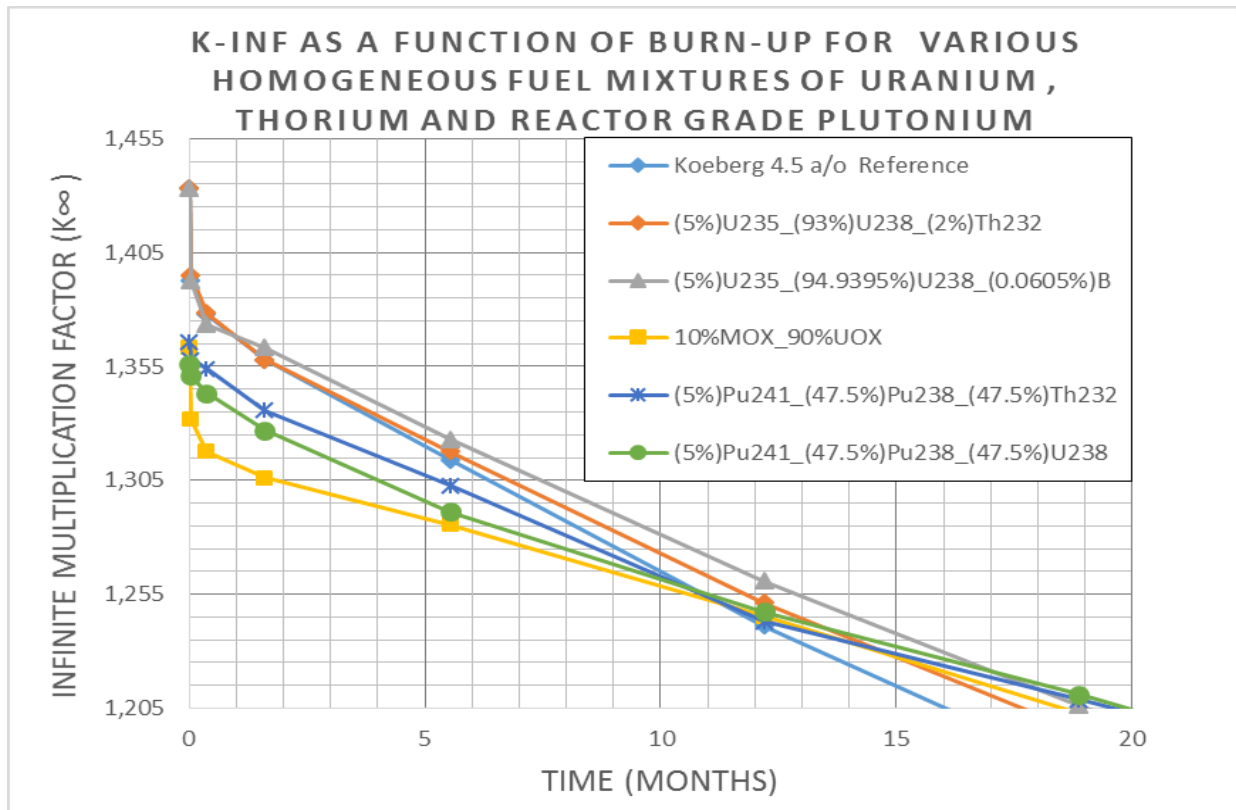


Figure 2-1: Comparison of the infinite multiplication factor over time due to burnup of various fuel mixtures courtesy of (Erlank, 2017).

The fuel pins containing Pu-238 and 5% Pu-241 performed quite exceptional, meeting the desired standards in terms of reducing the initial k_{∞} and keeping it well above 1.205 for periods longer than 16 months as seen in Figure 2-1. In terms of feasibility, it would be difficult to implement fuel pins with such compositions mainly because of the nuclide ^{238}Pu .

Pu(PWR) of spent nuclear fuel contains a small percentage ($\leq 2\%$) of Pu-238 (Nuclear chemistry, 2018). Because it comes in such small quantities in spent fuel and it possess such a brief half-life (87.7 years), it is dangerous and not worthwhile recovering it from spent fuel. It contributes to the decay heat produced by spent fuel and usually fully decays while in the intermediate storage. Through emitting an alpha particle, Pu-238 decays to U-234 which has a half-life of tens of thousands of years technically making ^{234}U stable.

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Pu-238 can also be produced by irradiating actinides Np-237 and Am in a nuclear core and again by bombarding U-238 with deuterons (2 protons) on a beam line. Though Pu-238 can be obtained in numerous ways it cannot be mass produced for nuclear fuel applications.

The following fuel mode compositions are implementable therefore meet the feasibility criteria:

- 5% U-235, 93% U-238 & 2% Th-232.
- 5% U-235, 94.9395% U-238 & 0.0605 % B-10.
- 10% MOX & 90% UOX (0.8% Pu(PWR) 4.5276% U-235 & 94.6724% U-238).

2.2 Fissile and Fertile material

Amongst a few other things, the main factor that determines the reactivity of a reactor is the fuel isotopic composition. The cross-section data of the fissile and fertile isotopes used in fuel can help with predicting which type of neutron- nuclei interactions are dominant and at which neutron energies are the interaction taking place. The reactivity of the fuel compositions changes with burn-up and temperature changes and by understanding the neutronic properties of the fissile and fertile isotopes with these changes we can be able to deduce the dynamic behaviour of a reactor. For that reason, it is important to know the properties of the fissile and fertile isotopes used in the designed fuel model compositions.

2.2.1 Fissile isotopes

Overview:

In this section we briefly discuss the neutronic properties of fissile isotopes (U-233, U-235, Pu-239 and Pu-241), looking at the similarities and distinct differences that make each one unique for purposes of maintaining criticality and stability in a nuclear core. Each fissile isotope is primarily distinguishable by its microscopic cross-sections for fission and capture (σ_f & σ_c), its capture to fission ratio (α) and neutron economy, which is best represented by the number of fission neutrons produced per neutron absorbed in that fissile isotope (η). From a safety standpoint it is important to consider the influences of each fissile isotope on reactivity as the neutron spectrum hardens. The delayed neutron fractions (β) of fissile isotopes play a crucial role considering reactor kinetics and safety. Each isotope has a unique delayed neutron fraction.

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Fission product poisoning, primarily by Xe-135, adds to the dynamic behaviour of a reactor and again each fissile isotope has a distinct yield of fission product poisons.

Any nuclear data given is at 0.0253eV (ideal thermal neutron energy) corresponding to neutrons with a speed $v=2200\text{m.s}^{-1}$ which is the most probable neutron speed at room temperature (20°C) (Lamarsh & Baratta, 2001).

2.2.1.1 Fissile isotope Fission cross-section description

U-235 is the only naturally occurring fissile isotope and it is scarce in nature (only $\approx 0.73\%$ of natural uranium). The rest of the isotopes U-233, Pu-239 and Pu-241 are born from the transmutation of certain fertile nuclides, after capturing a neutron in a reactor core, U-233 from Th-232, and Pu-239 and Pu-241 from U-238 (Lamarsh & Baratta, 2001).

The fission cross-sections of all fissile isotopes resemble three distinct regions in accordance with the three neutron energy domains, namely thermal, epithermal and fast neutrons. At thermal energies a $1/v$ relationship can be observed with the fission cross-sections, at epithermal energies (slowing down range) the resonances dominate and above threshold energies (fast fission range) the fission cross-section diminishes (Lamarsh & Baratta, 2001). The fission cross-section of the plutonium fissile isotopes (Pu-239 and Pu-241) is much higher than that of the uranium isotopes (U-233 and U-235), the highest being for Pu-241.

For thermal reactor applications the best fissile material to use is U-233. According to (Greneche, 2018) U-233 has the highest value of (η) for thermal and epithermal neutron energies and thus possesses much more favourable properties than U-235, Pu-239 and Pu-241 for thermal reactors.

2.2.1.2 Spectrum shift

In a thermal reactor, thermal neutrons are the dominant group because of the presence of a moderator, but it is also important to look at the fission cross-section of fissile isotopes at higher neutron energies considering that the neutron spectrum becomes slightly harder with increasing power and temperature in the system. Two different mechanisms are involved:

- Neutrons that are already thermalized are by definition in thermal equilibrium with the water moderator. So, if the water temperature increases, the average kinetic energy of the thermal neutrons increases to the same temperature and thus the Maxwellian energy spectrum moves up to higher energies, i.e. it hardens (Lamarsh & Baratta, 2001).

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Since the plutonium fissile isotopes, both (Pu-239 and Pu-241) both have a fission resonance peak between 0.2eV-0.4eV, the high energy tail of the Maxwellian spectrum distribution will push up into these resonances, which will increase the number of fissions in them and thus will add undesired reactivity to a plutonium dominated core (Erlank, 2017). On the other hand, the reactivity of both U-235 and U-233 tend to decrease with increasing water temperature-(non-1/v factor pg78, Table 3.2 (Lamarsh & Baratta, 2001)) this makes these fuels more stable at thermal energies in terms of desired reactivity behaviour (Du Toit, 2013).

- The liquid moderator is very sensitive to temperature changes and becomes slightly less dense with increasing temperatures. Therefore, higher temperatures mean less moderation in the fuel core, which reduces the rate at which the fast neutrons are moderated down to thermal energies. This reduces the fraction of neutrons that are already thermalized and increases the fraction of epithermal neutrons that are still in the process of being moderated. Since in e.g. LEU fuels, fission dominates in the thermal energy region and radiative captures in the epithermal capture resonances of U-238 dominates in the epithermal energy region, this means more captures and less fissions, which will reduce the reactivity. (Edenius, 1976).

2.2.1.3 Neutron economy

At thermal energies, U-233 has the highest neutron yield factor ($\eta=2.287$) compared to other fissile isotopes U-235 ($\eta=2.068$), Pu-239 ($\eta=2.108$) and Pu-241 ($\eta=2.145$). This neutron economy given by U-233 is advantageous more especially for thermal reactors because it is in excess for breeding ($\eta > 2$). U-233 has the lowest capture to fission ratio ($\alpha= 0.089$) in comparison to the other fissile isotopes, U-235 ($\alpha=0.169$), Pu-239 ($\alpha=0.362$) and Pu-241 ($\alpha=0.365$). This means that in the case where a thermal neutron is absorbed the probability of fission in U-233 would be higher than for any other fissile isotope though relatively the actual fission cross-section of U-233 is lowest amongst fissile nuclides (Nuclear-power.net, 2018). This translates that the capture cross-section in U-233 is much lower compared to its fission cross-section that undesired neutron captures that could lead to undesired transmutations are less likely to occur (Du Toit, 2013).

2.2.1.4 Capture to fission ratio

As the neutron spectrum hardens the capture to fission ratio of the fissile uranium isotopes (U-233 & U-235) gradually increases up to neutron energies of 100KeV, indicating that the probability of fission gradually decreases while capture slowly increases. An opposite trend is observed in Pu-239 where the capture to fission ratio drastically decreases up to 100keV indicating an increase in the fission probability as opposed to capture (Nuclear-power.net, 2018).

2.2.1.5 Delayed neutron fraction

Considering reactor kinetics, the time behaviour of a reactor and its controllability are very much dependant on the delayed neutron fraction (β). It can be declared that the higher the delayed neutron fraction is the higher the reactor period and the easier reactor controllability becomes. The fission products of U-235 produce a much higher delayed neutron fraction, compared to the other fissile isotopes. For this reason, U-235 fuelled reactors are much less prone to uncontrollable reactivity swings. Therefore, they require less safety and control measures during transients. The substantially lower delayed neutron fractions of U-233 and the fissile plutonium isotopes (Pu-239 & Pu-241) are very similar to each other, which makes them behave kinetically similar (Du Toit, 2013).

2.2.1.6 Fission poisons

The accumulation of neutron poisons (Sm-149 & Xe-135) in a core arising as daughter products of fissionable isotopes must be considered. The two aforementioned poisons are aggressive neutron absorbers because of their exceptionally large absorption cross-sections which have a dynamic effect on reactivity throughout the fuel life cycle but a more drastic dynamic effect with power level changes (Stacey, 2007). Each fissile isotope has a unique fission product poison yield (Lamarsh & Baratta, 2001). Being the lighter fissile isotope, U-233 has the lowest fission poisons yield which translates to less absorption effects and lower reactivity changes that could subsequently lead to premature subcriticality. This further translate to longer fuel life cycles (Du Toit, 2013).

The average number of prompt neutrons emitted per fission (ν) by each fissile nuclide increases with incoming neutron energy (Stacey, 2007). This effect is negligible for small changes in neutron energy, e.g. a harder or a softer thermal spectrum. In fact, epithermal fissions normally produce worse neutron economies than thermal fissions. However, large increases in neutron energy

gives substantial increases in ν . Therefore, fast reactors produce much better neutron economies than thermal or epithermal reactors and therefore fast reactors are the best breeder reactors.

2.2.2 Fertile isotopes

Fertile nuclides possess unique neutronic properties that are of primary importance in reactor safety and again have proven to prolong the fuel life cycle. In this section general neutronic properties of fertile nuclides are discussed comparing the two predominant nuclides, namely Th-232 and U-238.

2.2.2.1 Cross-sections

There is a small, but significant probability for fast fission in the fertile nuclides Th-232 and U-238. Fast fissions do occur in thermal reactors and they contribute a significant fraction toward the total fission energy, approximately 8% fission energy from U-238 and 2% from Th-232. It is clear from this that the probability of fast fissions for Th-232 is unfortunately much smaller than for U-238, which is a definite disadvantage for Th-232. This is because U-238 has a higher fission cross-section in the fast neutron spectrum than Th-232 (Du Toit, 2013). Fast fissions could be advantageous for the neutron economy of a thermal reactor because generally when a fertile isotope fissions at fast energies, it produces more neutrons than when a fissile isotope fissions at thermal energies but thermal neutrons are the dominant group in thermal reactors (Erlank, 2017). Furthermore, fast fissions of fertile isotopes consume only the almost worthless fertile isotopes, as opposed to thermal fissions that consumes the expensive fissile isotopes.

The fission cross-section of U-238 becomes significant only above 1MeV where it slowly approaches 1barn (10⁻²⁴ cm²) with increasing neutron spectrum till it goes slightly above 1barn around 10MeV. A similar trend can be observed for the fission cross-section of Th-232. The Th-232 fission cross-section slightly increases in a stepwise manner from 1MeV to 10MeV. But the ²³²Th fission cross-section is a factor of 3-5 times lower than that of U-238 so the Th-232 fission cross-section barely approaches 1barn at 10MeV neutron energies (Stacey, 2007).

The radiative capture cross-section (σ_γ) for both Th-232 and U-238 shows the $1/\nu$ dependence below the resonance region (thermal energies). With a lack of consistency, a similar trend ($1/\nu$ trend) can be observed above the resonance regions between certain neutron energies, but overall capture both in Th-232 and U-232 becomes insignificant above threshold energies (Stacey, 2007).

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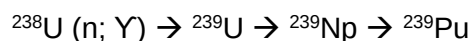
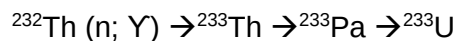
According to (Talamo & Gudowski, 2005), to bring the capture reaction rate of Th-232 and the fission reaction rate of fissile nuclides to equilibrium at thermal energies, a quantity of Th-232 25-30 times greater than that of the fissile material is required. The aforementioned shows how small the capture cross-section of fertile isotopes is compared to the fission cross-section of fissile nuclides at thermal energies considering that the capture cross-section of Th-232 is larger compared to that of U-238 for the same thermal neutron energy.

From the abovementioned it can be drawn that thermal reactors fuelled with Th-232 will generally require larger enrichments of fissile material to be critical compared to those fuelled with U-238. This is mainly because Th-232 possess a larger capture cross-section than U-238 at thermal energies (Du Toit, 2017). However, a larger capture cross-section gives Th-232 a favourable breeding/conversion ratio at thermal energies as compared to U-238 (Du Toit, 2013).

With both fertile isotopes, the ideal energy range for breeding is in their epithermal spectrum range where the resonance peaks dominate. Noting that the resonance peaks rise to above 1000 barns, when the neutron spectrum hardens capture in resonances will dominate thus making conversion/breeding more favourable. A slight spectrum shift towards the epithermal range is advantageous in that sense (Du Toit, 2013).

2.2.2.2 Transmutation

The time it takes for a fertile nuclide to transmute into a fissile nuclide is an important factor to consider for the reactivity of the system. The thorium fuel cycle (Th-232 to U-233) takes roughly 27 days because of the unstable isotope Pa-233 (half-life=27days) in the transmutation chain. And the uranium fuel cycle (U-238 to Pu-239/ U-238 to Pu-241) takes only 2.3 days because of the isotope Np-239 (half-life=2.3days) in the transmutation chain (Greeneche, 2018).



It seems one of the reasons the thorium fuel cycle requires such high fissile enrichments is because of its delayed transmutation chain to produce fissile U-233. The delay long enough for undesired transmutations to occur where Pa-233 captures a neutron and produces an undesired nuclide hampering the production of fissile U-233 (Du Toit, 2013).

However, the weaker starting reactivity of the thorium fuel cycle can be a gift as opposed to being curse. The long delay is a huge advantage in the sense that fissile material slowly accumulates

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overtime and most of this fissile material will fission at a later stage in the reactor provided a thermal neutron is absorbed. That alone can increase the fuel life cycle length for a higher plant availability. It can potentially increase the fuel cycle length by almost 6 months (Van der Walt, 2016).

The uranium cycle is advantageous in a sense that it has a quicker stronger starting reactivity because it produces more fissile material in 2-3 days and again the uranium fuel cycle has the potential to produce two fissile nuclides Pu-239 and Pu-241.

One way of to produce nuclear energy with low radio-toxic nuclear waste and most importantly proliferation resistant nuclear waste is through the thorium fuel cycle. The biggest problem with the thorium fuel cycle is the yield of isotope U-232 which produces a decay chain that is a dangerous gamma ray emitter. This may be seen as a huge problem in cases where the fuel has to be recycled but potentially this is what also makes thorium-based fuels proliferation resistant (Du Toit, 2013).

Thorium based fuels produce U-233 which has a number of advantages over Uranium fuels, they have Higher fissile conversion rates, Higher neutron economy, Low capture to fission ratio Low actinide production and Pu and Chemical stability (Tucker, et al., 2014).

2.3 Burnable absorbers

Burnable absorbers (BAs) or burnable poisons (BPs) are materials which absorb neutrons very aggressively because of their very high neutron absorption cross-sections, where after they transmute into materials of low neutron absorption cross-sections. BAs play an important role in the control and safety of a reactor primarily by lowering or completely shutting down the fission chain reaction, they add the negative reactivity necessary for stable reactor operation (criticality) over time and the necessary negative reactivity when it calls for shutdown.

There's a wide range of BA materials the most commonly used in nuclear reactors is boron-10 in the form of boric acid (H_3BO_3) and boron-carbide (B_4C). Boric acid is uniformly distributed in the moderator in a variety of concentrations and it is controlled by the Boron Thermal Regeneration System (BTRS), where boron-carbide is mostly in the form of mechanical control rods which are submerged in or withdrawn from the top of the core to fulfil their function.

Another form of BA that is widely used is gadolinium (Gd) in the form of gadolinia (Gd_2O_3). Gadolinia is a type of BA that is embedded directly in the fuel, there's a wide range of

these type of BAs and they are commercially known as Integral Fuel Burnable Absorbers (IBAs or IFBAs). IBAs can be homogeneously mixed with the fuel material or applied as a layer on the fuel pellet itself like Zirconium Boride (ZrB_2) and Erbium Oxide (Er_2O_3). In the case of gadolinia, it is homogeneously mixed with the fuel pellet material (Bjork, 2012).

Moving the control rods (B_4C), changing the concentration of the boric acid (H_3BO_3) and adding integral burnable absorbers (IBAs) Gd_2O_3 , in the fuel are the main ways in which the reactivity of a PWR is controlled. Because of the type of model used in this study, (model described in section 2.1.1) (Erlank, 2017), the relevant measures of reactivity control in this study includes only boron in the form of boric acid and gadolinium in the form of gadolinia.

2.3.1 Soluble boron

Soluble boron compensates for reactivity changes during burn-up (long term reactivity control). During burn-up the reactivity goes through many inevitable changes because of the changes in fuel composition overtime, fission products build-up, fissile material depletion and actinides build-up. These changes have an adverse influence on the reactor dynamics and soluble boron comes in handy to compensate for most of these changes to keep the reactor critical (Lamarsh & Baratta, 2001). Soluble Boron controls the slow reactivity changing process during a cycle (Rui, et al., 2017). The boron added is varied in concentration and the boron concentration required to compensate for a specific positive reactivity during the cycle burn is called the critical boron concentration (CBC) and it is usually measured in parts per million (ppm).

Soluble boron significantly increases the absorption in the moderator and therefore significantly changes the thermal/fuel utilization factor (f) affecting the multiplication factor (K) (Lamarsh & Baratta, 2001). Unlike control rods, soluble boron uniformly concentrates in the moderator, flattening out the neutron flux making the power in the core more uniform (Rui, et al., 2017).

As much as soluble boron is responsible for reactivity control and compensating for any positive reactivity, too much concentration of it in the moderator can have an adverse effect on the reactor's inherent safety characteristics. A large concentration of boron in the

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moderator tends to produce a positive MTC, an undesirable trait for a PWR as mentioned in quite a number of studies on LWRs (Nyalunga, 2016). It is important to take note that the concentration of soluble boron in PWR's is limited to 1500 ppm (part per million) for safety reasons (Rui, et al., 2017).

The effect of boric acid concentration on reactivity is described by the boron worth/boron reactivity coefficient (BRC) expressed by formula 2.0 below. Typical boron worth in a PWR (-10.86; -5.71 pcm/ppm) (IAEA, 2003).

$$W_b \cong \frac{\partial \rho}{\partial C_b} \quad (2.0)$$

W_b = boron worth/ boron reactivity coefficient (BRC) (pcm/ppm)

C_b = boric acid concentration in parts per million (ppm)

Typical boron worth in a PWR (-10.86; -5.71 pcm/ppm)

The two naturally occurring isotopes of boron are B-10 (19.6 % abundance) and B-11 (80.4 % abundance) (Lamarsh & Baratta, 2001). The high absorption abilities of soluble boron result from the isotope B-10 ($\sigma_a=3460$ barns at 0.025eV) which undergoes an (n, alpha) reaction with thermal neutrons, resulting in Li-7 (Lamarsh & Baratta, 2001). B-10 has a high absorption cross-section with approximately $1/v$ dependence at thermal neutron energies as shown in the figure below.

The influence of thorium, uranium and plutonium fuel mixture composition on the reactivity coefficients of pressurised water reactors

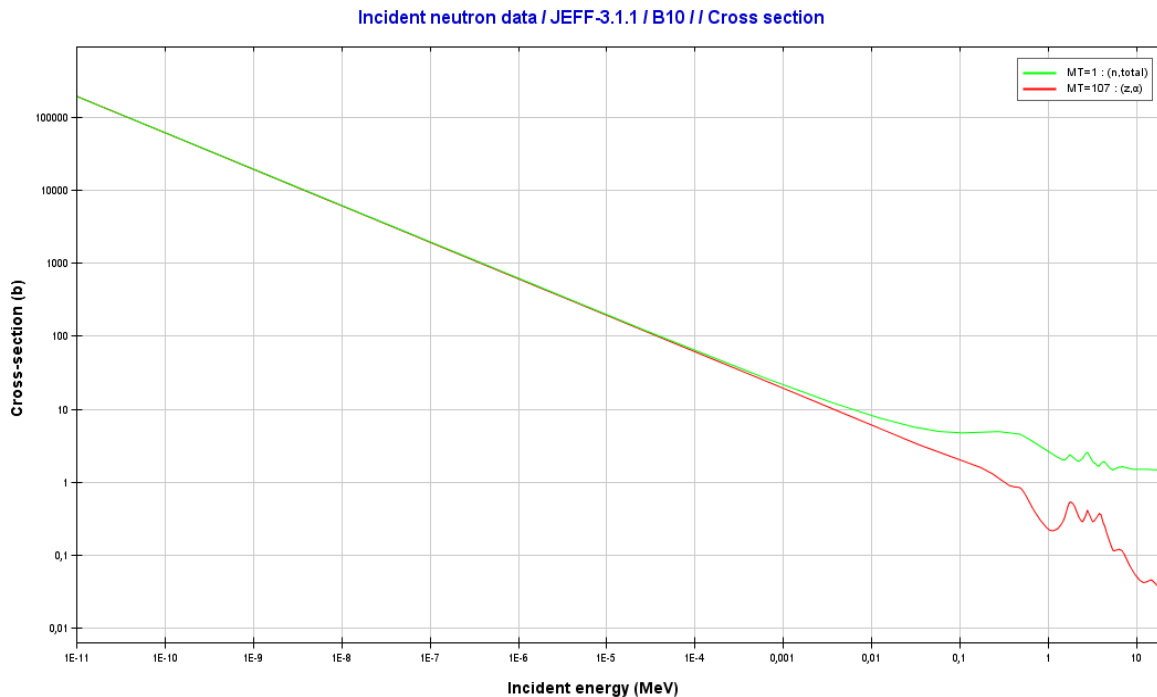


Figure 2-2: Comparison of total cross-section and (n, alpha) reaction cross-section for boron 10 (Nuclear-Power, 2017).

2.3.2 Gadolinia

Natural gadolinium contains seven isotopes and only two out of the seven have the desirable neutronic properties for a burnable absorber (BA) characterised by a significantly high thermal neutron absorption cross-section. Gd-155 with a natural abundance of 14.8 a/o and a thermal neutron absorption cross-section of 61 000 barns and Gd-157 with a natural abundance of 15.65 % a thermal neutron absorption cross-section of 254 000 barns. Amongst all stable isotopes and amongst burnable absorbers in general, the two gadolinium isotopes have the highest thermal neutron absorption cross-sections and compared to their BA counterparts, they burnout relatively fast (Nuclear-power.net, 2018).

Gadolinium is added in the form of gadolinia (Gd_2O_3) in fresh fuel as an IBA to compensate for the high initial reactivity associated with fresh fuel assemblies (Galahom, 2016). And because gadolinia is an aggressive neutron absorber it is added in small amounts, not more than 5% of the fuel material - all depending on the fissile enrichment of the fuel. Again only a few fuel pins in the fuel assembly contain gadolinia (Gd_2O_3) and

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these fuel pins are carefully distributed in the fuel assemblies to even out the neutron flux and therefore the power in the core during operation.

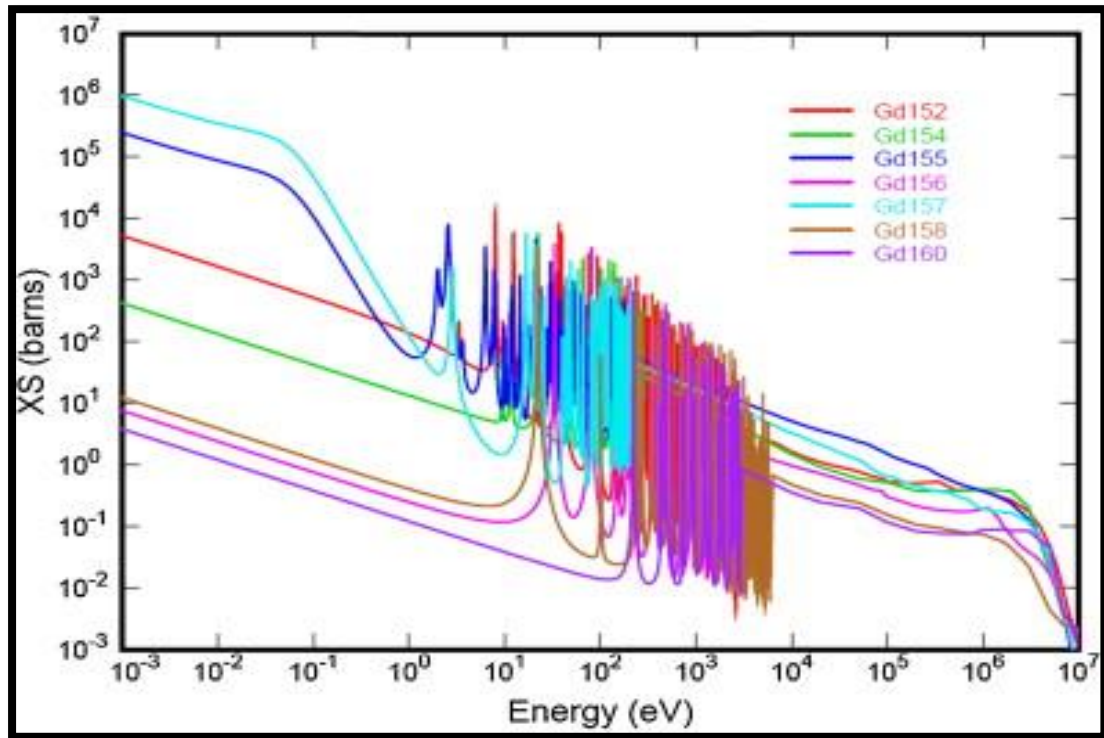


Figure 2-2: Neutron absorption cross-section (σ_a) spectrum for all seven isotopes of Gadolinium (Gd).

Figure 2-2 above shows the Neutron absorption cross-section (σ_a) spectrum for all Gd isotopes. The absorption cross-section of the two predominant Gd isotopes (Gd-155 and Gd-157) is substantially high at thermal neutron energies with the dominant reaction being radiative capture ($n; \gamma$). As mentioned in (Nuclear-power.net, 2018), BAs particularly Gd behaves like a black body, using the analogy to describe the absorption characteristics of Gd-155 and Gd-157 at thermal neutron energies.

The spectrum shifts and gets harder with increasing temperature due to the moderator becoming less dense and thus moderating less. In such cases, the absorption in Gd decreases because the capture cross-section at high neutron energies for Gd-155 and Gd-157 is substantially low. - The absorption decreases because of the non- $1/v$ behaviour of the Gd isotopes cross-section at thermal energies, i.e. the fact that the capture cross-section decreases faster than $1/v$ with increasing neutron energy.

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After the two prominent Gd isotopes have burnt out approximately after 10 months (Du Toit, 2013), some negative reactivity remains for some time arising from the remainder Gd isotopes (Galahom, 2016). The isotope ^{152}Gd has quite a significant absorption cross-section (1050 barns) but its abundance is quite small, about 0.2 % so its impact on absorption is not very significant. The other isotopes are quite abundant, but their absorption cross-sections put together is barely 90 barns.

One other key function of gadolinia as an IFBA (integral fuel burnable absorber) in fuel assemblies is to decrease the initial concentration of soluble boron at BOC. Excess amounts of soluble boron could have an adverse effect on the MTC/VRC if its concentration is too high (Nuclear-power.net, 2018). The beauty of using soluble boron together with burnable absorbers is that the critical boron concentration can be reduced to allowable limits (<1500 ppm). Figure 2-3 below displays the boron let-down curve (boron burn-up) over a 12 months cycle. The two curves demonstrate the CBC at BOC with BA (red) and without BA (blue) (Nuclear-Power, 2017).



Figure 2-3: The CBC in a core without burnable absorbers (blue curve) and core with burnable absorbers (red curve) (Nuclear-Power, 2017).

2.3.2.1 Gadolinia study

In a study by (Galahom, 2016) on the effects of different BA's (particularly BA's Gd_2O_3 and Er_2O_3) on the neutronics of a Westinghouse PWR fuel assembly, burnup calculations investigating the behaviour of K_{eff} during burnup were ran in MCNPX. In one case the change in K_{eff} with burnup of UO_2 fuel of 3.6 a/o enrichment containing 2% Gd_2O_3 was investigated and it was found that K_{eff} starts very low and increases almost exponentially with burnup to a maximum K_{eff} , then there's a tipping point where K_{eff} eventually steadily decreases with burnup almost linearly- This is illustrated in Figure 2-4. K_{eff} starts off very low because Gd_2O_3 is such a strong neutron absorber due to the two Gd isotopes ^{155}Gd and ^{157}Gd . As Gd_2O_3 absorbs neutrons and slowly depletes, K_{eff} increases slowly at the same rate that Gd_2O_3 depletes. The tipping point as seen in the figure corresponds to the point in burnup where Gd_2O_3 is nearly completely depleted and K_{eff} peaks. Just after the tipping point we can assume that all the Gd_2O_3 has completely depleted and K_{eff} decreases at the same rate that UO_2 depletes until complete subcriticality.

Similarly, in the same study UO_2 fuel of 3.6 a/o enrichment WoBA (without burnable absorbers) was simulated and the behaviour of the fuel's K_{eff} as a function of burnup can be seen in Figure 2-4. It can be seen that the change in K_{eff} of the 3.6 a/o enriched UO_2 Fuel model Behaves similar to the change in k_{∞} of the fuels simulated in the reference study by (Erlank, 2017) as seen in Figure 2-1.

By comparing the initial K_{eff} values of the 3.6 a/o enriched UO_2 with burnable absorbers (BAs) and without burnable absorbers (WoBAs) at BOC we can see just how much of an effect 2% of Gd_2O_3 can have on the initial k_{eff} value. In this particular case, the difference in reactivity between the fuel with BAs and WoBA is approximately 1.3×10^5 pcm which is quite a large reactivity. In a nutshell, this is a clear indication of how IBAs can drastically decrease the initial CBC of a reactor at BOC.

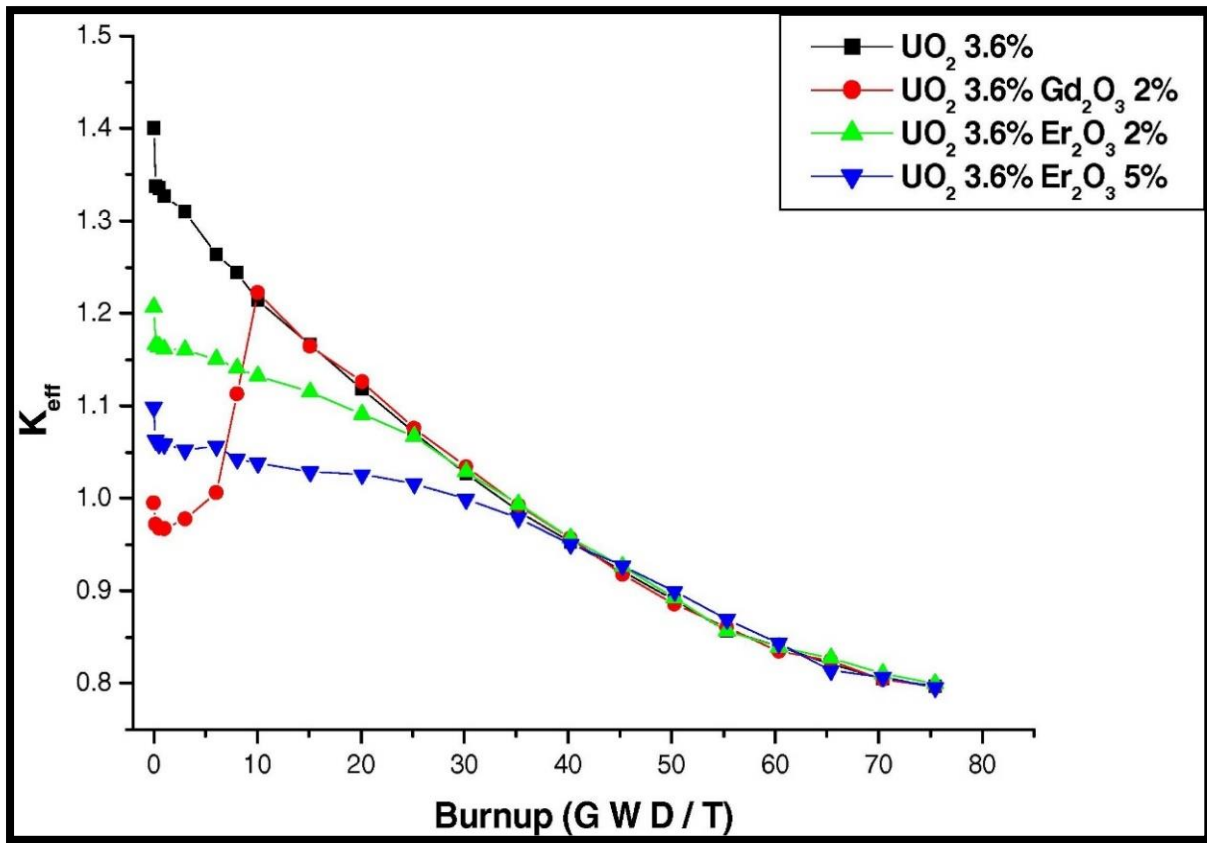


Figure 2-4 :Depiction of the change in k_{eff} with burnup of UO_2 fuel WoBA (black) and fuel with BA gadolinia (red) (Galahom, 2016).

2.3.2.2 Challenges with Gd_2O_3 in fuel

The general desire by utilities for PWRs is to optimize their fuel cycle length by prolonging it and also to decrease boric acid concentrations required for stable operation (Choe, et al., 2016). Optimizing the cycle length calls for increased heavy metal loading including increased fissile fuel enrichments, subsequently also calling for increased loadings of BAs. Since soluble boron has a limit and it is desirable for its concentrations to be as low as possible, it means that Gd_2O_3 loading in the fuel has to be increased to have enough leverage of negative reactivity. The problem with increasing IBA loading is that in LEU fuel for example, Gd_2O_3 in the fuel reduces the amount of UO_2 loading resulting in a reduced cycle length. Again, in UO_2 fuel Gd_2O_3 reduces the heat conductivity abilities which will result in overheating of the fuel (Campolina, et al., 2018). To avoid such, the enrichment of fuels containing Gd_2O_3 is limited to 2 a/o ^{235}U so that the amount of Gd_2O_3 is also limited (Choe, et al., 2016).

The influence of thorium, uranium and plutonium fuel mixture composition on the reactivity coefficients of pressurised water reactors

In the case of adding gadolinia to mixed oxide fuels, example Th/U or Pu/U fuels, it is quite a challenge to create a homogenous ternary mixture with such fuels (Bjork, 2012).

It is important to note that the use of gadolinia in this study is not for cycle length optimization but more for safety optimization in terms of reactivity. Nor does this study focus on the thermal conductivity implications of Gd_2O_3 in fuel, but more on Gd_2O_3 as a solution to bring down the CBC of each fuel model composition to avoid running into a positive moderator temperature coefficient problem.

The fuels by (Erlank, 2017) have high enrichments and therefore possess high positive reactivities at BOL. The fuels varying in composition, it is predicted that each will have different BA requirements in terms of Gd_2O_3 % and soluble boron concentration to add the necessary negative reactivity for criticality.

2.4 The neutron multiplication factor

The multiplication factor (k_{eff}) signifies the ratio of the number of neutrons born in one generation due to fission (in the reactor core) to the number of neutrons born in the preceding generation due to fission. When the ratio is equal to unity ($k_{eff}=1$) we say that, that particular reactor is critical because the fission reaction is steadily sustained, the number of neutrons in each generation remains constant and so is the energy produced (Lamarsh & Baratta, 2001).

In a real reactor core fission neutron can either interact with the materials inside the reactor and undergo various nuclear reactions upon interaction or can leak from the vicinity of the reactor core. A finite reactor core depicts a real core and is described by the effective multiplication factor (k_{eff}). An infinite reactor model assumes that no neutrons leak from the reactor core and the infinite multiplication factor (k_{inf}) is used to describe such a system (Lamarsh & Baratta, 2001).

In this particular mini-dissertation an infinite fuel pin is modelled and therefore the infinite multiplication factor (k_{inf}) is applicable.

2.4.1 Infinite multiplication factor (k_{inf})/ four factor formula decoded

The infinite multiplication factor (k_{inf}) is also known as the four-factor formula as it is defined by four parameters:

$$k_{\infty} = \eta f \epsilon p \quad (2.1)$$

Generally, the four parameters describe the multiplication of neutrons in an infinite reactor core and can be decoded as follows:

1. **The neutron yield factor (η_T)**- is the ratio of the number of fast neutrons from fission to thermal neutrons absorbed in the fuel.
2. **The thermal/fuel utilization factor(f)**- Is the ratio of the number of thermal neutrons absorbed in the fuel to the total number of neutrons absorbed in the system. In this case the reactor core refers to the fuel, moderator and burnable absorbers as these are the key materials in the fuel pin model to be simulated.
3. **The fast fission factor (ϵ)** – Is the ratio of the total number of neutrons produced by both fast and thermal fission to the number of neutrons produced by thermal fission.
4. **The resonance escape probability (p)** – Is the fraction of neutrons that manage to slow down to thermal energies “escaping the resonances” a region where neutron capture is highly probable in fuel atoms.

In a critical and yet stable reactor, a balance exists between the number of neutrons produced and neutrons lost through absorption (non-fissionable material and parasitically in fissionable material) and leakage - In an infinite model we can rule out leakage. An increase in temperature in the reactor core is triggered by the neutron population (n_T) inducing fission. If an increase in temperature increases the thermal neutron population n_T to a point where those produced exceed those being absorbed, the reactivity of the reactor core will increase meaning a positive TRC exists. A negative TRC will result in a reduced n_T with increasing temperature, thus an increase in temperature results in a loss of more thermal neutrons through absorption than those produced (Richards, 2012).

2.5 Reactivity coefficients

The temperature's effects on reactivity is an important factor to consider both for the operation of a reactor and importantly for the safety of a reactor. Many parameters that determine the criticality of a reactor are mostly dependant on temperature (Lamarsh & Baratta, 2001).

The amount of heat generated in a reactor largely depends on the number of induced fissions which can be traced back to the neutron population in the reactor. So logically the genesis of changes in temperature in an operating reactor is changes in the neutron population which further describes changes in reactivity (Stacey, 2007).

It is important to note that the components in the reactor are not in thermal equilibrium with each other and heat is transferred from the fuel to the moderator/coolant. The first and most important temperature reactivity coefficient is the doppler coefficient (DC) also known as the prompt temperature coefficient or the fuel temperature coefficient (Lamarsh & Baratta, 2001). Each of the names given to the Doppler Coefficient describe its specific characteristic behaviour to changes in temperature. Heat is generated in the fuel giving instantaneous rise to fuel temperature and influencing the reactivity of the fuel instantly. For that reason, the DC will be the first to respond to these changes in fuel temperature. When the temperature increases, based on its magnitude and sign (positive or negative) the DC will promptly lead to a power excursion (if positive) or promptly bring the reactor safely back to stability (if negative). An increase in fuel temperature is characterised by a phenomenon called Doppler broadening associated with the absorption of neutrons in the resonances, hence the name doppler temperature coefficient. This phenomenon is of crucial importance in nuclear reactor safety.

Heat generated in the fuel eventually disperses to the other components of the reactor coolant/moderator raising the temperature in these components. The moderator possesses a delayed temperature reactivity coefficient simply because it takes several seconds for the moderator temperature to rise after the fuel temperature increased (Stacey, 2007). The feedback mechanism of moderator is given by the value and sign of the MTC characterised by change in moderator density and the moderator to fuel ratio. An over moderated core will possess a positive MTC with increase in temperature while an under moderated core possesses the desired negative MTC which brings stability in the reactor (Nuclear-power.net, 2018). However, a positive MTC is less dangerous because unlike the DC its feedback is delayed. In most cases the prompt response of a negative DC brings the reactor to stability quicker than heat could reach the moderator and raise the moderator's temperature to a potentially adverse value for the positive MTC to kick in.

The influence of thorium, uranium and plutonium fuel mixture composition on the reactivity coefficients of pressurised water reactors

The reactivity coefficients of interest in this study are the DC and the MTC. Maintaining criticality and adequate safety in the operation of a reactor is of most importance and inherent safety as provided by the negative TRC's is of first priority.

Reactivity (ρ) can be described as the measure of the deviation of a reactor from criticality ($k=1$) during operation. Reactivity can be altered by a number of different factors, by factors that are introduced in the reactor by external mechanical means and those that occur spontaneously with time. Absorber rods and chemical shim are examples of factors that influence reactivity from being mechanically introduced into a reactor. Those that alter the reactivity with time can be referred to as "Time dependant factors" and these include changes with burn-up such as the neutron flux and the build-up of fission products particularly neutron poisons (Lamarsh & Baratta, 2001). The SI unit for reactivity (ρ) is percent mill (pcm) where $1 \text{ pcm} = 10^{-5}$.

$$\rho = \frac{k_{eff} - 1}{k_{eff}} \quad (2.2)$$

- If $K_{eff} > 1$ then $\rho > 0$ Supercritical reactor.
- If $K_{eff} < 1$ then $\rho < 0$ Subcritical reactor.
- If $K_{eff} = 1$ then $\rho = 0$ Critical reactor.

The **temperature reactivity coefficient/TRC (α_T)** is a quantity that describes the **reactivity feedback**- how the reactivity of the fuel corresponds to changes in temperature. Mathematically it can be described as the partial change in reactivity over the partial change in temperature expressed as follows (Stacey, 2007):

$$\alpha_T \cong \frac{\partial \rho}{\partial T} \quad (2.3)$$

Substituting the definition of ρ and simplifying:

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$$\alpha_T \cong \frac{1}{K} \frac{\partial \rho}{\partial T} \quad (2.4)$$

The manner in which the reactivity of a reactor responds to changes in temperature is solely determined by whether the TRC of a reactor is positive ($+\alpha_T$) or negative ($-\alpha_T$) (Lamarsh & Baratta, 2001). A negative TRC (α_T) is desirable trait in a reactor because when the temperature of the core increases, it (the negative TRC) reduces the reactivity to retain stability (ENS,2017). A positive TRC would produce an opposite effect to the negative TRC and possibly lead the reactor into a power excursion. A negative TRC is required by the regulatory commission for licensing purposes (Tucker, et al., 2014). The only instance where a reactor is allowed to have a positive TRC is at the BOC during start-up. 5 pcm/*F is the maximum allowable positive TRC for a Westinghouse PWR at BOC (Tucker, et al., 2014).

Neglecting the structural components, the main components of a thermal reactor include the fuel, moderator and the coolant - in a LWR the moderator and the coolant are one and the same. Generally, heat is generated in the fuel and is transferred to the other two components (moderator and coolant). Each component is at a different temperature and responds differently to temperature changes and therefore each has a unique individual coefficient of reactivity (Richards, 2012). The TRC of a reactor is the algebraic sum of the reactivity coefficients of the fuel and the moderator. The DC and the MTC are the individual reactivity coefficients of the fuel and the moderator respectively. To meet standards set by the licensing commission, the algebraic sum of all reactivity coefficients must give a negative value to ensure stability during reactor operation. Typical TRC values of a PWR are (-5 to -30 pcm/*C) (Nyalunga, 2016).

Structural components have an expansion coefficient so does the fuel, the moderator and the cladding (Edenius, 1976). The expansion coefficient is not important in this study.

2.5.1 DC/ Prompt reactivity coefficient/Doppler coefficient

Consider a monochromatic beam of neutrons approaching a group of hot nuclei (hot fuel atoms). The hot nuclei are highly energetic and therefore vibrate or move randomly at different kinetic energies and therefore different speeds. From a point of relative motion, the group of hot nuclei with different kinetic energies will perceive the monochromatic beam of neutrons as having a wide range of energies though all the neutrons in the beam have the same energy. The hotter the nuclei become with increasing fuel temperature the wider the range of energies perceived by the

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nuclei becomes. This is rather a complex phenomenon, but it explains the broadening of the resonance peaks of fertile fuel atoms with increasing fuel temperature. Since fertile material makes up a large portion of the fuel, the DC of most thermal reactors is negative ($DC < 0$) owing to the phenomenon described above which happens to be dominant in fertile fuel atoms (Lamarsh & Baratta, 2001).

Absorption of neutrons in the resonances of fertile isotopes U-238, Th-232 and Pu-240 usually occurs at epithermal neutron energies (Stacey, 2007). When the temperature of the fuel increases the consequence is that the reactivity of the reactor increases too. But due to the negative DC imbedded in the fertile isotopes, the shape of the resonances changes by becoming lower at the peak and broader at the base as seen in figure below, a phenomenon known as “the nuclear Doppler effect”.

When the resonances become broader and lower it simply means that the probability of absorption in the resonances is increased and resonance absorption slightly extends into the thermal spectrum absorbing thermal neutrons too. Since a large percentage of the fuel is fertile this robs the thermal neutrons away from fissile isotopes therefore decreasing the number of induced fissions reducing the reactivity in the core to retain criticality. Though the shape of the resonances changes the area under the peak remains the same. Linking the aforementioned to the four factor formulae, it is evident that the total DC is based on the resonance escape probability (Stacey, 2007). In a nutshell, an increase in fuel temperature decreases the number of neutrons reaching thermal energies as most of the neutrons end up captured in the resonances (Richards, 2012). Typically, the DC of a PWR is in the range (-4.9; -2.4 pcm/k) (IAEA, 2003).

2.5.1.1 Factors affecting the doppler coefficient (DC)

Fuel expansion

The fuel expands when fuel temperature increases due to its thermal expansion coefficient. The expansion increases the fuel volume which logically reduces the fuel density. This effect directly affects the reactivity by decreasing resonance absorption which adds a positive reactivity. On the other hand, the reduced fuel density may decrease the thermal utilization factor.

An increase in fuel temperature may harden the neutron spectrum. The thermal neutrons are therefore reduced meaning less interactions take place at thermal energies with material that have cross-sections with a good $1/v$ dependence. This effect decreases the reactivity (Stacey, 2007).

Change in resonance absorption

In UOX fuel there's a change in neutron resonance absorption which arises when ^{238}U transmutes to Pu-240. Pu-240 has quite a significant absorption cross-section in the thermal range. Changes in burnup have the biggest effect on the DC solely because of the changes in fuel composition such as transmutations of certain fuel isotopes and the build-up of fission products during the burnup cycle (O. Safarzadeh, 2015).

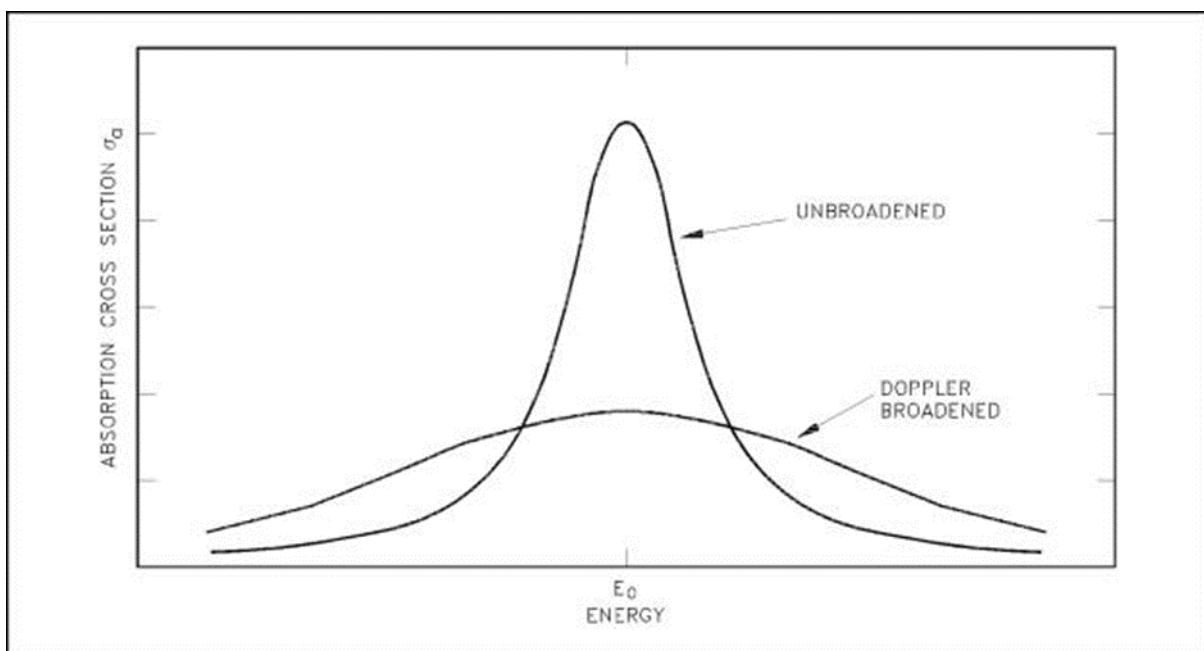


Figure 2-5: Doppler broadening of fertile isotopes.

The DC may also be referred to as the Doppler coefficient because of the nuclear doppler effect. It may also be referred to as the prompt reactivity coefficient simply because it responds instantaneously to any reactivity changes associated with increasing temperature (Stacey, 2007).

2.5.1.2 Mosteller benchmark (Doppler coefficient in a fuel pin cell)

The Mosteller benchmark solely investigates the doppler coefficient of reactivity (DC) in a PWR infinite fuel pin cell with reflective boundaries, a depiction of the model is given in

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Figure 2-6. The infinite fuel pin cells are from various PWRs based on optimized fuel assembly designs (Mosteller, 2007).

The doppler coefficient calculations are carried out at the BOC with 1400 ppm soluble boron concentration at two fuel temperature conditions, HZP (600k) and HFP (900k) (Jo & Kim, 2015). A doppler feedback change of 1000 pcm has been found between the reactivity at HZP and at HFP conditions (Mosteller, 2007).

Initially the benchmark study looked into UO₂ fuels of different U-235 enrichments and nearly a decade ago started looking into MOX fuels of Pu(RWR) and of weapon grade plutonium in PWR. A requirement by the benchmark is that the fuel being studied should be pure in composition and should not contain actinides and minor actinides arising from fission nor should it contain any other impurities. UO₂ should be pure with isotopes U-235, U-238 and U-234 for uranium. The MOX fuel should be pure and only contain PUO₂ and UO₂ with the isotopes Pu-239, Pu-240, Pu-241 and Pu-242 for plutonium. Another requirement by the benchmark is for the cladding material to be pure zirconium instead of zircalloy (Mosteller, 2007). However, the requirements of the benchmark are mostly minor requirement which are idealizations. But according to the benchmark, these idealizations should not have any major influences on the doppler coefficients as they've mainly been put in place for the simplification of calculations.

The doppler coefficient between HZP and HFP is determined as follows:

$$\alpha_{DC} = \frac{\left(\frac{k_{HFP} - k_{HZP}}{k_{HFP} k_{HZP}} \right)}{\Delta T_f} \quad (2.5)$$

Formula 2.6 below is used to estimate doppler coefficients of lattices which have been made about critical and it gives more realistic approximation of the net neutron leakage spectrum according to a Mosteller based benchmark study (De Kruijf, 1995). This is because it incorporates the buckling correction which considers the transverse net neutron leakage spectrum rate - spectrum between adjacent regions of the core.

$$\alpha_{DC}^B = 2 \frac{(k_{HFP} - k_{HZP})}{(k_{HFP} + k_{HZP}) \Delta T_f} \quad (2.6)$$

For an infinite lattice, **Formula 2.6** gives a much better approximation of the DC than **Formula 2.5** when the calculated kinf value is not about equal to unity (k=1) (De Kruijf, 1995).

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To produce a conservative model for the benchmark a 10% uncertainty is approximated for the Doppler coefficients in LWRs. This means a standard deviation of approximately 10% for the DC values calculated (Mosteller, 2007).

Table 2-1 to **Table 2-3** gives the conditions and requirements of the benchmark and the specifications of the fuel pin cell model used and the soluble boron, courtesy of (Mosteller, 2007).

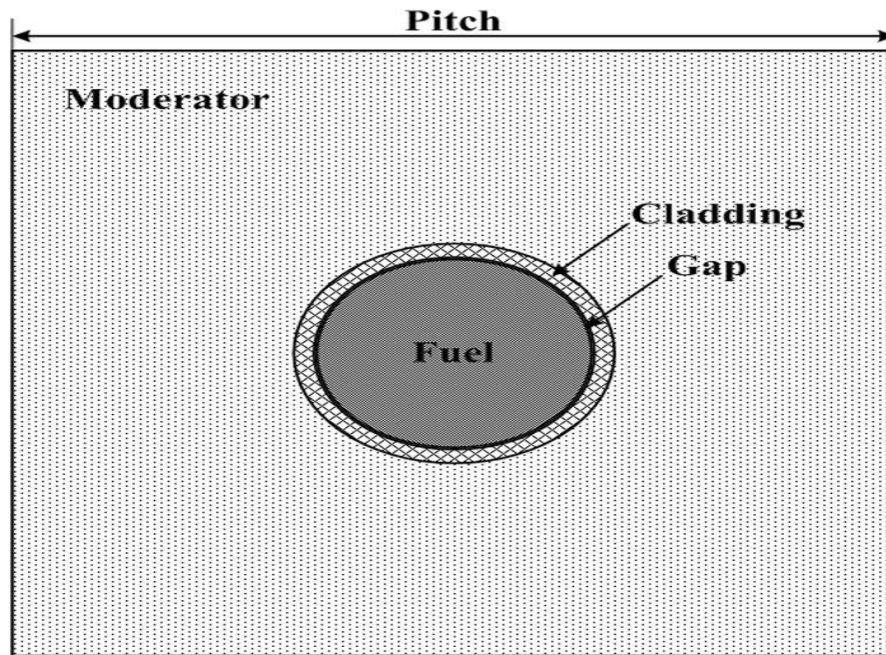


Figure 2-6: Model of a unit cell fuel pin in a square control volume.

Table 2-1: PWR Fuel pin specifications.

Parameter	HZP	HFP
Radius of fuel [cm]	0.39398	0.39433
Cladding inner radius [cm]	0.40226	0.40226
Cladding outer radius [cm]	0.45972	0.45972
Cladding thickness	0.05746	0.05746
Cell Pitch [cm]	1.26678	1.26678

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Table 2-2: 1400ppm Borated water at 600K.

Isotope	Number density [atoms/b-cm]	Atom fraction
H	4.42326E-02	6.66E-01
O	2.21163E-02	3.33E-01
10B	1.02133E-05	1.54E-04
11B	4.11098E-05	6.19E-04

Table 2-3: Zirconium cladding number density.

Material	Number density [atoms/b-cm]
Pure zirconium cladding	4.21838E-02

2.5.2 Moderator temperature coefficient (MTC)

The PWR is water moderated and changes in moderator density due to changes in temperature has a direct effect on reactivity and this is important to consider for the TRC of a reactor core (Edenius, 1976). In essence water becomes less dense with increasing temperature. With the moderator being a crucial component in thermal reactors and the density of the moderator playing a key role in the moderation process, the slightest change in density can change the entire state of the core considering neutrons are uncharged subatomic particles. The number of thermal neutrons will decrease because of the lack of moderation causing a large group of neutrons to remain in the fast region of the spectrum (shift of spectrum) and with neutrons remaining at such high energies more will easily leak from the vicinity of the core considering scattering and absorption with the water molecules is reduced (Edenius, 1976). Though a change in moderator temperature does change the reactivity of a reactor by changing thermal cross-sections, the change in moderator density with temperature has a more significant effect on the reactivity (Lamarsh & Baratta, 2001).

In terms of the four-factor formula, the factor that influences the MTC greatly is the resonance escape probability. This is due to the spectral shift to the epithermal region where resonance absorption dominates and therefore reducing the number of neutrons reaching the thermal region. The nuclides that makeup the fuel all have different resonance structures all of which affect the

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MTC quite differently because the MTC is very sensitive to fuel isotopic composition (Morrison, et al., 2018).

Since thermal reactors are designed for thermal fission with the decrease in the number of thermal neutrons in the core and increase in neutron leakage, the reactivity in the core will eventually decrease. Subsequently the reactor power and temperature will decrease. In many ways this explains the decrease in reactivity due to decrease in moderator density as a result of increase in the core's temperature.

The above mentioned describes a negative reactivity feedback mechanism provided by the moderator having a negative MTC ($MTC < 0$). A negative MTC is quite vital in PWR's and the reactor must be designed in such a way that the MTC is negative to avoid an adverse power excursion (Nyalunga, 2016). According to the IAEA the MTC in a PWR should range from -70pcm/°C to 0pcm/°C (IAEA, 2003). The MTC can be estimated using the formula below from (Jitka Zakova, 2008):

$$\alpha_m \cong \frac{\partial \rho}{\partial T_m} \quad (2.7)$$

Considering an infinite core, the MTC will be a function of the resonance escape probability and thermal utilisation factor (Nuclear-power.net, 2018). The fast fission factor can depend on moderator density, but according to (Lamarsh & Baratta, 2001), the fast fission factor can be taken to be independent of temperature.

2.5.2.1 Factors affecting the MTC

Below are the factors mainly affecting the MTC according to a study by (Zainuddin, et al., 2016).

2.5.2.1.1 Thermal and resonance absorption

Due to decreased moderation, fewer fission neutrons will be thermalized thus more of the neutrons will remain in the fast and epithermal regions. In the epithermal region the resonance absorption of neutrons is highly probable thus more neutrons will be lost through resonance capture and the overall effect will yield a decreased k_{∞} .

In the case of the fuel model compositions investigated in this dissertation which may include mixtures of plutonium, the spectral shift of neutrons to epithermal energies may decrease the reactivity or increase it. This uncertainty arises considering that the common fissile isotopes of

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plutonium Pu-239 and Pu-241 have a fission peak between 0.2eV-0.4eV which can add a positive reactivity to the system (Erlank, 2017). In a study by (Bjork, 2012) where Th-MOX fuels with various plutonium contents were the subject, it was found that fuels with a higher content of plutonium had a less negative MTC.

2.5.2.1.2 Hydrogen absorption (parasitic absorption):

Not all reactions of neutrons with the hydrogen atoms in water molecules are of scattering nature. According to (Lamarsh & Baratta, 2001) the scattering cross-section in hydrogen is constant up to 10keV and at all neutron energies radiative capture is $1/v$ noting that hydrogen isotopes exhibit no resonances. In the epithermal and fast regions the radiative capture cross-section of hydrogen is much lower than in the thermal region and thus captures in these regions are insignificant, therefore these captures increase strongly with increasing moderation. In most cases neutrons are lost through radiative capture in hydrogen atoms at standard reactor conditions with a standard moderator density. Once the moderator density changes with increase in temperature a counter effect can be observed (reduced radiative capture of neutrons) due to a decrease in moderator density. This effect adds a positive reactivity thus increase the value of k_{∞} (Zainuddin, et al., 2016).

2.5.2.1.3 Burnable absorbers

Being a parasitic absorber of neutrons to hold down the excess reactivity at BOC and to balance the reactivity throughout the fuel life cycle, any change in moderator density due to temperature will have an effect on the absorption as boric acid is uniformly concentrated in the moderator. As moderator density decreases so will the neutron absorption in boron, this will further unbalance the reactivity in the core making it more positive and resulting in an increased k_{∞} . In essence soluble boron has the biggest effect on the MTC, and more emphasis should be put on the investigation of the effects (Zainuddin, et al., 2016).

Burnable absorbers embedded in the fuel are highly effective at absorbing thermal neutrons. The neutron spectrum shifts to higher energies when the moderator temperature increases therefore moving neutrons away from the absorption vicinity of BAs. This adds a positive reactivity in the core (Zainuddin, et al., 2016).

2.5.3 Void reactivity coefficient (VRC)

A void can be described as the fraction of a volume of liquid coolant that has turned into vapour due to boiling (Lamarsh & Baratta, 2001). In many studies on LWRs the coolant void is mainly associated with the BWR and much less so with PWRs. One of the reasons this is, is that the liquid coolant in a PWR is subject to a high pressure (15.5 MPa) to prevent it from boiling. However, under circumstances where voids do form in PWRs. Voids in a PWR are merely a result of nucleate boiling. When the fuel rod surface temperature gets too hot there comes a point where the coolant in contact with it reaches its saturation point and begins to boil forming bubbles of vapor around the fuel rod (Lamarsh & Baratta, 2001). Due to the high pressure in the core (15.5MPa), the liquid coolant will boil at very high temperatures (350°C/ 623K) and because of adequate coolant flow in the core the vapour bubbles should collapse in time meaning this local nucleate boiling poses no serious threat to normal reactor operation (Nuclear-power.net, 2018).

In a PWR the moderator is the coolant and vice versa, so when voids form it means the density of the liquid coolant is reduced and this will have the same effect as the decrease in moderator density due to temperature increase (Nuclear-power.net, 2018). Similar to the MTC, an increase in void fraction should decrease the reactivity provided that the soluble boron content in the moderator/coolant isn't too large and within allowable limits (Morrison, et al., 2018).

On one hand, if the boiling persists as a result of coolant flow interruptions in the primary loop for instance, then the void fraction becomes significant this will have an impact on the reactivity of the system. The effect of the void on reactivity is described by the void reactivity coefficient VRC (Lamarsh & Baratta, 2001).

The voids are represented by a void fraction (x) defined in the first paragraph of this section. Any change in the void fraction (x) directly alters the average density of the coolant/moderator. In that sense as written in (Lamarsh & Baratta, 2001), the density of the liquid/vapour mixture as function of void fraction is:

$$\rho_d = (1 - x)\rho_l \quad (2.8)$$

Where ρ_d and ρ_l represent the density of the liquid/vapor mixture and the density of the liquid coolant/moderator respectively.

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The VRC is given by:

$$\alpha_v = \frac{d\rho}{dx} \quad (2.9)$$

It is important to note that voids rarely occur in PWR's (unless under accidental conditions) if they do they're very small that they're almost negligible- In a PWR voids are about 0.5%. Typical values of the VRC in a PWR are in the range of -100pcm/%void (Nuclear-power.net, 2018). And in case a core becomes fully voided, the FRV (Fully voided reactivity coefficient) becomes relevant and typical values of the FVR are in the range -1000pcm/%void (Morrison, et al., 2018).

It is important to note that in a PWR, the VRC is related to the MTC simply because changes in the void fraction means there's a change in the coolant/moderator density. The density of water is very temperature sensitive and the effect thereof cannot be ignored when calculating the MTC. So as x increases the moderator density decreases meaning less moderation and this effect should bring the reactivity down. This sequence describes the effect of a negative VRC (Lamarsh & Baratta, 2001). The VRC is related to the moderator to fuel ratio because the formation of voids decreases this ratio when the voids formed partially displace the moderator reducing it in that sense. Voids harden the neutron spectrum shifting it to the epithermal range where resonance absorption is dominant. This decreases the resonance escape probability (Nuclear-power.net, 2018).

Though the VRC and MTC are mostly similar, there are slight differences between the two. In essence, the perturbation of the density for the VRC is far greater than the perturbation of density for the MTC. Therefore, the neutron spectrum of the VRC is very different from that of the MTC in the sense that the spectrum of the VRC is much harder. This is because the VRC only comes into play under boiling where the coolant/moderator is almost completely vapour (Morrison, et al., 2018). In terms of fuel isotopic composition, for a boiling situation where the void fraction is about 0%, a harder spectrum is observed in Pu-MOX fuels than in UOX or Th-based fuels. This is because the spectrum under a large void is completely away from thermal energies, 1eV (in particular) where Pu-240 dominates in capturing neutrons (Foad & Takeda, 2015). With respect to burn-up, the VRC becomes more negative while the MTC becomes less negative with burn-up particularly in Pu-MOX fuels (Morrison, et al., 2018). A situation where the core is fully voided but not scrammed is very unlikely to occur (Morrison, et al., 2018).

3 METHODOLOGY AND THEORY

3.1 Reference PWR core

The PWR core referenced in this study is the South African owned, but French built Koeberg NPP reactor core. All its operational parameters, core geometry, material specifications, refuelling procedures and cycles were gathered from studies by (Erlank, 2017) and (Du Toit, 2013). From the gathered information we can summarise the Koeberg reactor core as 2785 MW thermal core with 157 fuel assemblies arranged in a 17x17 array each consisting of 264 fuel rods. The reactor utilizes UO_2 fuel with various enrichments ranging from 2.5- 4.5a/o%% U-235 enrichment at the axial blanket and centre respectively. 12 out of the 264 fuel elements contain 2.5wt% gadolinia to compensate for excess reactivity at BOL, also IBA ZrB_2 is embedded in some of the fuel pins. Furthermore, the reactor has a mean discharge burnup of 51 GWd/TU, 1/3 refuelling fraction and 12-18 months cycle (Du Toit, 2013). Tables (**Table 3-1, Table 3-2 & Table 3-2**) below gives design parameters which are important in this study.

Table 3-1: Fuel rod specifications.

Unit cell fuel rod	
Parameter	Value
Pitch [cm]	1.26
Fuel material	UO2
Enrichment [wt%]	2.5- 4.5 a/o% (UO2)
Fuel radius [cm]	0.4096
Cladding material	Zircalloy-4
Cladding outside radius [cm]	0.4750
Cladding thickness [cm]	0.0572
Cladding density [g/cc]	6.55
He gap thickness [cm]	0.0082

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Table 3-2: Assembly specifications.

Core	
Parameter	
Fuel model Assemblies in core	157
Assembly array	17 x 17 lattice
Fuel rods/assembly	264
Gd₂O₃ rods/assembly	12
Gd₂O₃/rod	2.5wt%
ZrB₃ rods/ assembly	~
Guide tubes/assembly (absorber rods)	24

Table 3-3: Core operating conditions.

Operating parameters (HFP)	Value & unit
Thermal power	2785 MW _{th}
Reactor pressure	15.5 MPa
Power density	102.8 kW/l
Moderator temperature	600 K
Moderator density	0.661 g/cc
Fuel pellet temperature	900 K
He temperature	600 K
Cladding temperature	600 K

3.2 Model and material description

An infinite unit cell fuel pin model in a square control volume was used. The model can be described as a square based rectangular prism with a cylindrical fuel pin right in the centre (Jo & Kim, 2015). Infinite models make use of reflective boundary conditions which enable them to simplify complex geometries into smaller and less complex geometries (Erlank, 2017). The material specifications, the geometry and the fuel pellet isotopic compositions is the input data needed to model a fuel pin (Uwaba, et al., 2014).

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3.2.1 Model materials and functions

Top view of the unit cell fuel pin in a square control volume is depicted in **Figure 3-1**. The fuel pin consists of a fuel pellet in the centre with zircaloy-4 fuel cladding around it. Between the cladding and the fuel pellet is a helium gap ring which gives provision for fuel pellet expansion and for heat transfer purposes. The dimensions of the fuel pin in the control volume are portrayed in **Figure 3-2** and also given in **Table 3-3**. These dimensions were adopted from a Koeberg size fuel pin.

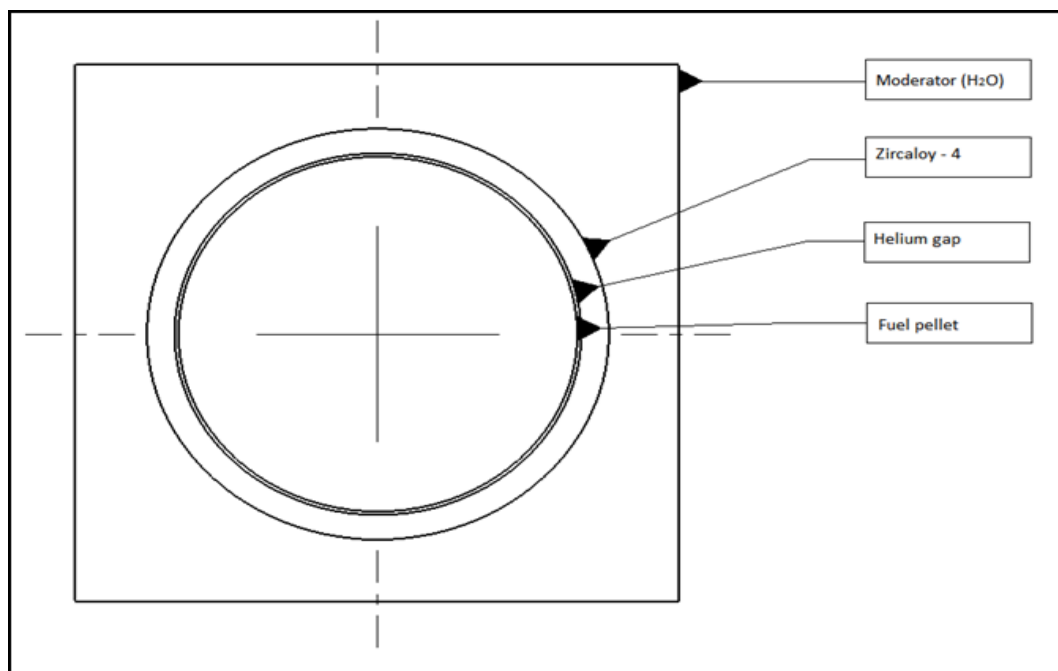


Figure 3-1: Material breakdown of the materials used for the simulated model.

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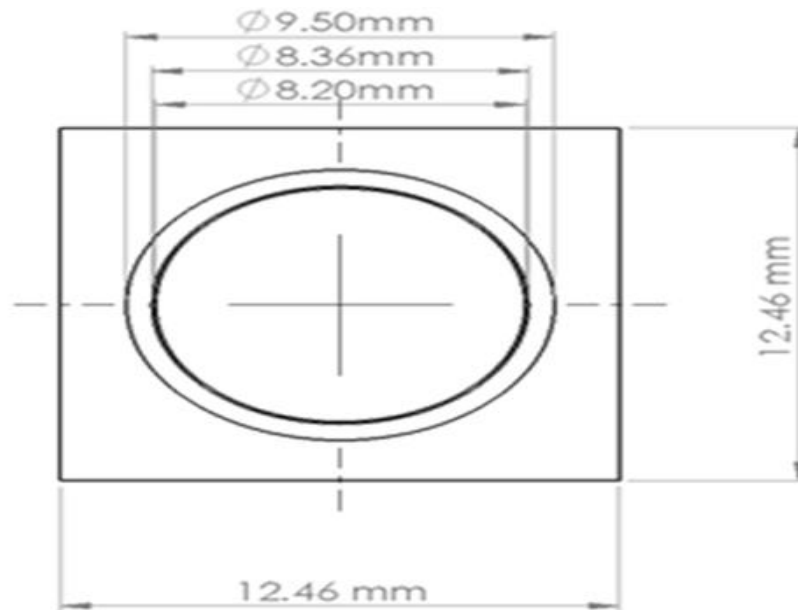


Figure 3-2: Top view of the square base rectangular prism model showing geometry specs of a Koeberg size fuel pin.

Table 3-4: Model geometry specifications.

Unit cell fuel pin	
Parameter	Value
Height [cm]	2.54
Pitch [cm]	1.26
Fuel radius [cm]	0.4096
Cladding outside radius [cm]	0.4750
Cladding thickness [cm]	0.0572
Cladding density [g/cc]	6.55
He gap thickness [cm]	0.0082

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3.2.1.1 Fuel pellet

Three fuel models are used, and their fuel pellets contain the following isotopic compositions:

- **Fuel model A:** (4.5% ^{235}U , 95.5% ^{238}U) O_2 (Koeberg)- reference fuel model
- **Fuel mode B:** (5% ^{235}U , 93% ^{238}U & 2% ^{232}Th) O_2
- **Fuel model:** (0.8% Pu(PWR), 4.5276% ^{235}U & 94.6724% ^{238}U) O_2

In some instances, burnable absorber Gd_2O_3 is included in the fuel model compositions.

3.2.1.2 Fuel model Cladding (Zircalloy-4)

The metallic fuel cladding/canning is one of the primary safety barriers for the retention of fission products. Furthermore, it serves as a medium for the transfer of fission generated heat from the fuel pellets to the coolant to produce steam. Zircalloy-4 is an alloy and contains the following metals, Zirconium (Zr – 97.56-98.27%), Tin (Sn- 1.20-1.70%), Iron and chromium (Fe+Cr- 0.28-0.37%) and pure Iron (Fe- w.18-0.24) (AZoM, 2018). Metals possess good thermal and mechanical properties for the mentioned applications - fission product retention and thermal conduction. In terms of neutronic properties, Zircalloy-4 has a very low absorption cross-section and a significant scattering cross-section. The low absorption is a desired neutronic property in fuel cladding because it is desired that more neutrons are absorbed in the fuel.

3.2.1.3 Moderator/coolant:

The moderator simulated is pure H_2O . The moderator is an important component of the model. It is necessary for slowing down neutrons to thermal energies where the desired interactions with the fuel nuclei are highly probable. The moderator is pure liquid (H_2O) with a density of 1g.cm^{-3} (at CZP, 293.6K). Liquids have quite a dynamic behaviour especially in an environment that is characterised by constant change in temperature (Stacey, 2007). Overall the moderator is an important variable which influences the type of neutron- nuclei interactions in the outcome as it is responsible for any shift in the neutron energy spectrum. Neutrons can lose their energy both through elastic and inelastic scattering reactions.

3.2.1.4 Quarter sized fuel pin model

Due to the nature of an infinite fuel pin with reflective boundaries, it is possible to reduce the computational time by simulating a smaller sized portion of the model. (Erlank, 2017) reduced the computational time by simulating a quarter sized fuel pin model. The reduced model is expected to drastically reduce computational time on the burnup calculations thus being more efficient in that regard.

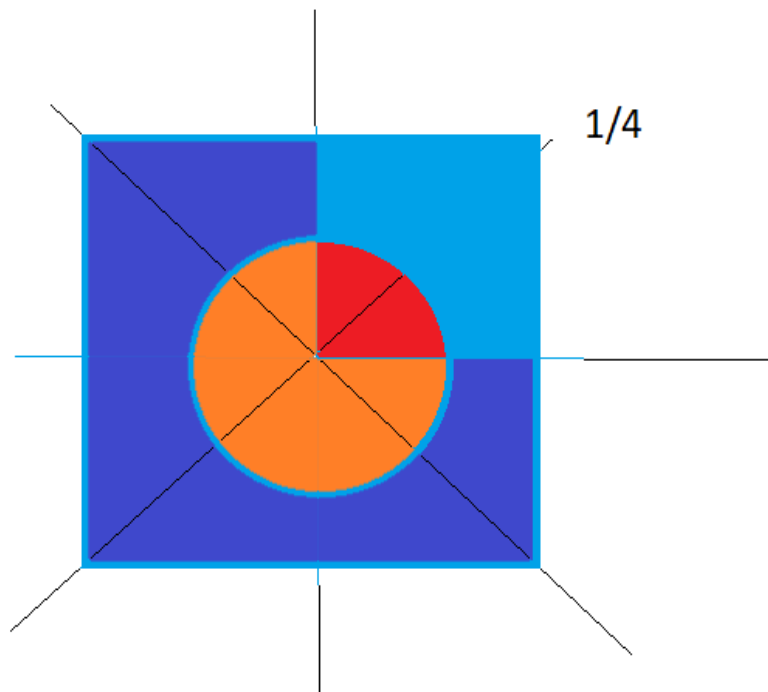


Figure 3-3: A depiction of a quarter portion infinite fuel pin model with reflective boundaries.

3.3 The use of numerical codes

The numerical code used in this mini-dissertation to model a fuel pin and carry out calculations on is Monte Carlo N Particle (MCNP 6.2). This is a code normally used to analyse the transport of neutrons and other sub-atomic particles including gamma ray photons (Shultis & Faw, 2008). This is a transport “Monte Carlo method” based code.

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Monte Carlo method is statistical and uses random sampling of probability distributions to simulate the random nature of neutron-nuclei interaction. The cross-section data of nuclei represent average probabilities of reactions to expect in the outcome. The Monte Carlo approach generates random values and calculates results from the generated random values many times over. Just like in physical reality the simulation creates millions of random particle interactions. While each interaction is completely random, the sum total of these interactions mimics the actual behaviour of the physical system (Shultis & Faw, 2008). Since each interaction is random, the sum total of a subset of interactions, e.g. the total number of fissions of U-235 in the fuel pin, will display a random statistical variation around an average value. Normally this be a Normal distribution, i.e. if we have a total of N interactions, the standard deviation around N will be $\sqrt{N}$. The fractional standard deviation thus $= \frac{\sqrt{N}}{N} = 1/\sqrt{N}$. Therefore, the larger N , the smaller the % error. This means that the more computation time is used, the more accurate the results will become, which always result in a trade-off between accuracy and computation time.

MCNP is capable of computing different types of geometries, resonances and conditions. Computational time usually depends on the size of the model at hand. For example, computational time for a full core is much longer than that of a fuel assembly and much more than that of a fuel pin. However, irrespective of the size of the model, computational time may be quite long and that's to produce converged reliable results (Pelowitz, et al., 2014).

To exploit the use of symmetry in order to simplify calculations on the actual model and to reduce computational time, a quarter of the fuel pin model is used. The model has reflective boundaries which resemble an infinitely large system with an endless number of similar fuel pins in all directions forming what we may call an infinite reactor core - calculations on an infinite model neglects neutron leakage as the simulated model does not have an outer boundary (Erlank, 2017).

Experimenting on an actual nuclear core is time consuming, it is expensive, and it is often dangerous because we are dealing with highly radioactive material. So numerical methods have made the design and modelling of nuclear systems more convenient.

3.3.1 KCODE (criticality calculations)

The KCODE is a general function in MCNP for nuclear reactor core calculations which involve criticality calculations. This function or card (in MCNP terms) is only for neutron problems and normally neutrons resulting from fission. In the card a criticality source is specified, then its

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neutron multiplication factor k_{eff} is generated (Pelowitz, et al., 2014). KCODE generates multiple values of k_{eff} for as many iterations as the user wants then gives the final value of k_{eff} which is the average of all iterations.

The criticality source specifications of the various KCODE calculations performed herewith involved 10 000-15000 neutrons for each cycle, 200-500 inactive cycles and 400-3500 active cycles.

3.3.2 Burnup/depletion

During burnup the isotopic composition of a fuel changes, so by the end of a cycle the fuel has a much different isotopic composition compared to its initial composition. The fuel model composition at the end of a cycle is mostly characterised by fission products, actinides and a lower enrichment of fissile U-235. In essence burnup measures the depletion and changes of the fuel and also the amount of heat energy produced per unit mass of fuel material (Du Toit, 2017).

Burnup/depletion calculations make use of MCNP and CINDER90. While MCNP focuses on steady state flux calculations and reaction rates, CINDER90 uses these rates to do depletion calculations of the nuclides involved. The CINDER90 library contains information such as decay chains of nuclide, the fission yields and the 63 group cross-section data. When CINDER90 generates new data arising from the depletion of material such as new number densities, MCNP extracts this new information and uses it to generate new fluxes and reaction rates (Pelowitz, et al., 2014). In this way the simulation iterates between MCNP and CINDER90.

The burnup specifications for the simulations at hand include constant power of 1.122×10^{-4} MW/cm³ and the atom fraction below which the fuel isotopes are no longer tracked in the calculations is 1×10^{-20} . The CINDER90 transmutation chain convergence criteria is 1×10^{-20} .

3.3.3 Customized cross-section and thermal scattering data

The temperature reactivity coefficients are evaluated at new temperatures which require new cross-section data as well as new alpha/beta thermal scattering $S(\alpha, \beta)$ data not given in the MCNP data libraries. The $S(\alpha, \beta)$ data is only available for a number of materials and temperatures (Goorlely, et al., 2013). To obtain new cross-section data corresponding to the new temperatures the *makxsf* code is used.

Given in Table 3-5 are the existing temperature dependant cross-section libraries for materials and existing thermal scattering data for uranium oxide and light water respectively. The new

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temperature dependant cross-section data and thermal scattering data libraries are generated by makksf and interpolated between temperatures given.

Table 3-5: Temperature corresponding ZAID identifiers for neutron cross-section libraries. The first table represents cross-section, the second table alpha,beta scattering in

ENDF-B VII.1 (cross-section identifiers)	Temperature (K)
.80c	293.6
.81c	600
.82c	900
.83c	1200

ENDF-B VII.0sab (alpha,beta scattering in UO ₂ identifiers for specified temperatures)		
O ₂ in UO ₂	U in UO ₂	Temperature (K)
o2/u.15t	u/o2.15t	800
o2/u.16t	u/o2.16t	1000
o2/u.17t	u/o2.17t	1200

ENDF-B VII.0sab (α,β scattering identifiers for specified temperatures)	
H ₂ in light water (H ₂ O)	Temperature (K)
lwtr.13t	300
lwtr.16t	600
lwtr.17t	650
lwtr.18t	800

3.3.3.1 Makxsf (make cross-section files) code

The makxsf code is a basic tool in MCNP that incorporates several operations from programmes NJOY [2] and DOPPLER [3] to create customized nuclide data libraries. This provides the option to create temperature dependant libraries which come in very handy for applications such as in temperature sensitivity studies (Brown, 2017).

In *makxsf* code operations behind the creation of customized nuclide data libraries involve interpolating unresolved resonance probability tables and $S(\alpha, \beta)$ thermal scattering data to new temperatures. The code also has the ability to extrapolate resolved resonance data to higher temperatures through doppler broadening (Brown, 2017).

In basic terms the *makxsf* (make cross-sections file) takes existing *xsdirs* (cross-section directories) and existing cross-section libraries and then creates new *xsdirs* and new cross-section libraries by interpolating between these. The *xmdir* is a file which locates and reads ZAID data from cross-section library files.

In this study the new cross-sections are interpolated at temperatures between 900K and 1200K (in steps of 50K) for the fuel material (including Gd) and between 500K and 620K (in steps of 20K) for the moderator material (including boron). Both the unresolved resonance data as well as the $S(\alpha, \beta)$ thermal scattering data at new temperatures are obtained through interpolation between the specified temperatures.

3.3.3.2 Free-gas model vs $S(\alpha, \beta)$ model

MCNP gives the option of using TMP (temperatures) cards to modify cross-sections by the free gas treatment of low energy neutrons and this option can be used together with Pseudo materials when working with new temperatures (Du Toit, 2017). This option though it more convenient it is only effective down to 4 eV where the $S(\alpha, \beta)$ thermal scattering kernels are effective at even lower neutron energies (Goorlely, et al., 2013). So not considering the $S(\alpha, \beta)$ thermal scattering data may make the results inaccurate in certain cases especially in water moderated systems where a bulk of the neutrons lurk around thermal energies. The $S(\alpha, \beta)$ thermal scattering is mostly effective in the thermal neutron region (below 2eV), where it perceives materials as molecular compounds (Du Toit, 2017).

Both the free-gas model and the $S(\alpha, \beta)$ model consider scattering and collision kinematics at certain nuclides and temperatures. However, the free-gas model assumes that the medium is a free-gas where the $S(\alpha, \beta)$ treats materials as molecules and crystalline solids (Goorlely, et al.,

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2013). Again, the free-gas model is effective only down to 4 eV where the $S(\alpha, \beta)$ is effective below 2 eV neutron energies.

The results of the free-gas treatment model with Pseudo materials and of the makxsf code with new cross-section and $S(\alpha, \beta)$ data are compared to observe the effects each have on the values of k_{inf} . KCODE INP (input) files that contain pseudo-materials and use the free-gas treatment of thermal neutrons are compiled and on the other hand KCODE INP files that contain new cross-section and $S(\alpha, \beta)$ data are compiled.

Formula 3.1 is the free-gas treatment formulae used to convert the thermal temperature of a cell into energy (MeV). The free-gas model specifies the relative energy between neutron-nuclei at a specific temperature. In this study the formula is only applicable the fuel pellet cell (fuel temperature) and the moderator cell (moderator temperature).

$$tmp = 8.617 \times 10^{-11} * T_{new} (k) (MeV) \quad (3.1)$$

Formula 3.1, formula 3.2 and formula 3.3 below are for pseudo- materials and are used to find the weighted fractions of a material at temperatures between two existing cross-sections. This formula is used to avoid using the makxsf code because there's no need to generate new cross-section data when using it (Du Toit, 2017). T_{new} is the new temperature we wish to perform calculation at for a material. T_{xs1} and T_{xs2} are temperatures of the lower and higher existing cross-sections respectively.

$$f_1 = \frac{\sqrt{T_{new}} - \sqrt{T_{xs2}}}{\sqrt{T_{xs1}} - \sqrt{T_{xs2}}} \quad (3.2)$$

$$f_2 = 1 - f_1 \quad (3.3)$$

3.3.4 FMESH card (Tallied reaction rates)

The FMESH card is a special function in MCNP which can be used to calculate reaction rates over a problem geometry. The function basically tracks the number of particles per unit area within a mesh cell - the default unit of measurement is particle/cm².

FMESH mesh tallies can be used in conjunction with FM tally multipliers. This collaboration between FM tally multipliers and FMESH mesh tallies is a special case which allows the user to calculate isotopic reaction rates within cells. The particle flux is multiplied by the cross-section of the isotope then multiplied by the atom fraction of the isotope in a material to give the reaction rate in unit of total reactions/cm³/s instead of in the default units.

Furthermore, the FMESH tally function can be used with energy bins. The results (reaction rates) can also be tallied in energy bins to see how the reactions vary with energy in the various isotopes, therefore to show energy dependant reactions within different isotopes. The energy bins represent energy groups or energy windows of the particle flux.

3.4 Verification and validation method

Like with any experimental work in the science community, it is generally advisable and important to evaluate the accuracy of a measuring tool to ensure that the results produced are reliable. To determine the accuracy and reliability of MCNP 6.2 code as the measuring tool in this particular dissertation, fuel models similar to those from previous studies are simulated in the code and results obtained are compared to reference models - Readily calculated models from previous studies are referred to as reference models - Section 4.1 gives the verification process and results.

The fuel model compositions in the study *Increasing the Fuel Cycle length of a PWR by means of a homogenous uranium, thorium and plutonium fuel design* by Erlank, were simulated in MCNP 6.1 Beta which is the older version of the MCNP 6.2 code. The bulk of the present study is based on the fuel model compositions simulated in the aforementioned study by (Erlank, 2017), so to verify the results of the MCNP 6.2 models to be used in this study, it makes sense to select at least one fuel pin composition from the study by (Erlank, 2017) and use it as a reference model. The selected reference model is the fuel model composition titled “Koeberg 4.5 a/o reference” mentioned under **section 2.1**. This reference model is basically UO₂ fuel of LEU of 4.5%. Erlank performed a burnup calculation on the aforementioned reference model and to cut a burnup calculation’s computational time it is important to note that he simulated a quarter (¼) of a fuel pin cell with a square geometry.

In this study, the same quarter sized model of 4.5 % enriched UO₂ from Erlank’s study is simulated in MCNP 6.2 and similarly at HFP conditions a burnup calculation is performed. To ascertain the results generated from the burnup calculation in MCNP 6.2, the results obtained are compared to

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those obtained by Erlank for his reference model (Koeberg 4.5 % enriched UO_2 reference) which he simulated in MCNP 6.1 beta.

In the current study, a fuel pin model of 4.5 a/o enriched UOX fuel is simulated in MCNP 6.2 and similar to what Erlank did in his study a burnup calculation on the fuel pin is performed. The results of the calculation are compared based on the change in the infinite neutron multiplication factor with time. The comparison process is what evaluates the accuracy of MCNP 6.2 as a measuring tool and furthermore the accuracy of the designed fuel pin models.

The full-sized fuel pin model and the quarter sized model are geometrically similar with both being rectangular prisms so therefore having a square geometry when viewed from the top. To further verify the accuracy of MCNP 6.2 a full-sized pin model and a quarter-sized pin model both containing 4.5 % enriched UO_2 are separately simulated using the burnup function in MCNP 6.2 and then the burnup results are compared based on k_{inf} as a function of time.

3.5 Three batch “full core” concept and refuelling method

Table 3-6 : The three- batch fuel terminology.

Terminology	
Fresh/fresh burn	Fresh fuel, more reactive
Once burnt	Slightly depleted fuel less reactive
Twice burnt	More depleted fuel much less reactive

In a PWR refuelling takes place every 12 – 18 months depending on whether the fuel is reactive enough to maintain criticality and during refuelling a third of the core (a third of the fuel assemblies) is removed and replaced while the remaining two thirds is reshuffled in the core. A single fuel assembly can be burned for at least four years and since refuelling is every 12-18 months this means once loaded, fuel goes through at least three burnup cycles on average before being totally removed from the core (Nuclear-power.net, 2018).

Generally, in a full core there's fresh fuel assemblies, slightly depleted fuel assemblies (fuel being burned for the 2nd time) and more depleted fuel assemblies (fuel being burned for the 3rd time or) and all three types of fuel assemblies contribute to the overall reactivity of the full core. The terms Fresh, once burnt and twice burnt are also used to refer to fresh fuel assemblies, slightly depleted

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fuel assemblies (burning for the 2nd time) and more depleted fuel assemblies (burning for the 3rd time) respectively. **Figure 3-4** below depicts a quarter of a full core with Fuel model Assemblies of fresh, once burnt and twice burnt fuel (Fridman & Kliem, 2011).

In order to try and mimic a full core where three types of fuel assemblies are in place, one type of fuel model composition is burned three times using the unit cell fuel pin model described in **section 3.2**. Firstly, it is burned as fresh fuel to represent fresh fuel assemblies in their first cycle, then for the second time to represent fuel assemblies that are reshuffled and burned for the second cycle and then for the third time to represent fuel assemblies near depletion being burned for the third and last cycle before their total removal from the core.

Each fuel depending on composition has a specific cycle length and the refuelling and reshuffling process length may be the same for each fuel model composition type. Using the unit cell fuel pin model and the burnup function in MCNP, each fuel model composition is burned for the duration of its cycle at HFP, then burned at zero power to simulate the refuelling outage. The burnup process is repeated for the second time and then for the third time keeping in mind that the refuelling process is only at the end of the first burn and second burn.

To simulate the refuelling outage periods, the burnup calculations are executed at zero power for 37 days. In a study by the USNRC on the risk assessment of refuelling outage days for 16 PWRs between 2000 and 2013 it was found that the average refuelling outage days for the 16 PWRs is 37 days (Shackelford & Jones, 2015).

As mentioned, the average of the results of the three burnups simulated in the unit cell fuel pin model is taken and it assumed to represent the overall reactivity in a full core. Each fuel model composition is simulated in this manner initially without any burnable absorbers in the fuel or moderator. In terms of different enrichments found in a real full core, it is assumed here that a full core has only three enrichments. Namely, the enrichment of the fresh fuel, the once burnt fuel and the twice burnt fuel.

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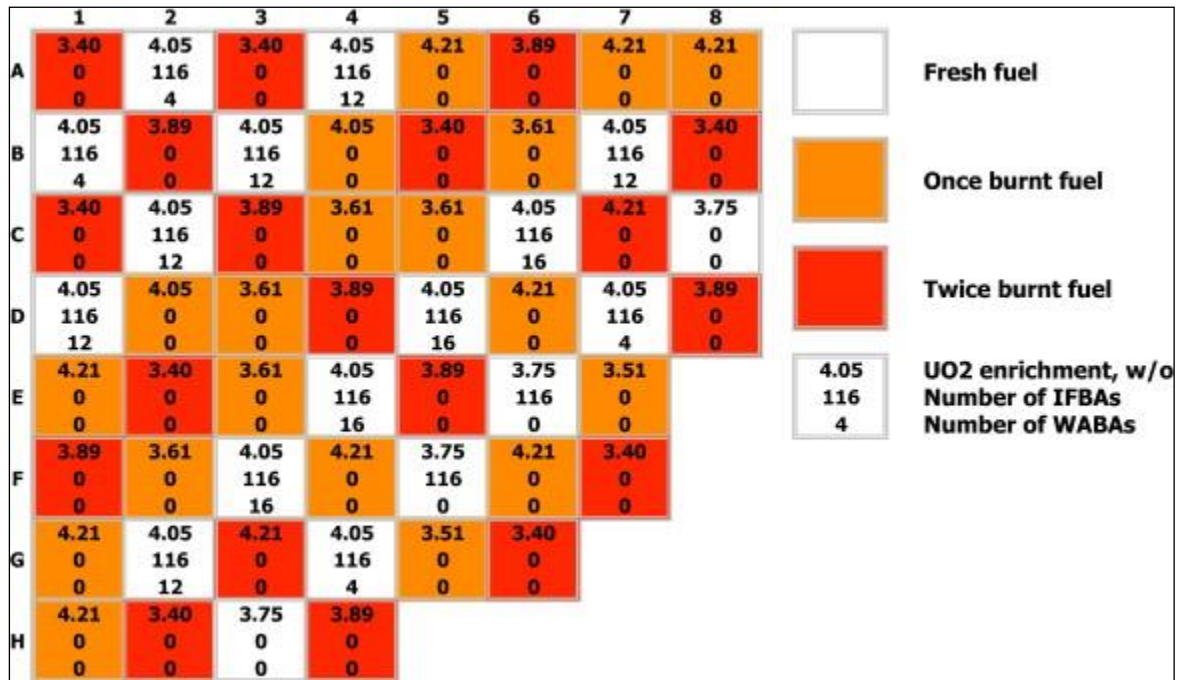


Figure 3-4 : $\frac{1}{4}$ of a full core UO₂ fuel showing fresh, once burnt and twice burnt Fuel Assemblies. courtesy of (Fridman & Kliem, 2011).

3.5.1 Lower operational limit and non-leakage probability

The lower operational limit can be described as the minimum infinite neutron multiplication factor (k_{inf}) value where fission is still sustainable for a finite reactor system. This value is assumed to be an equivalent to $k_{eff}=1$ for a finite reactor (Erlank, 2017). **Figure 3-5** below shows the lower operational limit used to estimate the cycle lengths of various fuel model compositions simulated in a fuel pin model by (Erlank, 2017).

The lower operational limit is used to estimate the non-leakage probability which can later be used to convert k_{inf} values to k_{eff} , therefore converting an infinite system to a finite system. The non-leakage probability of each fuel model composition is expected to be same meaning the lower operational limit value of each fuel model composition is the same. This is because the fuel model compositions are simulated at the same conditions (HFP) in the same model of the same size and geometry. The geometric buckling is a function of size and geometry of a reactor and leakage can be modelled as a function of buckling in the diffusion equation (Lamarsh & Baratta, 2001). The non-leakage probability (P_{NL}) is a function of the size of the reactor and other important design parameters - but largely depends on the size of the reactor (Stacey, 2007).

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Many studies that simulate infinite PWR reactor systems take the lower operational limit as 1.03 assuming 3% leakage (Du Toit, 2017). Typical non-leakage probability in a PWR is 0.97 (Stacey, 2007).

In the study by (Erlank, 2017) it was estimated that the lower operation limit for the simulated infinite fuel pins is where $k_{\infty} = 1.205$. This value of k_{∞} corresponds to where the finite models becomes subcritical and the fuel model compositions are thus no longer able to sustain a fission chain reaction. In a nutshell, $k_{\infty} = 1.205$ is assumed to be the same as $k_{\text{eff}} = 1$ for a finite reactor. This value ($k_{\infty} = 1.205$) was drawn using 4.5% ^{235}U Koeberg reactor fuel model composition type as the reference model. An infinite model of the 4.5% ^{235}U was simulated and its burnup k_{∞} calculated with time. Since the real Koeberg reactor is shut-down and refuelled every 16 months (approx. 480 days), k_{∞} for the infinite model after 16 months can be assumed to be the k_{∞} that corresponds to criticality ($k_{\text{eff}} = 1$) in a finite reactor (Erlank, 2017). A similar approach is undertaken in a study by (Van der Walt, 2016)

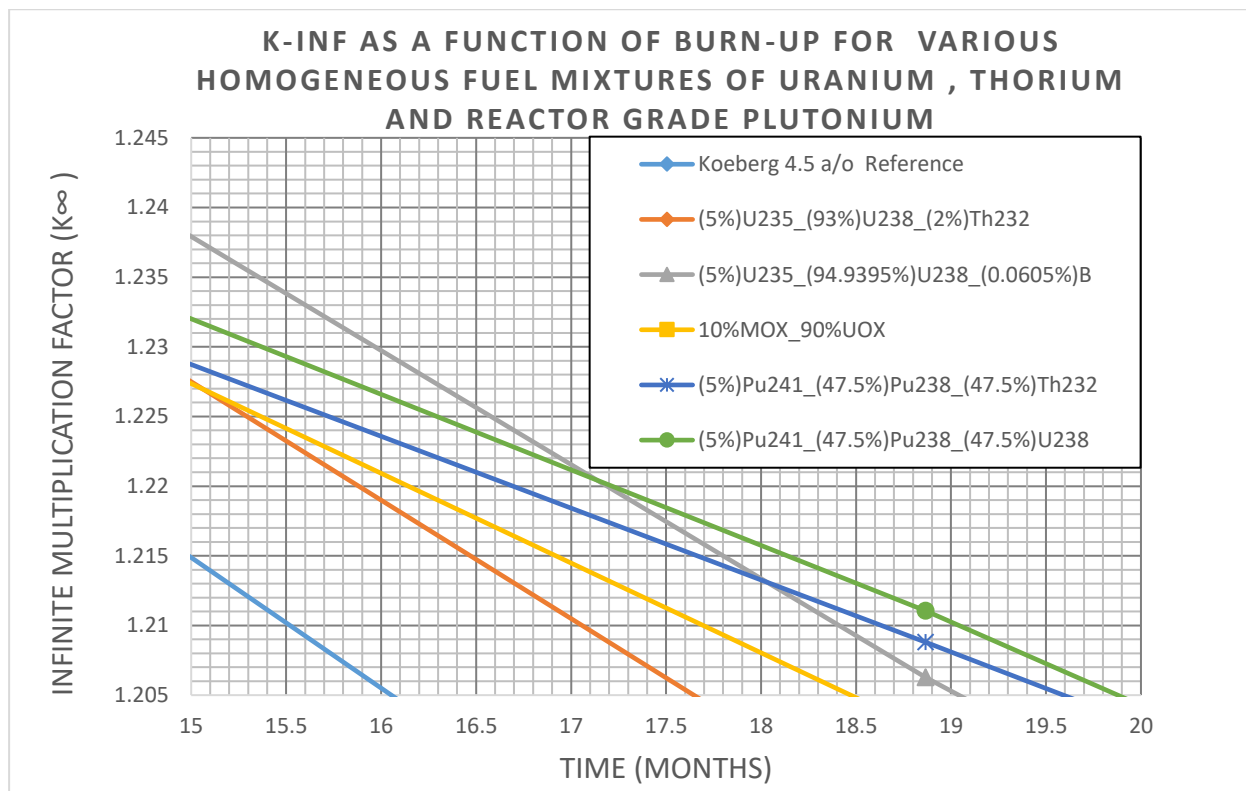


Figure 3-5: The lower operational limit of various fuel compositions simulated in the same geometric model courtesy of (Erlank, 2017).

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The non-leakage probability is the ratio of k_{eff} critical ($k_{eff(crit)} = 1$) and k_{∞} critical ($k_{inf(crit)} > 1$). In general, $k_{inf(crit)}$ is the lower operational limit of an infinite reactor system, corresponding to a point in time where the finite version of the reactor system is shut-down for refuelling.

$$P^{NL} = \frac{k_{eff(crit)}}{k_{\infty(crit)}} \quad (3.4)$$

k_{∞} is translated into k_{eff} by using the expression below (**formula3.5**) which contains the non-leakage probability (P^{NL}): k_{eff} depends on k_{∞} and P^{NL}

$$k_{eff} = K_{\infty} P^{NL} \quad (3.5)$$

The non-leakage probability described here assumes both thermal and fast leakage.

3.6 Critical boron concentration and critical gadolinia percentage

This section discusses the method explored to find how much soluble boron is required to give criticality to each of the fuel model compositions.

The idea is to find combinations of soluble boron in the moderator and gadolinia in the fuel that gives criticality ($k_{eff}=1.00$) for each of the fuel compositions of interest assuming they're in a full core as a combination of fresh, once burnt and twice burnt fuel assemblies. This is done keeping in mind that the maximum allowable soluble boron concentration in a PWR is 1500 ppm as stated by (Rui, et al., 2017) and that in 17x17 array full core, only a few fuel assemblies contain Gd₂O₃ and only 12 out of 264 fuel pins in such an assembly contain gadolinia (Bjork, 2012).

Since the amount of soluble boron and gadolinia mixed into the moderator and the fuel respectively, to compensate for the excess reactivity at BOC, solely depends on the fuel compositions, it is necessary to calculate the CBC of the different fuel model compositions because each fuel model composition has a different fissile enrichment and therefore different excess reactivity at BOC.

3.6.1 Gadolinia modelling

First it is necessary to scale down the gadolinia percentage to the unit cell fuel pin model since a full core isn't used in this study.

To scale down the gadolinia percentage to the infinite unit cell fuel pin model with the information gathered about the Koeberg reactor, **formula 3.6** is used. The formula considers that the reference core has a 17x17 lattice with 157 fuel assemblies with 264 rods per fuel assembly. 12 of the 264 rods contain gadolinia (Gd_2O_3) and the reactor has a 1/3rd batch refuelling fraction (Du Toit, 2013). Gadolinia can only be added in fresh fuel and it is assumed here that 1/3 of the core is fresh fuel with gadolinia.

$$Gd\%(model) = \frac{157 \times 12 \times 1}{157 \times 264 \times 3} \times Gd\%(actual) \quad (3.6)$$

Where $Gd\% (actual)$ is the gadolinia percentage used in an actual core, 2.5wt% Gd_2O_3 is used in the reference core (Koeberg PWR).

Though a quarter of an infinite unit fuel pin cell model is used for simulations in this study it is important to note reflective boundaries make it seem like it is infinitely large unit fuel pin cell model in all directions (Van der Walt, 2016). It's not necessary to scale down to the actual size of the fuel pin cell model in terms of its physical dimensions.

Gadolinia is only added at the BOC in the fresh fuel model and it is assumed that all the gadolinia burns away within a few months. This can be assumed because in reality burnable absorber gadolinia burns out quite rapidly to a point where it completely depletes at around the 10th month in the cycle of UOX fuel (Du Toit, 2013). In the thorium and the MOX fuel model compositions the gadolinia will have a distinct depletion rate from the UOX fuel model composition.

3.6.2 B-10 enriched soluble boron

To compensate for the absence of control rods and other burnable absorbers such as IFBA ZrB_2 which could not be simulated due to the limitations of the model used in this study, enriched soluble boron is used. By enriching boron, the yield of isotope B-10 which is the main neutron absorber in natural boron is increased. Naturally B-10 is 19.6 % of boron and in this instant the yield is doubled to 40 % B-10. This enrichment is adopted from a full core study on Th-MOX fuels in a PWR by (Fridman & Kliem, 2011).

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As for other IFBAs such as ZrB_2 their specific quantity in fuel assemblies is unknown and therefore this BA is not added in the unit fuel pin cell model.

3.6.3 Critical boron concentration method

The method explored involves the linear interpolation method used for estimations (Nyalunga, 2016). Soluble boron concentrations between 500ppm and 1500 ppm are randomly selected. The selection keeps in mind the maximum allowable soluble boron concentration in a PWR and it assumes that the minimum can't be below 500 ppm. An initial soluble boron concentration is guessed. Running a KCODE calculation with the guessed soluble boron concentration k_{eff} is obtained. By observing whether k_{eff} is subcritical ($k_{\text{eff}} < 1$) or supercritical ($k_{\text{eff}} > 1$) a second soluble boron concentration is guessed and with that a KCODE calculation is ran to find another k_{eff} value. Once the KCODE calculations yield a k_{eff} value that is slightly subcritical and another that is slightly supercritical then using the linear interpolation method the soluble boron concentrations that gives criticality ($k_{\text{eff}} = 1$) can be found. Because the model used here is infinite, the KCODE calculations yield k_{∞} and using the non-leakage probability factor calculated using **formula 3.4** in section 3.5.1, a theoretical k_{eff} of the system can be obtained. The non-leakage probability is found to be different for each fuel model compositions.

3.7 The use of perturbation method

The perturbation method based on the perturbation theory involves deviating a system from its normal state and then observing the effects of the deviations on how the system operates. In reactor analysis, perturbation is a convenient tool for calculating changes in reactivity with certain conditional changes in the system to estimate reactivity coefficients. In so doing, the perturbation method mimics the effects of changes during burn-up on the multiplication factor. This method is very useful in the sense that it can be used to study changes in reactivity when certain conditions change (temperature, density etc), however the method ignores the influence of changes in the neutron flux distribution caused by the changed conditions. The reactivity coefficients calculation method is based on the perturbation theory (Stacey, 2007).

3.7.1 Reactivity coefficients calculations method

The reactivity of the various fuel model compositions will be calculated. Each fuel design with different isotopic compositions will be simulated to determine the respective K^∞ . Then the reactivity of each fuel design will be determined analytically using the **formula 3.7**

$$\rho = \frac{k_{eff} - 1}{k_{eff}} \quad (3.7)$$

Reactivity can simply be described as the deviation of a reactor from criticality (when $k_{eff}=1$). Noting that an infinite fuel pin model with reflective boundaries is simulated in this study, k^∞ is expected from the MCNP burnup and KCODE calculations. The expression (**formula 3.8**) below is used to convert k_{inf} from the KCODE calculations to k_{eff} .

$$k_{eff} = k^\infty P_{NL} \quad (3.8)$$

The reactivity is calculated at BOC. Reactivity is strongly influenced by reactor conditions especially, those associated with temperature. Each of the designed fuel model compositions will have a different reactivity. The reactivity is dependent on a number of parameters. When certain conditions change and consequently change the reactivity, the degree of change in reactivity with these factors/parameters can be measured to determine the respective reactivity coefficient. In this particular case we investigate how the change in factors such as the core temperature and moderator density alter the reactivity.

$$\alpha(y) = \frac{\Delta \rho}{\Delta y} \quad (3.9)$$

α_y is the reactivity coefficient as function of parameter y , and ρ is the reactivity. Parameter y represents the fuel temperature (T_f) and the moderator temperature (T_m).

- ❖ The Doppler Coefficient, reactivity changes of each fuel model composition with step changes in fuel temperature (T_f) from room-temperature/ CZP (293.6k) through to HZP(600k), then at HFP (900K) and then at 1200K - All other materials (cladding, moderator etc) are kept constant at 600k. Changes in fuel density are insignificant as they're so small they can be neglected. Large temperature gaps for the Doppler coefficient can be taken as a 300k

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temperature difference is expected to produce a reactivity change close to ∓ 1000 pcm (Mosteller, 2007).

- ❖ The MTC, reactivity changes with step changes in moderator temperatures (T_m) given in **Table 3-7**. The changes in moderator density at each temperature step are considered as the change is significant.

All the other materials (fuel, cladding etc) are kept at 900k. The assumption here is that the fuel is already at (HFP-900k), then the moderator temperature slowly rises. In this study the changes in reactivity brought about the moderator temperature are observed from room-temperature/ CZP-293.6k through to LOCA-620k as heat is transferred from the fuel to the moderator. A large temperature step of 206.4k is taken from 293.6k to 500k, then from 500k to 620k temperature steps of 20k are taken.

It should be noted that the moderator density is very sensitive to temperature changes because of the large volumetric thermal expansion of water, which is largely due to nucleate boiling that takes place in PWRs (Elert, 2018). For this reason, changes in moderator density with temperature is considered. Given in **Table 3-7** is the moderator density corresponding to a specific temperature at a pressure of 155 bars.

Table 3-7: Moderator conditions at an operating pressure of 155bar.

Moderator temperature (k)	Moderator density(g/cc)
293.6 (room temperature)	1.003
500	0.843
520	0.816
540	0.787
560	0.753
580	0.712
600	0.661
620 (boiling)	0.098

The above described methodology is for KCODE calculation (10000 source neutrons, 200 inactive cycles and 500 active cycles) carried out at BOC, assuming each fuel model composition is in a full core, consisting of a combination of as fresh, once burnt and twice burnt fuel, with gadolinia only in the fresh fuel, but the critical soluble boron (CBC) used for all three fuel stages. This combination is mimicked by taking a fresh fuel pin and burning it all the way to the end of life of the twice burned fuel. This continuous history of k_{∞} is then chopped up into equal lengths corresponding to the fresh, once burnt and twice burnt stages. These three histories are then laid out alongside each other and averaged, so that the average k_{∞} at BOC is then the average of k_{∞} at BOC for the fresh, once burnt and twice burnt cycles respectively, etc.

3.7.2 Doppler coefficient for fresh fuel pins

The doppler coefficients of the various fuel model compositions are calculated based on the Mosteller benchmark. This benchmark solely investigates the doppler coefficient of reactivity (DC) in an infinite fuel pin cell, which is the model of choice in this study. The doppler coefficient calculations are carried out at the BOC with 1400 ppm soluble boron concentration at two fuel temperature conditions, HZP (600k) and HFP (900k) (Jo & Kim, 2015). Just as in the study by (Jo & Kim, 2015), KCODE calculations of 20000 neutrons, 500 inactive cycles and 3500 active cycles is set up. Conditions of the benchmark and the specifications of the fuel pin cell model used are given in tables in section 2.5.3, courtesy of (Jo & Kim, 2015). The doppler coefficients at each condition are estimated as follows:

$$\alpha_{DC} = \frac{\left(\frac{k_{HFP} - k_{HZP}}{k_{HFP} k_{HZP}} \right)}{\Delta T_f} \quad (3.10)$$

$$\alpha_{DC}^B = 2 \frac{(k_{HFP} - k_{HZP})}{(k_{HFP} + k_{HZP}) \Delta T_f} \quad (3.11)$$

Formula 3.10 is born from substituting **Formula 3.7** into **Formula 3.9** and expressing **Formula 3.9** as a function of fuel temperature (T_f). **Formula 3.11** is used to estimate the Doppler coefficients of lattices which have been made about critical and it gives a more realistic approximation of the net neutron leakage spectrum according to a Mosteller based benchmark

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study as it incorporates the buckling correction which considers the transverse net neutron leakage spectrum rate (spectrum between adjacent regions of the core) in a pin cell model. The buckling correction gives quite a realistic estimate of the net neutron leakage (De Kruijf, 1995).

3.7.3 MTC technical evaluation method

For a technical evaluation of the factors that contribute to the MTC feedback, MCNP type 4 FMESH tallies which give isotopic reaction rates (reactions/cm³) are used. These are used to evaluate the dominant reactions rates in the fuel and the moderator at three energy bins corresponding to the three energy spectrums (thermal, epithermal and fast). The MTC is very sensitive to fuel isotopic composition and therefore it is important to understand the effect of each predominant isotope on the MTC (Morrison, et al., 2018). **Table 3-8** gives the moderator conditions to be used in the simulation and **Table 3-9** gives the energy bins.

Table 3-8 : The three moderator conditions for the MTC technical analysis.

	Temperature	Density (g/cc) @155bars
CZP (Room temperature)	293.6	1.003
HZP/HFP (operating condition)	600	0.661
Boiling	620	0.098

Table 3-9: The energy bins.

Neutron energy spectrum	Energy range
Thermal	0-2eV
Epithermal	2.5 keV
Fast	10MeV

3.8 Limitations and challenges of the methods and models

3.8.1 Limitations and challenges of the model

One of the reasons why BA (burnable absorbers) are inserted in fuel assemblies in a PWR's is to decrease the initial concentration of soluble boron which could have an adverse effect on the MTC/VRC if concentrations are too high (Nuclear-power.net, 2018). So, as a precaution, the soluble boron concentration in PWRs is limited to 1500 ppm (Rui, et al., 2017). BAs are inserted in two forms in Fuel Assemblies, as IBAs (Integral burnable absorbers) and control rods/ BPRs (Burnable poison rod (Galahom, 2016)). In IBA form, a few of the UOX fuel pins (12 in a Koeberg fuel assembly) contain a mixture of BAs mainly gadolinia (Gd_2O_3) and zirconium diboride (ZrB_2) (Du Toit, 2013). These fuel pins containing IBAs are placed at various locations in the fuel assembly. Control rods which normally contain B_4C (Boron Carbide), or composites of Hafnium and Cadmium are movable and can be inserted or withdrawn from a fuel assembly via guide tubes situated at various locations in the fuel assembly. These two mechanisms of BAs (IBA and BPR) are specifically designed to hold down the initial high reactivity associated with fresh fuel pins at the BOC and importantly to keep the initial concentration of soluble boron at acceptable levels as mentioned.

Firstly, a portion of a fuel pin is used to represent a whole reactor core which normally contains complex geometry and contains many materials and structural components compared to a fuel pin. To simulate a full core with a three-batch refuelling scheme of fresh, once burnt and twice burnt fuels, at least a model resembling at least a $\frac{1}{4}$ of a full core is required since most cores are symmetrically packed.

Due to limitations of the model used in this study, control rods could not be included in the simulations. In order to include control rods in the simulations, a fuel assembly or at least a portion of a fuel assembly is required. In 17x17 lattice Koeberg PWR fuel assembly, 12 of the 264 LEU UOX fuel pins will contain IBAs Gadolinia (Gd_2O_3) and zirconium diboride (ZrB_2) (Du Toit, 2013). Gd_2O_3 is homogenously blended in the Fuel and ZrB_2 is a thin layer coated on the surface of the fuel pellet (Bjork, 2012). For the control rods, 24 guide tubes are in place for their insertion.

3.8.2 Limitations and challenges of the methods

Stochastic code

Because of the nature of the Monte Carlo method which is statistical and involves probability distributions it should be noted that uncertainties will arise and in return may cause the values of the calculated neutron multiplication factors (k_{eff}) to diverge (Rui, et al., 2017). Generally, the neutron-nuclei interactions (which are the main interactions considered in this study) involved in producing the neutron multiplication factor are drawn from statistical sampling technique. In every KCODE calculation there's an uncertainty attached, which is the reason why MCNP also gives the standard deviation for each and every neutron multiplication factor calculated. Again, MCNP is a stochastic code and challenges with such codes is the computing time (Nyalunga, 2016).

Interpolation method and model

The interpolation method for finding the CBC firstly assumes a linear relationship between the neutron multiplication factor and the highest and lowest guessed boron concentrations. The k_{eff} values corresponding with the guessed boron concentrations may have converged insufficiently thus cause huge deviations in the results of the interpolation method (Rui, et al., 2017).

If more accurate results are to be found using this method, firstly the number of cycles in the KCODE calculations will have to be increased. And secondly the number of iterations with the boron concentration that give a k_{eff} value that is close to unity will have to be increased to determine the exact relationship between k_{eff} and boron concentration. Even if the relationship is linear, but with more iterations a more converged value may be obtained.

Gadolinia and ternary fuel mixtures

In the case of adding gadolinia to mixed oxide fuels (e.g Th/U or Pu/U fuels), it is quite a challenge to create a homogenous ternary mixture with such fuels (Bjork, 2012). A ternary mixture is a mixture achieved from a blend of three different materials.

It is important to note that the use of gadolinia in this study is not for cycle length optimization but more for safety optimization in terms of lowering initial reactivity. Nor does this study focus on the thermal conductivity implications of Gd_2O_3 in fuel, but more on Gd_2O_3 as a solution to lower the excess reactivity of the fuel compositions at BOL and to bring down the CBC of each fuel composition to avoid running into a positive moderator temperature coefficient problem.

The fuels by (Erlank, 2017) have relatively high enrichments and therefore possess high positive reactivities at BOL. The fuels vary in composition and it is predicted that each will have different

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requirements in terms of soluble boron concentration to add the necessary negative reactivity for criticality.

4 RESULTS AND DISCUSSION

4.1 Model verification

In the following verification process firstly two versions of MCNP are compared, version 6.1 beta and version 6.2. Secondly an eighth fuel pin model is compared to a quarter of a fuel pin model of the same fuel isotopic composition. The simulated models here for data verification include an infinite fuel pin model and its quarter size described in section 3.2. **Figure 3-2** and **Figure 3-3** respectively depict the two models. Both models are composed of fuel pellets of UOX fuel of LEU of 4.5 a/o enrichment. These two models will be referred to as verification models in the proceeding of this study.

The reference burnup data for the two simulated models is data from a published study by (Erlank, 2017). The data was drawn from a model he titled “Koeberg 4.5 a/o reference” in his study which by description is an infinite fuel pin model comprised of UOX of 4.5 a/o enrichment. This described model is referred to as the reference model and it is a replica of the South African Koeberg reactor standard fuel pin without burnable absorbers. It is important to note that the burnup data referenced herewith from (Erlank, 2017) study comes from a quarter (1/4) sized infinite fuel pin reference model simulated in MCNP 6.1 beta coupled with ENDF/B-VII.1. Because a different version simulation code was used in the study by (Erlank, 2017), this allows to verify MCNP 6.2 as a newer version of the code by comparing results obtained by MCNP 6.2 with those obtained by (Erlank, 2017) where version 6.1 beta is used.

The burnup data from the two verification models simulated is compared to the existing burnup data of the reference model. The models are verified by comparing the value(s) of the infinite multiplication factor (k^∞) during burnup at three points in the burnup cycle, at BOC (beginning of cycle), MOC (middle of cycle) and EOC (end of cycle). The models are simulated under the Koeberg conditions given in **Table 4-2**. **Table 4-1** gives the geometry and material specifications of the South African Koeberg nuclear reactor fuel pin. The geometric specifications of the reference fuel pin and operating conditions of the reactor are important parameters for the modelling process.

Verifying the geometry of the models at hand is equally an important step in assuring that the desired shape and material configuration of a model is simulated. The geometry of the models is verified using MCNP X Visual editor (version X_24E) VISED. When constructing an MCNP

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INP file (input file) as a new user, it is quite easy for one to mess up the geometry. A visual tool such as VISED aids an MCNP user by ensuring that the INP file gives the desired model.

Discrepancies are expected between results obtained from MCNP 6.2 and the MCNP 6.1 reference mode. This is because MCNP is statistical in nature and involves using probability distributions as mentioned in **section 3.3**. As part of the verification process it is important to quantify these discrepancies and determine their magnitudes to make proper comparisons regarding accuracy.

The 4.5% reference model is an important model to have and reference mainly because part of the purpose of this study is to show changes in reactivity coefficients that can be expected in a PWR when UOX fuel of 4.5% enrichment is substituted with thorium and plutonium fuel mixtures.

Table 4-1: Core and fuel pin design parameters of the South African Koeberg PWR.

Parameter(s)	
Fuel rod radius	0.475 cm
Fuel pellet radius	0.4096 cm
Cladding thickness	0.0572 cm
He gap thickness	0.0082 cm
Pin pitch	1.26 cm
Fuel lattice	17x17
Fuel density	10.607 g/cm ³
Cladding density (Zr-4)	6.55 g/cm ³
Coolant density	0.661 g/cm ³ (H ₂ O)
Enrichment (235U)	4.5 at%

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Table 4-2: Operating parameters of the Koeberg PWR used in simulation.

	Conditions
Power density	102.8 kW/l (1.028×10^{-4} MW/cm ³)
Coolant temperature	600 K
Fuel pellet temperature	900 K
He temperature	600 K
Cladding temperature	600 K

4.1.1 MCNP 6.2 verification model v/s MCNP 6.1 reference model

The quarter infinite fuel pin model comprised of 4.5 a/o enriched UOX (1/4 verification model) simulated here in MCNP 6.2 is a direct duplicate of the 4.5 a/o enriched UOX ¼ infinite fuel pin model (reference model) simulated in MCNP 6.1 beta by (Erlank, 2017). A burnup calculation is performed on the ¼ varication model in MCNP 6.2 under the Koeberg reactor conditions given in **Table 4-1** . The burnup calculations are accompanied by KCODE calculations of 500 cycles and 10 000 neutrons per cycle. From the burnup calculation the changes in the infinite neutron multiplication factor (k_{∞}) over time a period of 25 months (computer time) at a constant power density is obtained.

The two models are verified based on the changes in the infinite neutron multiplication factor over time particularly at BOC, MOC and EOC to see the direct correlation of the values of k_{∞} . A close correlation between the k_{∞} value(s) of the verification model and the reference model indicates that the burnup data results generated by MCNP 6.2 are reliable and of good quality. Furthermore, this verifies that the infinite fuel pin model can be trusted to simulate other fuel model composition mixtures.

Figure 4-1 depicts the visual verification of the ¼ verification model's geometry and material configuration. **Figure 4-2** depicts and compares the change in the infinite neutron multiplication factor of the two models. Furthermore, the infinite neutron multiplication values of the two models are compared at various points in burnup in terms of the average percentage difference

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in **Table 4-3**. The average percentage difference between the infinite neutron multiplication factor values is expressed as follows.

$$average\ (\%)difference = \left(\frac{k_{\infty(ver)} - k_{\infty(ref)}}{\frac{k_{\infty(ver)} + k_{\infty(ref)}}{2}} \right) \times 100 \quad (4.1)$$

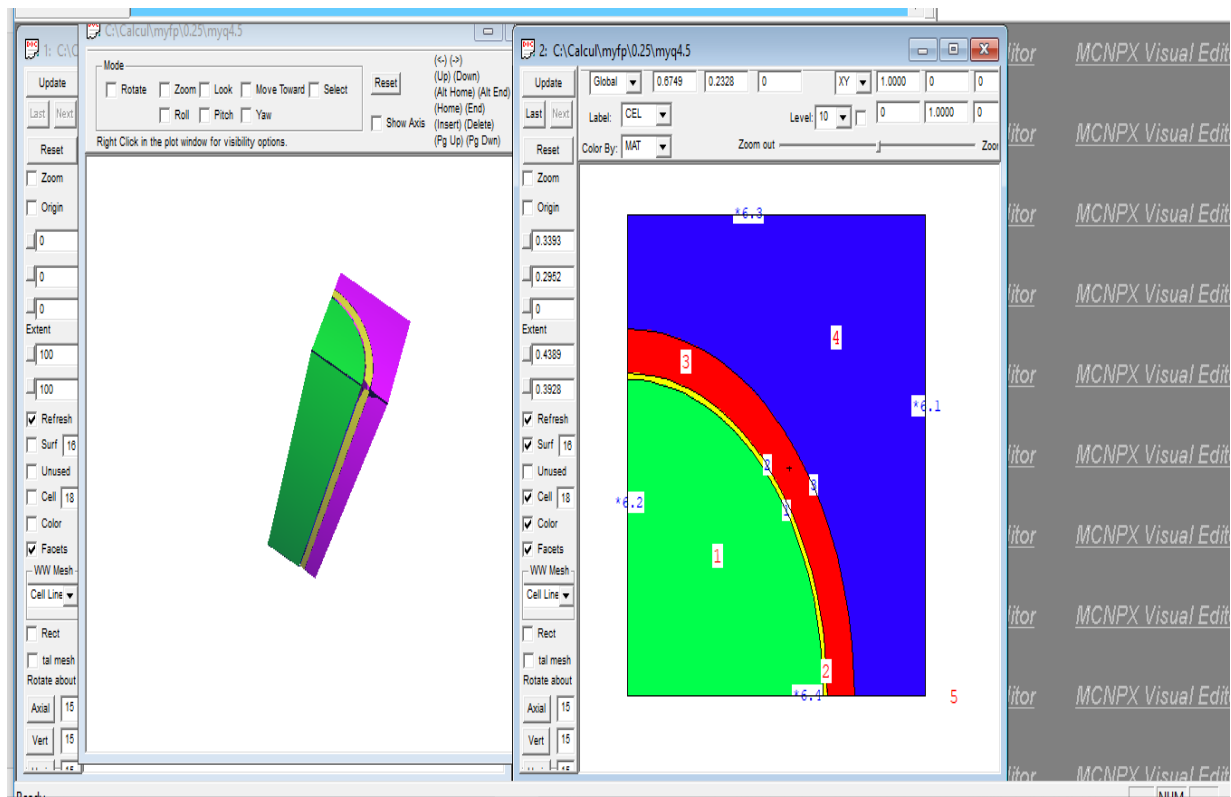


Figure 4-1: A 3D view and top view of the $\frac{1}{4}$ verification model of 4.5 a/o enriched UOX in VISED.

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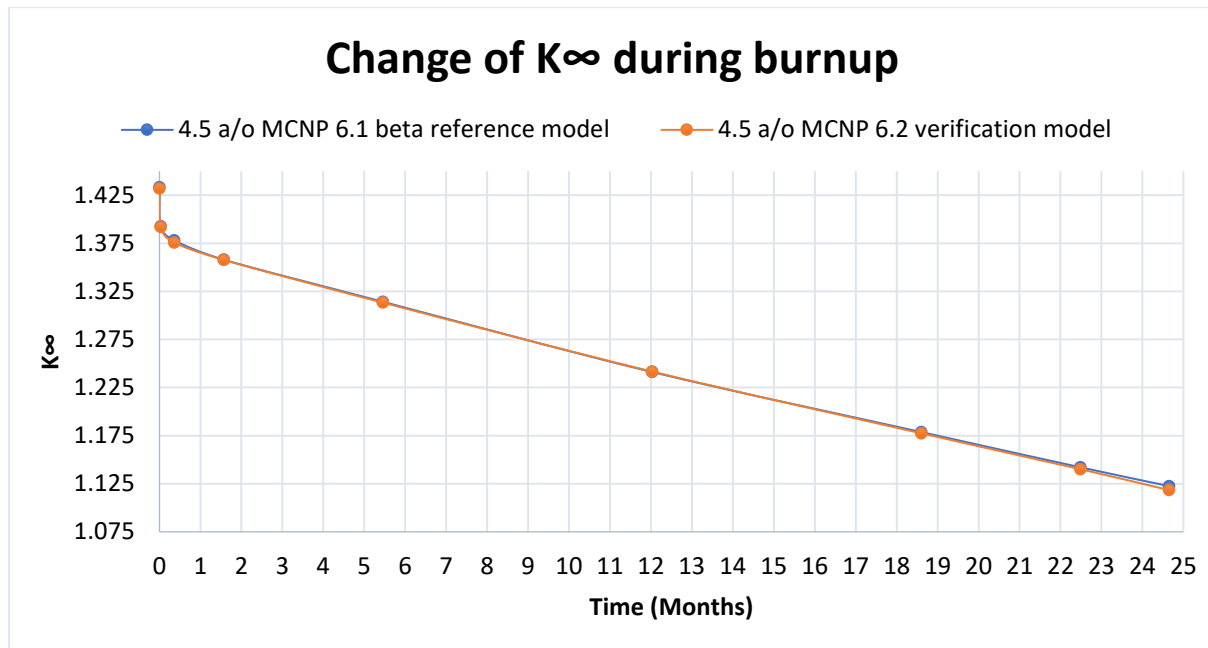


Figure 4-2: Graph comparing the change in k_{∞} with time of the $\frac{1}{4}$ MCNP 6.2 verification model and $\frac{1}{4}$ MCNP 6.1 beta reference model both of 4.5 a/o enriched UOX fuel.

Table 4-3: K_{∞} comparison at various points in burnup of the MCNP 6.2 verification model and the MCNP 6.1 beta reference model of same fuel model composition.

Point in burnup	K_{∞} (verification) (std dev)	K_{∞} (reference)	K_{∞} difference (pcm)	Average (%) difference
BOC	1.4318 (0.0018)	1.4332	140	-0.09773
MOC	1.2416 (0.0020)	1.2411	50	0.040279
EOC	1.1183 (0.0020)	1.1224	410	-0.36596
Average difference (pcm & %)			200	0.17

**** std dev (standard deviation)**

As seen in **Figure 4-2** the results highly correspond as it seems that each data point in the plot lies perfectly on top of the other. The average percentage difference values given in **Table 4-3** indicates that there's a very close correlation between k_{∞} values of both the reference model and verification model at the selected points in time during burnup. This very close correlation between the k_{∞} values highlights the accuracy of the MCNP 6.2 as a measuring tool and further verifies the quarter fuel pin model to be quite accurate. Furthermore, the average difference in K_{∞} values given (pcm) indicates that on average throughout the burnup cycle, the K_{∞} values differ on the third decimal.

There are no burnable absorbers present in both models which is why k_{∞} decreases at an average steady rate of 0.0144/month. The sharp drop in k_{∞} at the BOC is mainly the result of two negative reactivity effects (Stacey, 2007). Fissile fuel burns away rapidly while fission product poisons, mainly Xenon-135 starts to build-up a few hours after the initial fission reactions in the fuel. There's an initial higher burnup rate in fresh fuel, which means the rate at which fuel is burned and consequently the rate at which fission product poisons build up is higher. Reduction in fissile material reduces reactivity so does the build-up of high absorption cross-section fission product poisons. The two negative reactivity effects persist at a rapid rate at the very start as seen in **Figure 4-2** until the Xenon-135 concentration reaches its saturation, assuming there's a high neutron flux during burnup in the fuel models (Lamarsh & Baratta, 2001). As soon as the Xenon-135 reaches equilibrium, k_{∞} starts decreasing at a rather slower and steady rate.

4.1.2 ¼ sized MCNP 6.2 verification model vs full size pin model

In this section a ¼ portion MCNP 6.2 verification model described above in section 4.1.1 is compared to a full-size infinite fuel pin. The significance of this comparison is to verify if a quarter sized version is sufficient enough to be used as many studies actually use an infinite unit cell fuel pin model (Mosteller, 2007). Furthermore, from this comparison, the difference in computational time between the two models can be obtained, but the important comparison is the compatibility of the k_{∞} values of the two models. The two models are compared by running burnup calculations as in section 4.1.1 simulating the depletion of 4.5 enriched UO_2 fuel in both model sizes. The burnup card for both models were identical except for the geometry and the power - the full-size fuel pin is burned at 4 time the power of the ¼ model. The burnup file for both models is accompanied by a KCODE card of 10 000 neutrons, 200 inactive and 500 active cycles. A 12 core machine with 3.2 GHz processing speed and 24.5 GB RAM is used for running both models.

The influence of thorium, uranium and plutonium fuel mixture composition on the reactivity coefficients of pressurised water reactors

The quarter sized model was visually verified in section 4.1.1 as seen in **Figure 4-1**. **Figure 4-3** below depicts the full-size infinite fuel pin verification model's geometry and material configuration with the aid of VISED.

The influence of thorium, uranium and plutonium fuel mixture composition on the reactivity coefficients of pressurised water reactors

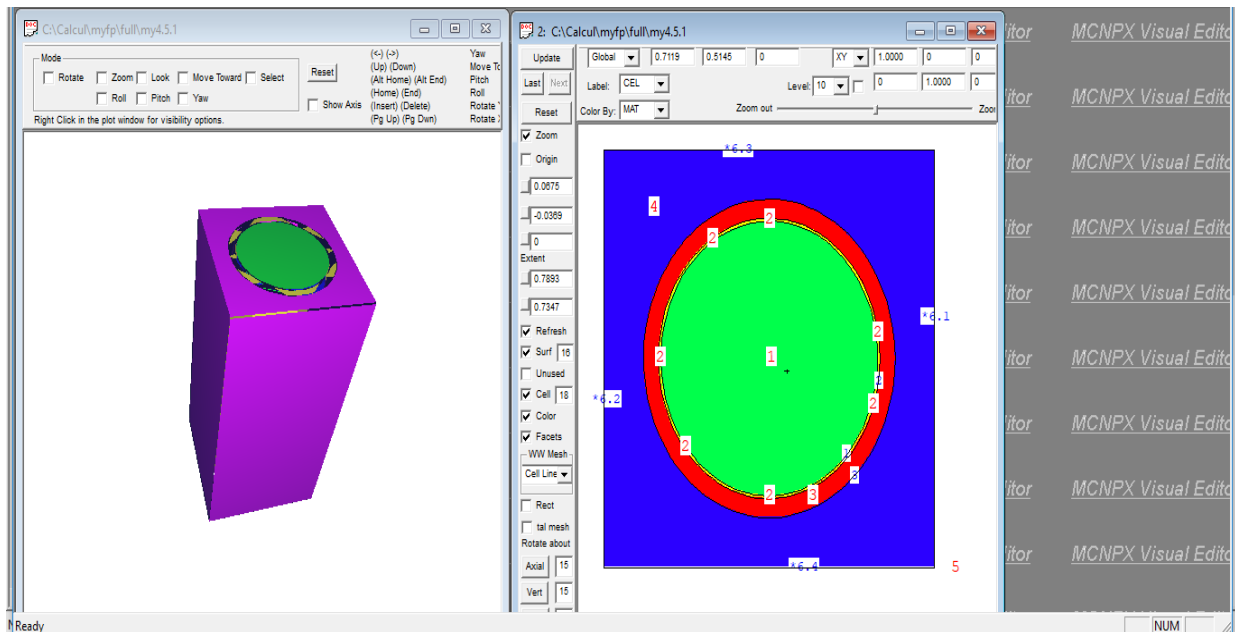


Figure 4-3: A 3D view and top view of the full-size fuel pin model of 4.5 a/o enriched UO_2 in VISED.

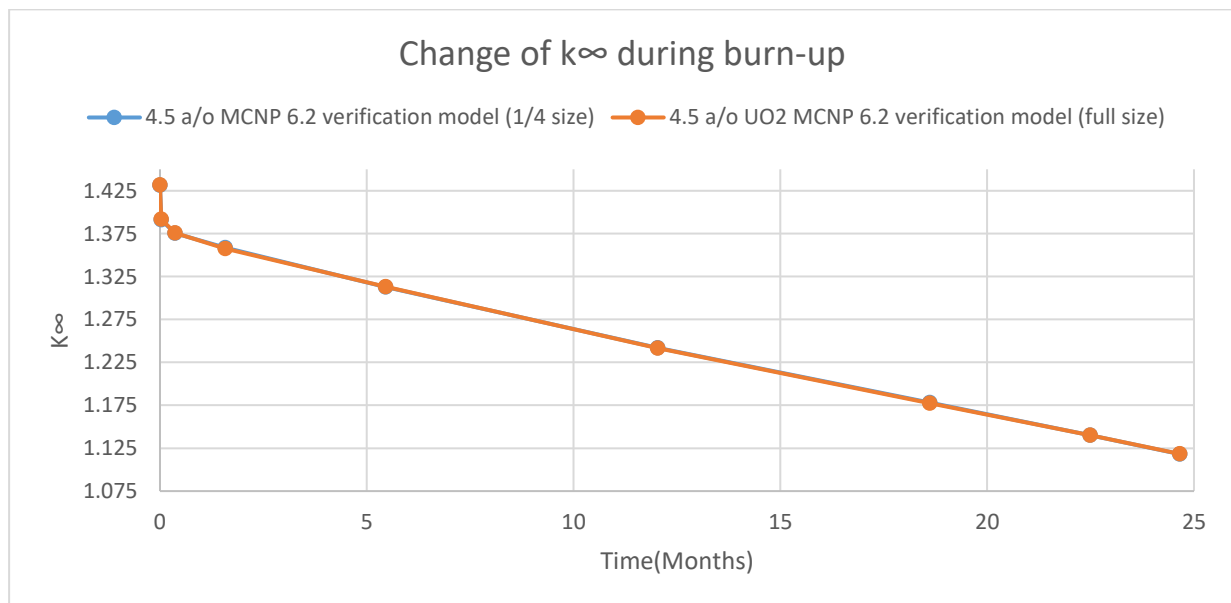


Figure 4-4: Graph comparing the change in k_∞ with time of the $\frac{1}{4}$ MCNP 6.2 verification model and full size MCNP 6.2 reference model both of 4.5 a/o enriched UOX fuel.

The influence of thorium, uranium and plutonium fuel mixture composition on the reactivity coefficients of pressurised water reactors

Table 4-4: K_{∞} comparison at various points in burnup of the $\frac{1}{4}$ pin verification model and the full pin verification model of same Fuel model Composition simulated in MCNP 6.2.

Point in burnup	K_{∞} (verification)- $\frac{1}{4}$ pin (std dev)	K_{∞} (verification)- full pin (std dev)	K_{∞} difference (pcm)	Average (%) difference
BOC	1.4318 (0.0018)	1.43163	17	0.011874
MOC	1.2416 (0.0020)	1.24181	-21	-0.01691
EOC	1.1183 (0.0020)	1.11811	19	0.016992
Average difference (pcm & %)			19	0.0153

**** std dev (standard deviation)**

Figure 4-4 and Table 4-4 clearly indicate that there's a very high correspondence in the K_{∞} values of the two models from BOC through to the EOC. Comparison of computational time of both models is given in **Table 4-5 below** and the $\frac{1}{4}$ fuel pin model cuts computer time by almost 4 days which is quite a significant margin. Overall the results obtained and presented herein verify that the $\frac{1}{4}$ model is as sufficient as the full pin model for calculations and that the theory given in **section 3.3** on the use of symmetry to simply or reduce models in order to reduce computational time is applicable.

Table 4-5: Computational time comparison of a $\frac{1}{4}$ pin model and a full pin model.

MCNP 6.2 Model	Computer time
$\frac{1}{4}$ fuel pin	8677.22 min (6.026 days)
Full fuel pin	14 358.88 min (9.97 days)

4.1.3 Subsection conclusion

From the high correspondence between the results compared, it is clear that the MCNP 6.2 models are sufficient and can be relied on to proceed with calculations. VISED has verified the models by visual means showing the desired geometries for both $\frac{1}{4}$ and full-size pins.

4.2 Three batch “full core” fuel burnup (new operational limit for k_{inf})

This section presents the results of the burnup calculations of the fuel model compositions listed under **Table 4-6** below. **Table 4-6** also gives the duration of the burnup (one full cycle) each fuel composition was subjected to as well as the refuelling period. The burnup calculation results of each fuel composition are portrayed in the form of the change in the infinite neutron multiplication factor with time ran at HFP conditions (**Table 4-2, section 4.1**). The results show fresh fuel, once burnt fuel and twice burnt fuel for each fuel model composition simulated in a quarter of a unit cell fuel pin model. Zero power is used to simulate the refuelling and reshuffling outage which takes place at the end of burning the fresh fuel and burning the once burnt fuel.

The results of the fresh, once burnt and twice burnt fuels is taken and it is assumed that the average of the results represent the change in the neutron multiplication factor with time in a full core as explained in **section 3.5**. From the average of the change in k_{inf} with time of the three burnt fuels the lower operational limit of each fuel composition can be drawn. The k_{inf} value at the EOC of each fuel model composition from the average of the three burns is the lower operational limit. This value can also be called k_{inf} criticality ($k_{inf(crit)}$) because it corresponds to minimum theoretical value of k_{inf} where fission is still sustainable for the models simulated here.

The non-leakage probability is the ratio of k_{eff} criticality ($k_{eff(crit)}=1$) and the lower operational limit ($k_{inf(crit)}> 1$) - see **formula 3.4, section 3.5.1**. Theoretically all the fuel model compositions should have identical lower operational limits and therefore the same non-leakage probability because they are all simulated in the same model of the same size and same geometry.

Note: No burnable absorbers have been added in the models for these burnup simulations (no gadolinia or soluble boron).

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Table 4-6: Simulated Fuel Compositions and their respective cycle lengths and refuelling period.

	Fuel/ Isotopic composition	Cycle length: (months)	Refuelling period (days):
Fuel model A	4.5% enriched UO ₂ (Koeberg)- reference fuel model	≈16	37
Fuel model B	(5% ²³⁵ U, 93% ²³⁸ U and 2% ²³² Th) O ₂	≈17.6-18	37
Fuel model C	(0.8% Pu(PWR), 4.5276 % ²³⁵ U and 94.6724% ²³⁸ U) O ₂	≈18.5- 19.5	37

Table 4-7: Full core fuel terminology.

Term	Meaning
Fresh/fresh burn	Fresh fuel model burning for the first time.
Once burnt fuel	Slightly depleted fuel model burning for the second time.
Twice burnt fuel	More depleted fuel model burning for the third time.

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4.2.1 Results and discussion 3 cycle burnup

Table 4-8, **Table 4-9** and **Table 4-10** present the burnup results of all three fuel model compositions respective fresh, once burnt and twice burnt fuels. It gives the k_{inf} values as well as the U-235 enrichment of each burn at BOC and EOC.

Table 4-9 and **Table 4-10** gives the k_{inf} values as well as the U-235 enrichment of each burn at BOC and EOC for Fuel model B and Fuel model C respectively. For both fuels, a similar trend as explained above for Fuel model A with respect to **Table 4-8** can be observed with the k_{inf} values and U-235 enrichments at BOC and EOC.

Figure 4-5 , **Figure 4-6** and **Figure 4-7** depict the change in k_{inf} over time for fresh, once burnt and twice burnt fuels of fuel model A (LEU UO₂ fuel) , fuel model B (LEU Th-UO₂ fuel) and fuel model C (LEU Pu-MOX fuel) respectively for the duration of each fuel's cycle length.

Table 4-8: Reactivity and enrichment comparisons of fresh, once burnt and twice burnt fuels at BOC and EOC for Fuel model A (4.5 a/o enriched UO₂, Koeberg reference).

Fuel model	Beginning of burn cycle		End of burn cycle		
A: 4.5% UO ₂ (Koeberg reference)					
Burn Cycle	U-235 enrichment (%)	K_{inf}	U-235 enrichment (%)	K_{inf}	Δk_{inf} per cycle (pcm)
Fresh	4.5	1.44012	2.67	1.20662	-23350
Once burnt	2.67	1.24065	1.41	1.05803	-18262
Twice burnt	1.41	1.08703	0.612	0.92447	-16256

The influence of thorium, uranium and plutonium fuel mixture composition on the reactivity coefficients of pressurised water reactors

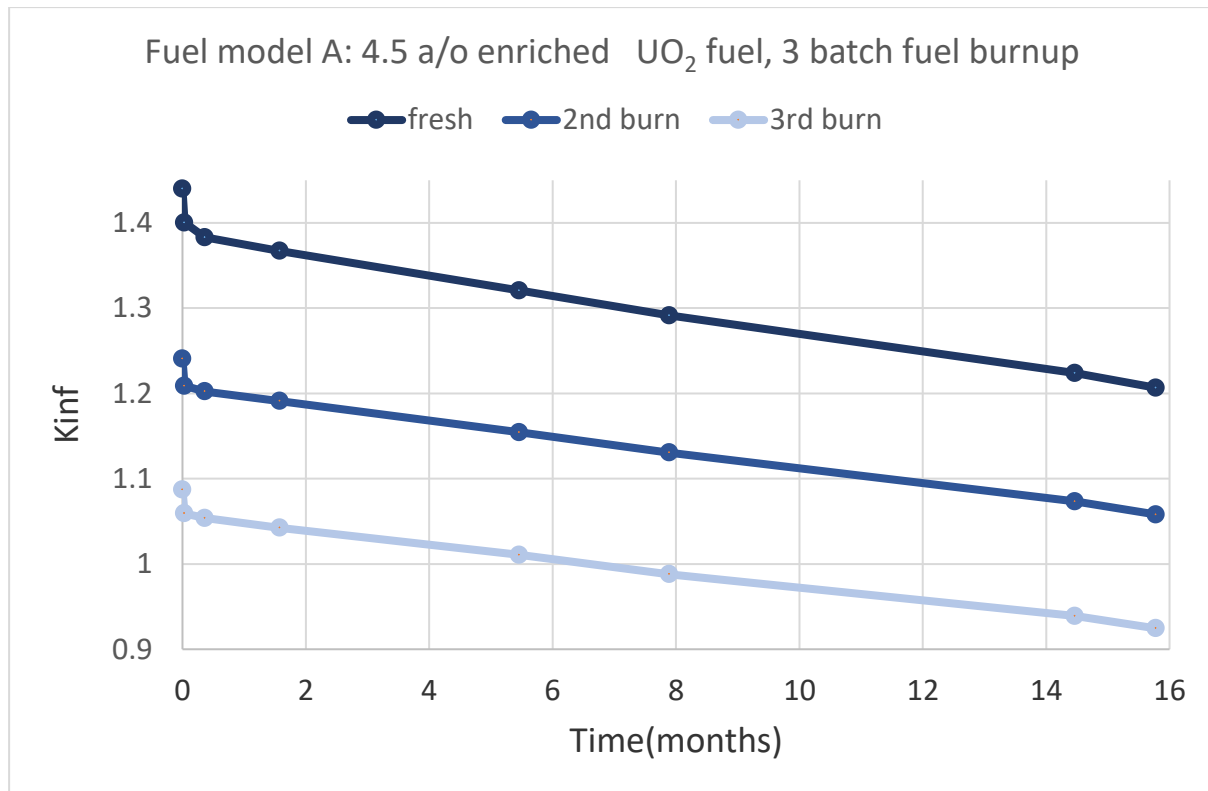


Figure 4-5: Change of k_{inf} with time for fresh, once burnt and twice burnt fuels of fuel model A (4.5 a/o enriched UO_2) for the duration of the fuel's cycle length.

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Table 4-9: Reactivity and enrichment comparisons of fresh, once burnt and twice burnt fuels at BOC and EOC for Fuel model B (5% ^{235}U , 93% ^{238}U & 2% ^{232}Th).

Fuel model B: (5% ^{235}U , 93% ^{238}U & 2% ^{232}Th) O_2	Beginning of burn cycle		EOC		
Burn cycle	U-235 enrichment (a/o)	K_{inf}	U-235 enrichment (a/o)	K_{inf}	Δk_{inf} per cycle (pcm)
Fresh	5	1.44262	2.95	1.20704	-23558
Once burnt	2.95	1.24051	1.53	1.05768	-18283
Twice burnt	1.53	1.08518	0.64	0.92204	-16314

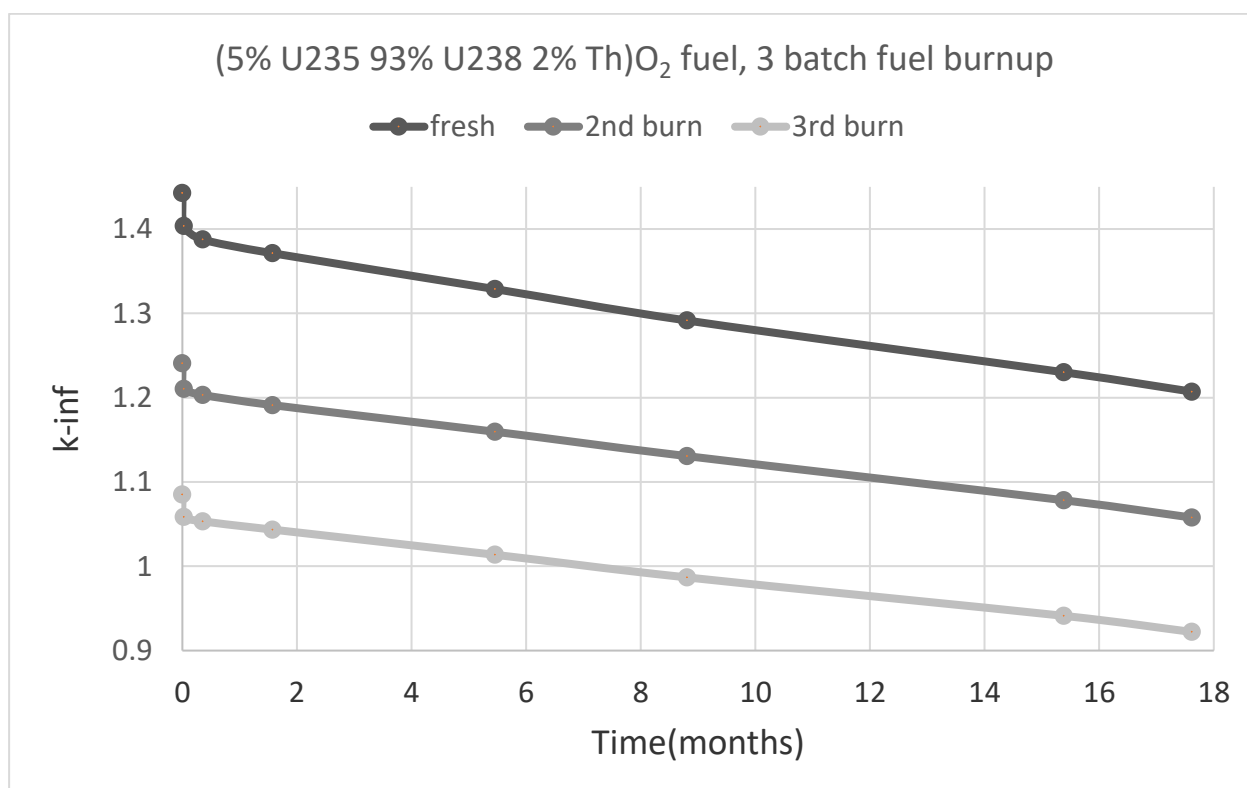


Figure 4-6 : Change of k_{inf} with time for fresh, once burnt and twice burnt fuels of Fuel model B (5% ^{235}U , 93% ^{238}U & 2% ^{232}Th) for the duration of the fuel's cycle length.

The influence of thorium, uranium and plutonium fuel mixture composition on the reactivity coefficients of pressurised water reactors

Table 4-10: Reactivity and enrichment comparisons of fresh, once burnt and twice burnt fuels at BOC and EOC for Fuel model C ((0.8% Pu(PWR), 4.752 % ^{235}U & 94.672% ^{238}U) O_2).

Fuel model C: (0.8% Pu(PWR), 4.5276% ^{235}U & 94.6724% ^{238}U) O_2	Beginning of burn cycle (BOC)		End of burn cycle (EOC)		
	U-235 enrichment (a/o)	K_{inf}	U-235 enrichment (a/o)	K_{inf}	
Fresh	4.5276	1.37979	3.084	1.20888	-17091
Once burnt	3.084	1.2389	1.79	1.08021	-15869
Twice burnt	1.79	1.1066	0.86	0.95035	-15625

The influence of thorium, uranium and plutonium fuel mixture composition on the reactivity coefficients of pressurised water reactors

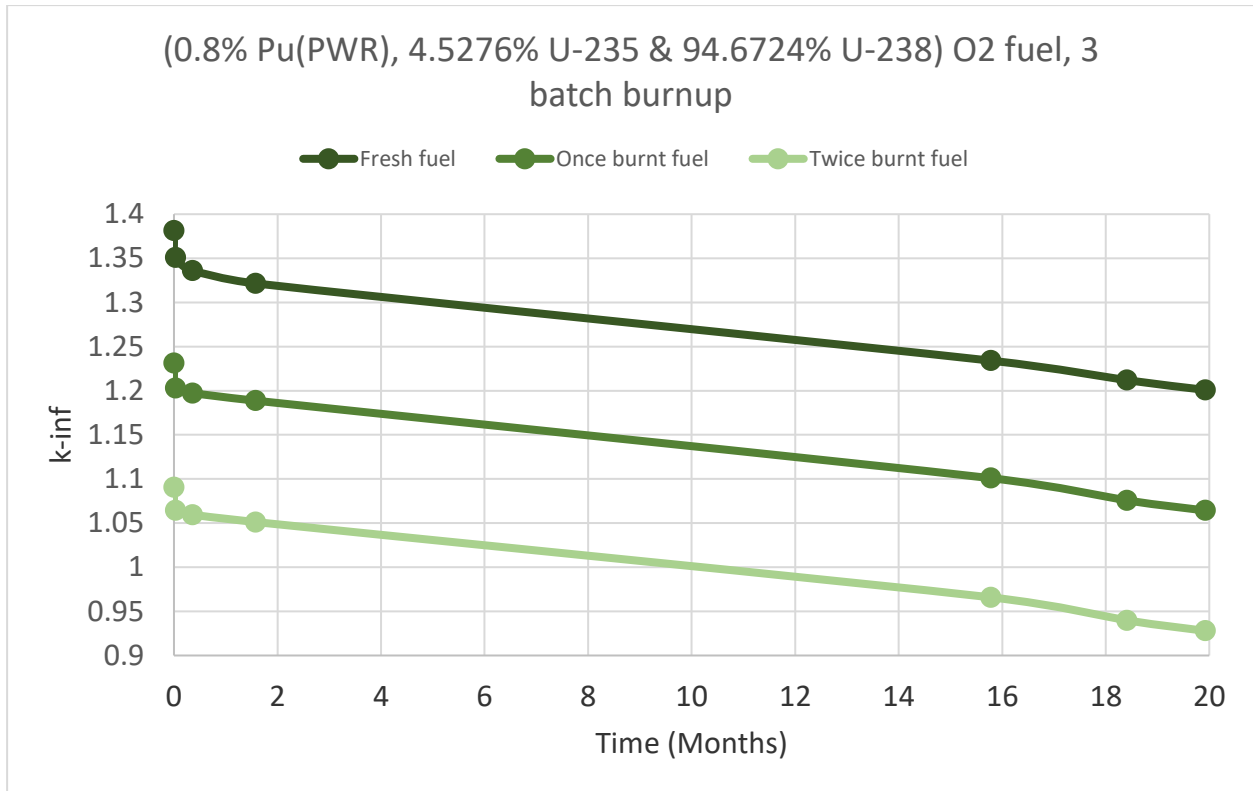


Figure 4-7: Change of k_{inf} with time for fresh, once burnt and twice burnt fuels of Fuel model C of LEU Pu-MOX ((0.8% Pu(PWR), 4.752 % ^{235}U & 94.672% ^{238}U)O₂) for the duration of the fuel's cycle length.

The change in k_{inf} (Δk_{inf}) from BOC to EOC is taken for each burn to deduce the degree of change in reactivity of the fuel for the duration of the cycle length. And as seen in **Table 4-8**, **Table 4-9** and **Table 4-10** the fresh fuel goes through the biggest change in reactivity for duration of its cycle followed by the once burnt fuel then the twice burnt fuel. The obvious explanation for the decrease in reactivity with burn-up is the resulting decrease in enrichment and increase in neutron poison concentration, which large come from the fission products.

4.2.2 Average burn-up behaviour of fresh, first burn and second burn fuel

During the simulated fuel shuffling, the fuel rod at the EOC of one cycle by definition becomes the fuel rod at the BOC of the next cycle. Normally very little transmutation of the fuel nuclides occurs during the relatively short refuelling outage and therefore the enrichments will be virtually identical at the start of the subsequent burn cycle than at the end of the preceding one. The only important exception is Th-based fuels since Pa-233 has a relatively long half-life of approximately 27 days (Hobart, 2013), a substantial amount of Pa-233 will still be present in the fuel at the EOC and thus will decay to U-233 during the outage and, therefore, the enrichment will be substantially higher at the start of the next burn cycle. Though the enrichment at the EOC of the fresh burn and once burnt corresponds to the enrichment at the BOC of the once burnt and twice burnt fuels respectively, as seen in **Table 4-10** the k_{inf} values are different. For example, the k_{inf} value at BOC of once burnt fuel is substantially higher than that at EOC of fresh burn fuel though both points have the same U-235 enrichment. At EOC where the reactor is eventually shutdown, at that instant the fuel is still “hot” due to the radioactive decay fission products. Substantial concentrations of aggressive neutron absorbing fission product poisons such as Xe-135 are present in the fuel at shutdown. In the case of Xe-135 it substantially builds up and peaks approximately six hours after shutdown, due to the remaining inventory of the fission product I-135 (iodine-135) which decays into Xe-135. However, once the I-135 fully depletes, the remaining Xe-135 decays to insignificant concentrations after a few days. Therefore, at the end of the refuelling outage there is practically no Xe-135 left in the fuel and therefore the reactivity increases substantially, even if the enrichment did not change.

By simulating the actual shutdown (37 days refuelling and reshuffling) by setting the power parameter at zero in the burnup code and burning the fuel for 37 days, the accumulated Xe-135 is expected to decay away almost completely by the end of day 37. Hence the k_{inf} value at BOC of the once burnt is higher than that at EOC of fresh burn despite both having the same U-235 enrichment. Similarly, for the EOC of the once burnt and BOC of the twice burnt the same U-235 enrichment is observed though the k_{inf} values are different.

4.2.3 Evaluation of the Lower operational limit

The cycle length of each fuel model composition is obtained from (Erlank, 2017). The cycle lengths of Fuel model B and Fuel model C given in **Table 4-6** may not necessarily be accurate as these were estimated from reading off a graph.

The Pu-MOX fuel has the highest fissile loading (because of the Fissile Pu-239 and Pu-241 which add to the overall enrichment), but yet the initial k_{inf} and k_{eff} are much lower than the counterpart fuels which have relatively lower fissile material loading as seen in **Figure 4-8** and **Figure 4-11**.

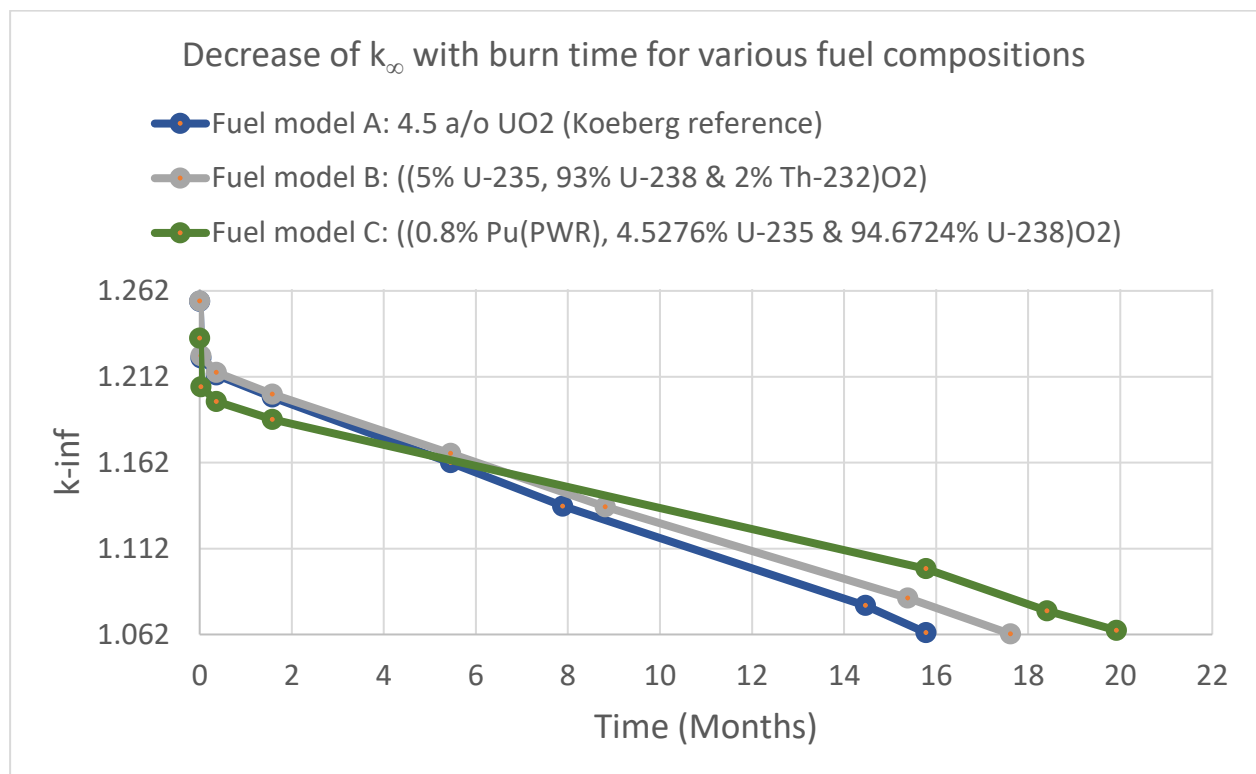


Figure 4-8: The change in k_{inf} with time showing the lower operational limit taken from the average of the fresh, once burnt and twice burnt fuels of fuel model compositions of LEU UO₂, LEU Th-UO₂ and LEU Pu-MOX at HFP.

Figure 4-8 is a depiction of the average of the results presented in **Figure 4-5**, **Figure 4-6** and **Figure 4-7**. This average is an ideal representation of the average depletion in a full core fuelled by either fuel model A (LEU UO₂), fuel mode B (LEU Th-UO₂) or fuel model C (LEU Pu-MOX). The average depletion is taken from the fresh, once burnt and twice burnt fuels of each fuel model

The influence of thorium, uranium and plutonium fuel mixture composition on the reactivity coefficients of pressurised water reactors

composition which represent fresh, once burnt and twice burnt fuel assemblies respectively in an actual core.

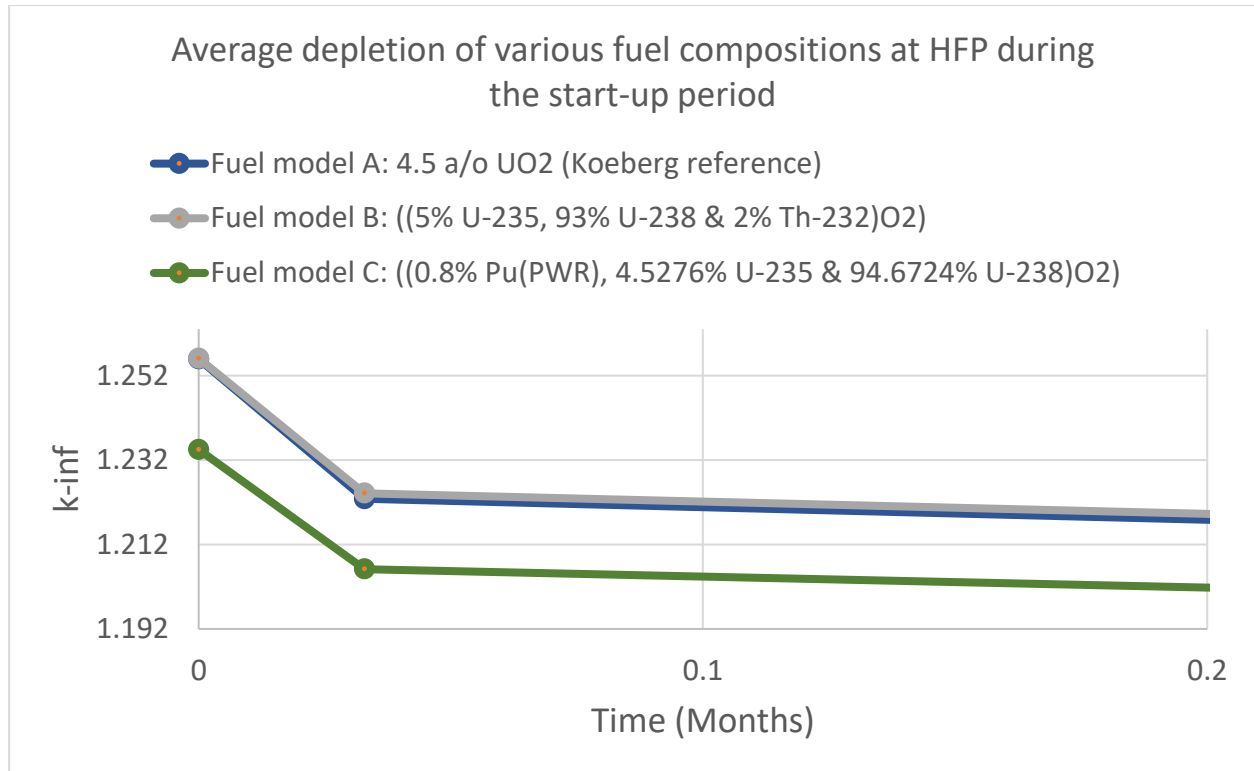


Figure 4-9: The change in k_{inf} with time during the first 0.2 Months taken from the average of the fresh, once burnt and twice burnt fuels at HFP.

As depicted in **Figure 4-8** and **Figure 4-9** Fuel model A and Fuel model B exhibit similar initial k_{inf} values, while Fuel model C (LEU Pu-MOX fuel) has a lower initial k_{inf} . Considering that the burnup calculations were ran hot full power conditions with the fuel at 900k and the moderator at 600k, it should be noted that Pu isotopes are very sensitive to temperature changes and experience large reactivity changes with increasing fuel and moderator temperatures (Mosteller, 2007). This temperature effect on the Pu fuel will be seen under the Doppler coefficient and moderator temperature coefficient result sections when the fuel and the moderator temperatures are varied from CZP conditions (cold zero power, room temperature, 293.6K) to higher temperatures.

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Table 4-11: Total captures and fissions reactions (reactions/second) on day zero of burnup in Fuel Model A, Fuel Model B and Fuel Model C at hot full power (HFP) conditions.

Fuel Composition	Total Capture (n, gamma) Reactions (capture/s)	Total Fissions (n, fission) Reactions (fissions/s)
Fuel model A: 4.5% UO ₂ (Koeberg reference)	1.35823E+13	1.03358E+13
Fuel model B: (5% ²³⁵ U, 93% ²³⁸ U & 2% ²³² Th) O ₂	1.36147E+13	1.0341E+13
Fuel model C: (0.8% Pu(PWR), 4.5276% ²³⁵ U & 94.6724% ²³⁸ U) O ₂	1.39023E+13	1.02961E+13

Presented in **Table 4-11** are the total capture and fission reactions occurring in the given fuel model compositions at HFP (hot full power), and it can be seen in the table that the total capture reactions in Fuel model C (LEU Pu-MOX fuel) are significantly higher than the capture reactions in both Fuel model A (LEU UO₂) and Fuel Model B (LEU Th-UO₂) furthermore the total fission reactions in Fuel model C are much lower than the fissions in both Fuel model A and Fuel model B. Both these effects observed result in a decreased reactivity therefore a lower initial neutron multiplication factor This explains why the initial k_{inf} in Fuel model C is much lower than the initial k_{inf} in Fuel model A and Fuel model C as seen in **Figure 4-8** and **Figure 4-9**.

It should be noted that the capture rate and fission rate presented in **Table 4-11** take into account the actinides and minor-actinides that have accumulated in the once burnt and twice burnt fuels of the given fuel model compositions hence a much larger capture rate than fission rate.

The influence of thorium, uranium and plutonium fuel mixture composition on the reactivity coefficients of pressurised water reactors

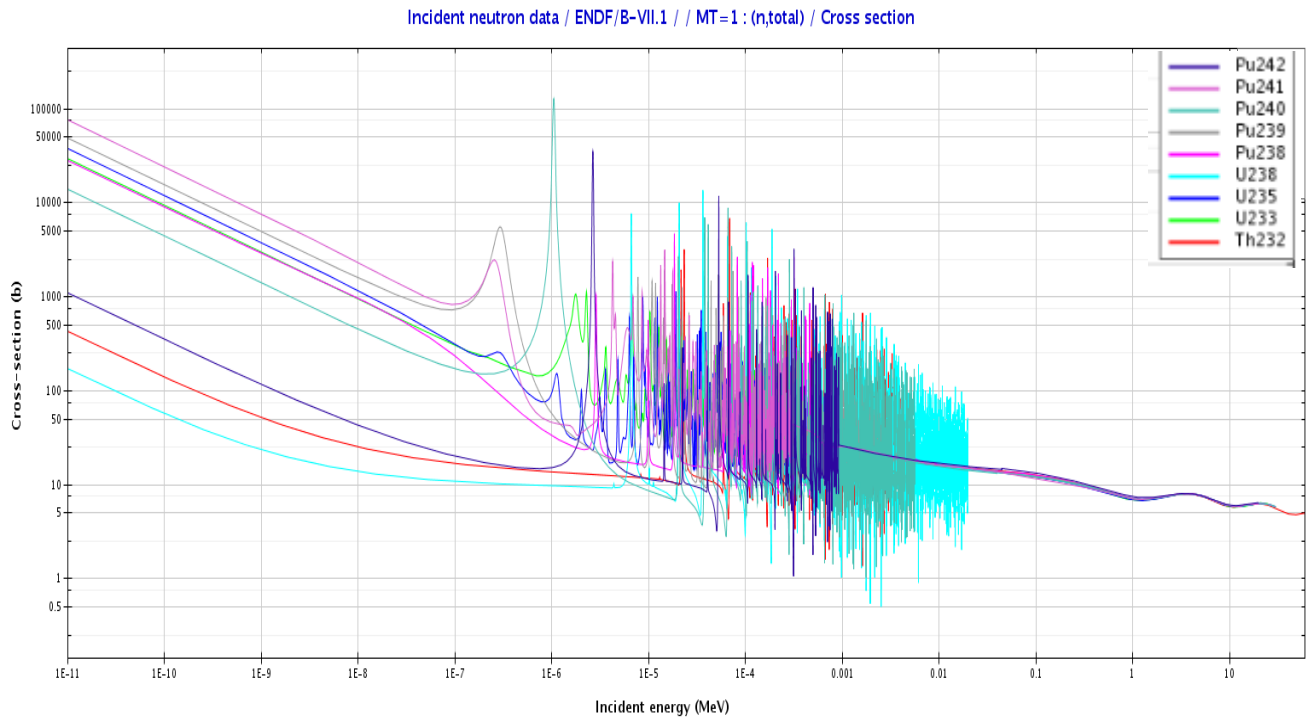


Figure 4-10: Plot showing how the total cross-section of various important isotopes found in Fuel Model A, Fuel Model B and Fuel Model C varies with incident neutron energy.

Table 4-12: The individual capture-to-fission ratio of various fissile nuclides taken from (Lamarsh & Baratta, 2001).

Fissile nuclide	Capture-to-fission ratio (σ_c/σ_f)
U-233	0.089
U-235	0.169
Pu-239	0.362
Pu-241	0.365

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In more depth, the reason for the total high capture reactions in Fuel model C is the high absorption probabilities in Pu isotopes. The fissile Pu isotopes, Pu-239 and Pu-241 have a high absorption peak (capture + fission) around 0.4 eV as seen in **Figure 4-10**, and at this peak the fission probability and capture probability compete for neutrons. The individual capture-to-fission ratios of both fissile nuclides twice as large as the capture-to-fission ratio of fissile U-235 as seen in **Table 4-12** and this shows that the Pu fissile isotopes would have a higher capture probability compared to U-235 (Lamarsh & Baratta, 2001). Again, the fertile isotopes Pu-240 and Pu-242 have their largest absorption peaks around 1eV and below 10 eV respectively. Because these are fertile isotopes, the dominant reaction in this absorption peak is capture. And because the fuel is hot, it is highly likely that the peaks in both isotopes (Pu-240 and Pu-242) have broadened due to the Doppler effect and therefore absorbing neutrons at a much higher rate. This further results in a lower initial reactivity as seen in the LEU Pu-MOX fuel (Fuel Model C) at HFP conditions.

Table 4-13: Comparison of the lower operational limits of fuel model A, fuel model B and fuel model C.

Fuel model composition	Lower operational limit: $k_{inf(crit)}$ (σ)	Lower operational limit difference from reference: (pcm)	Non-leakage probability (thermal and fast) $1/k_{inf(crit)}$	Leakage (%)
Fuel model A: 4.5% UO_2 (Koeberg reference)	1.06304 (0.00046)	0	0.941	5.9
Fuel model B: (5% ^{235}U , 93% ^{238}U & 2% ^{232}Th) O_2	1.062253 (0.00043)	78.7	0.941	5.9
Fuel model C: (0.8% Pu(PWR), 4.5276% ^{235}U & 94.6724% ^{238}U) O_2	1.06436 (0.00045)	132	0.940	6

** σ (standard deviation)

The influence of thorium, uranium and plutonium fuel mixture composition on the reactivity coefficients of pressurised water reactors

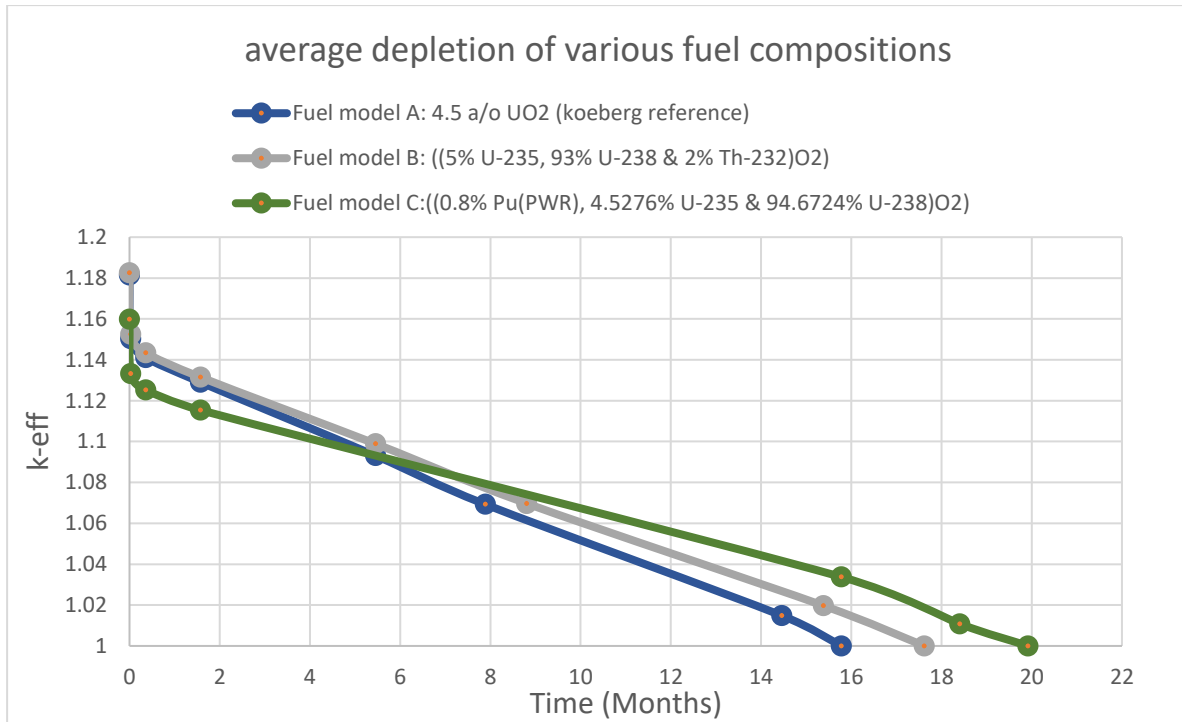


Figure 4-11: The change in k_{eff} for a finite model, over time, for the average of the fresh, once burnt and twice burnt fuels of Fuel model A (LEU UO₂) , Fuel model (B LEU Th-UO₂) and Fuel model C (LEU Pu-MOX).

Ideally, the lower operational limits of fuel model B and fuel model C is supposed to be equivalent to that of Fuel model A (the reference fuel) as the exact cycle length of Fuel model A is known from the reference study by (Erlank, 2017). Ideally all the fuels should have the same non-leakage probability because they are all simulated in the same model of the same size and same geometry. **Table 4-13** gives the lower operational limit and the non-leakage probability computed from the lower operational limit using **Formula 3.4** in **section 3.5.1**. According to the reference fuel's (Fuel model A) results given in the table the model has a very high neutron leakage of almost 6 %. As seen in **Table 4-13** the lower operational limit of Fuel model B and Fuel model C deviates by an average of 100 pcm from that of Fuel model A (reference fuel). Using the non-leakage probability of each fuel, k_{inf} is converted to k_{eff} and **Figure 4-11** depicts depletion in terms of change in k_{eff} with time.

4.2.4 Subsection conclusion

The estimated leakage is quite large compared to typical values. Normally for infinite systems the lower operational limit is taken as 1.03, and this leaves room for 3 % leakage assuming the infinite system is to be converted into a finite system (Du Toit, 2017). This may be due to the estimations of the lower operational limit. As mentioned, the slight deviations of fuel model B's and fuel model C's lower operational limits from that of fuel model A may arise from inaccurately estimated cycle lengths. Overall there's a high correlation between the lower operational limits of all the models and from this the theoretical non-leakage probability of the model is estimated. From the non-leakage probability, k_{inf} is converted to k_{eff} to assume a finite model.

Furthermore, it should be borne in mind that these calculations where the leakage was estimated from simulations for single infinite fuel rods. Such oversimplified calculations ignore important physical effects, such as the influence of fuel assemblies, as opposed to naked fuel rods, on the neutron physics. Another important ignored effect is the influence of older and fresher fuel assemblies that are loaded together in a real core and thus affects the neutron energy spectrum and thus the reaction rates in each other, as opposed to the single fuel rod for which the burn-up from fresh to depleted was simulated in isolation, where after the average core physics behaviours were assumed to represent the behaviour of assemblies that are burning together. For instance, in the present simulations the fresher and older fuel rods were burnt at identical power densities, while in an actual reactor the fresher fuel assembly will burn at much higher power densities than the older ones, which will increase the Xe-135 neutron poison concentrations in the fresher fuel assemblies.

4.3 Achieving criticality at start-up

This section presents the soluble boron concentrations (CBC) that give criticality to each of the fuel model compositions at BOC once at HFP conditions. A fixed amount of burnable absorber gadolinia (Gd_2O_3) is used for each fuel model composition to see how far it brings down the excess reactivity, then soluble boron is used to shave off the rest of the excess reactivity to give criticality ($k_{eff}=1$). The 2.5wt% Gd_2O_3 used in a small selection of fuel rods in the Koeberg PWR core was scaled down to 0.038 wt% to fit the average unit cell fuel pin model used here (see section 3.6.1). Enriched soluble boron 40 a/o% B-10 (20.4 a/o% more B-10 than natural boron) was used and this was to compensate for the absence of one other IBA which is normally added in the fuel ZrB_2 which could not be included in the present simulations due to limitations of the model and for the sake of simplicity. 40 a/o% B-10 enriched soluble boron is commercially available, and it used in several PWRs in Germany and Switzerland (Fridman & Kliem, 2011).

Two dilemmas should be kept in mind. The soluble boron concentration should be as low as possible (<1500ppm) to avoid a positive MTC and the gadolinia percentage should preferably be low enough to avoid having poor heat conductivity in the fuel (Choe, et al., 2016).

The linear interpolation method discussed in section 3.7 is used to predict the critical soluble boron concentration of each fuel model composition with 0.038wt% Gd_2O_3 . Fuel model B and C already contain two chemically distinct fuel materials - i.e Fuel model B contains uranium and thorium while fuel model C contains uranium and plutonium. It should be noted that in practice it is quite difficult to create a completely homogenous ternary mixture of fuel model B with Gd_2O_3 and of fuel model C with Gd_2O_3 (Bjork, 2012). It is important to note that in this section k_{eff} is used.

4.3.1 Influence of Gd_2O_3 on reactivity

The 2.5 wt% Gd_2O_3 used in a few individual fuel rods in an actual core is estimated to be 0.038 wt% Gd_2O_3 on average for the infinite unit cell fuel pin model. This fixed amount of Gd_2O_3 is used in all the Fuel model Compositions and Table 4-14 and shows how much excess reactivity this burnable absorber shaves off from each of the Fuel model Compositions at the BOC at HFP conditions.

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Table 4-14: Reduction in reactivity for each Fuel model composition without burnable absorbers (WoBA) and by adding burnable absorbers (0.038wt% Gd₂O₃) at BOL.

Fuel model composition	K_{eff} (WoBA) BOL	K_{eff} (0.038wt% Gd₂O₃) BOL	Δρ (pcm)
Fuel model A: 4.5% UO₂ (Koeberg reference)	1.1815	1.0267	-12 754
Fuel model B: (5% ²³⁵U, 93% ²³⁸U & 2% ²³²Th) O₂	1.1825	1.0288	-12 630
Fuel model C: (0.8% Pu(PWR), 4.5276 % ²³⁵U & 94.6724% ²³⁸U) O₂	1.1598	1.0227	-11 561

As seen in **Table 4-14**, an excess amount of reactivity still remains for all fuel model compositions. Fuel model A and B have similar amounts of excess reactivity before Gd₂O₃ is added while fuel model C has a lower reactivity. Gd₂O₃ seems to be more effective in bringing down the reactivity of fuel model A as the largest change in reactivity (Δρ) is in fuel model A, the least change in reactivity can be observed in fuel model C. Fuel model A has less fissile content compared to fuel model B and fuel model C. Fuel model C is of Pu-MOX composition and on average has a higher fissile content and would require a higher burnable absorber concentration to achieve the same reactivity change observed in fuel model A.

4.3.2 Critical boron concentration (CBC)

Figure 4-12 and **Table 4-15** displays the excess reactivity left over from the 0.038wt% Gd₂O₃ and the boron concentration needed to reach criticality (k_{eff} =1) for the three fuel model compositions. For all fuels the critical boron concentration is within allowable limits (below 1500ppm).

Fuel model C yields the highest soluble boron concentration to reach criticality followed by fuel model B then fuel model A. Fuel model C is of PU-MOX (Pu(PWR)) composition and PU-MOX fuels are known to have higher critical boron concentrations because of higher average fissile content from isotopes Pu-239 and Pu-241 (Mahrus & Chang, 2018).

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Fuel model B is of thorium composition but has a higher critical boron concentration than fuel model A when thorium fuels are known to have less soluble boron concentration requirements (Du Toit, 2017). But in this case, fuel model B only has 2 a/o% Th-232 and 5 a/o% U-235 enrichment, so it has more fissile content than fuel model A which only has 4.5 a/o% U-235 enrichment. This explains the high critical boron concentration of fuel model B compared to that of fuel model A. Generally, thorium cores may require higher soluble boron concentrations at BOC provided that they have a higher initial fissile content (Dziadosz, et al., 2004).

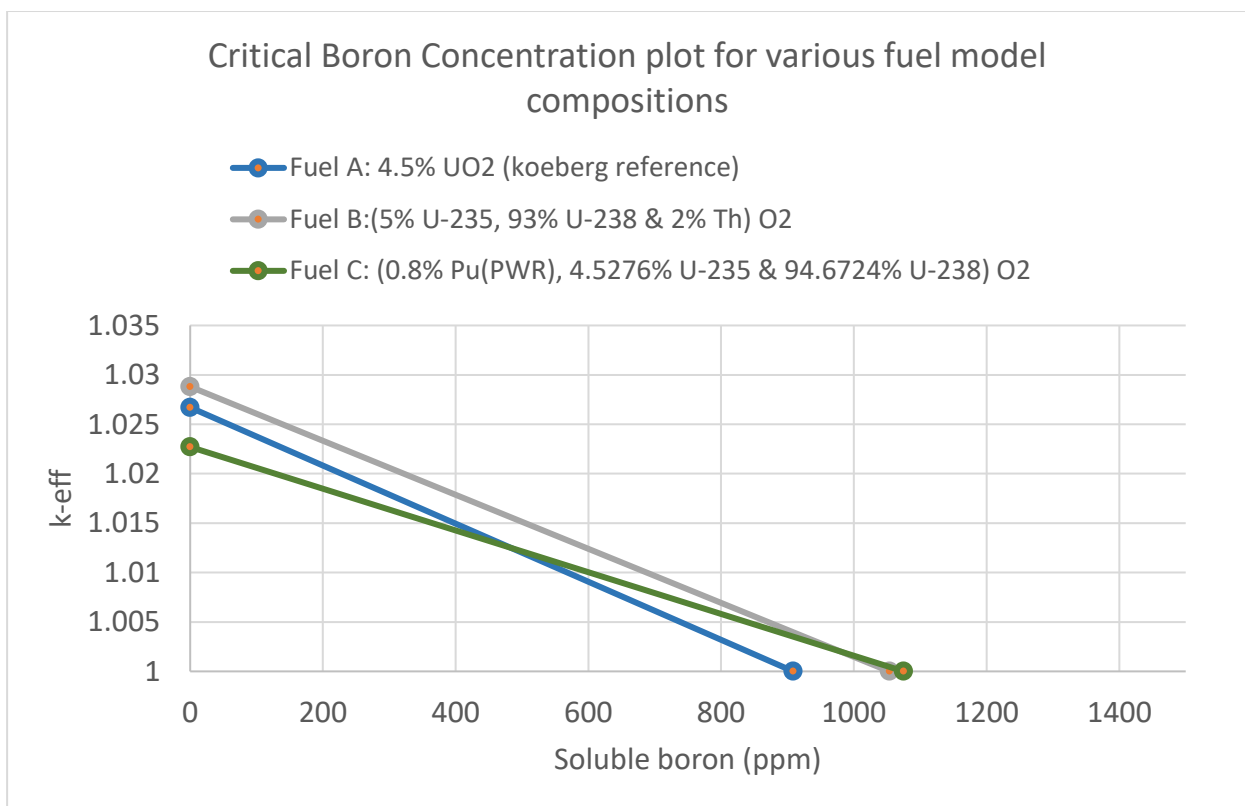


Figure 4-12: Change in k_{eff} as a function of soluble boron concentration for Fuel model A (LEU UO₂), Fuel model B (LEU Th-UO₂) and Fuel model C (LEU Pu-MOX), all compositions with 0.038wt% Gd₂O₃.

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Table 4-15 :The critical boron concentration (CBC) and Change in reactivity of fuel model A, B and C.

Fuel model + 0.038wt% Gd₂O₃	CBC(ppm) ($k_{eff}=1$)	$\Delta\rho$ (pcm)
Fuel model A: 4.5% UO₂ (Koeberg reference)	908.61	-2604.45
Fuel model B: (5% ²³⁵U, 93% ²³⁸U & 2% ²³²Th) O₂	1053.47	-2801.83
Fuel model C: (0.8%Pu(PWR), 4.5276 % ²³⁵U & 94.6724% ²³⁸U) O₂	1075	-2220.09

4.3.3 Critical boron concentration verification

Since the linear interpolation method is used to find the critical boron concentration of each fuel model composition there could be large discrepancies in the obtained critical boron concentrations based on this method. To verify the critical boron concentrations obtained and the method used, new KCODE calculations are ran using the obtained critical boron concentrations from the previous section (section 4.4.2). From these new KCODE calculations the CBC is verified on the basis that $k_{eff(crit)}$ should be equal to one ($k_{eff} = 1$) with the utmost accuracy (Nyalunga, 2016).

Calculation are carried out at HFP conditions. The fuel is at 900k while the rest of the materials (moderator, clad etc) are at 600k. This is at BOC with 0.038wt% Gd₂O₃ in each fuel model composition and the respective CBC in each.

Presented in Table 4-16 below are the $k_{eff(crit)}$ results (rounded off to 4 decimal places) showing that the estimated CBCs from the previous section gives values very close to criticality ($k_{eff} = 1$).

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The CBC of fuel model B might've been a bit under estimated by the method as the $k_{eff(crit)}$ value deviates from criticality by 70 pcm.

Table 4-16 :Critical boron concentration (CBC) verification results for each fuel model composition.

Fuel model +0.038wt% Gd₂O₃ + CBC	CBC(ppm)	$K_{eff(crit)}$ (σ)	Difference from criticality ($K_{eff(crit)} - 1$) (pcm)	Average percentage difference (%)
Fuel model A: 4.5% UO₂ (Koeberg reference)	908.61	0.9999 (0.00011)	-10	0.01%
Fuel model B: (5% ²³⁵U, 93% ²³⁸U & 2% ²³²Th) O₂	1053.47	1.0007 (0.00012)	69	0.07%
Fuel model C: (0.8%Pu(PWR), 4.5276 % ²³⁵U & 94.6724% ²³⁸U) O₂	1075	1.0000 (0.00011)	0	0.00%

4.3.4 Subsection conclusion

Gd₂O₃ has the biggest influence on reactivity, compared to soluble boron, even though the soluble boron has been enriched. Gd₂O₃ substantially reduces the initial excessive reactivity and thus also reduces the soluble boron concentration (Nuclear-Power, 2017). This is in line with the theory discussed in **section 2.3** and depicted in **Figure 2-3** by the boron let down curve. The linear interpolation method turned out to be good enough to estimate the critical boron concentration of each fuel model composition. The results given in **Table 4-16** shows that the estimated critical boron concentrations for the three fuel models gave k_{eff} values that were very close to unity ($k_{eff}=1$).

4.4 Analysis of the Doppler Coefficients

In this section the doppler coefficients of the three fuel model compositions were analysed. Two separate sets of calculations were performed for the doppler coefficients. The first set calculates the Doppler coefficients of only the fresh fuels of the three fuel model compositions based on the Mosteller benchmark for fresh fuel pin compositions. The second set of calculations used the MCNP simulations, according to the full core model concept described in **section 3.5**, to estimate the Doppler coefficients of the three fuel model compositions (Fuel model A, fuel model B and fuel model C), mimicking a full core with a three- batch loading scheme (fresh, once burnt and twice burnt fuel).

The significance of using the Mosteller benchmark and analysing only the fresh fuel models of Fuel model A, Fuel model B and Fuel model C is to gauge the Doppler coefficients of the three fuels without any interference from their respective once and twice burnt fuels which may contain similar species of actinides and minor actinides. The Mosteller benchmark provides Doppler coefficients for fresh UO_2 and PU-MOX (Pu(PWR)) fuels with a range of fissile material content which can be used for comparison purposes for the Doppler coefficients calculated for Fuel model A and Fuel model C which are of similar UO_2 and MOX composition respectively. Fuel model B is of thorium composition and unfortunately a study calculating the Doppler coefficients in thorium-based fuels based on the Mosteller benchmark wasn't found during this study.

4.4.1 Mosteller benchmark (Doppler coefficient analysis)

This section presents the doppler coefficient results of the fuel model compositions. The results are matched against a reference study found simulating 4.5 % enriched UO_2 fuel under the Mosteller benchmark specification to investigate the change in reactivity in terms of the Doppler coefficient between hot zero power fuel conditions (HZIP-600k) and hot full power fuel conditions (HFP-900k) fuel temperature conditions using ENDF/B-VII.1 cross-sections in MCNP 6.1 (Jo & Kim, 2015).

All necessities of the benchmark are given under section 2.5.2 which include the model and the simulation conditions. A pin cell model is sufficient for studying the Doppler coefficients of fuels and a full core is not really necessary (De Kruijf, 1995).

Since Fuel model A (4.5% UO_2 (Koeberg reference)) is the reference for the other fuel model compositions, for extra legitimacy its results are compared to a similar model of similar fuel composition and enrichment by (Jo & Kim, 2015) and as well as the (Mosteller, 2007) model. A

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similar model to fuel model C ((0.8% Pu(PWR), 4.5276 % ^{235}U & 94.6724% ^{238}U) O_2) is also found in the study by (Mosteller, 2007) and is also used for reference purposes. No Mosteller benchmark results that could be used for reference purposes were found for thorium fuels to compare fuel model B ((5% ^{235}U , 93% ^{238}U & 2% ^{232}Th) O_2) which is of $\text{ThO}_2\text{-UO}_2$ composition.

It is important to note that the doppler coefficients calculated are only for the fresh fuels of each Fuel composition (Fuel model A, Fuel model B & Fuel model C). The benchmark assumes only pure fuel compositions with no fission products nor impurities (Mosteller, 2007).

Table 4-17 gives the benchmark's UO_2 and PU-MOX fuels fissile enrichment and reactor grade Pu(PWR) content ranges respectively, as well as the doppler coefficient ranges corresponding to the specific content.

Table 4-17: Mosteller benchmark's Pure UO_2 Fuel And pure PU-MOX fuels content and DC ranges corresponding to the given content (Mosteller, 2007).

Fuel type	Fissile content	Doppler Coefficient range (pcm/k)
UO_2	4%-5% U-235 content	(-2.5; -2.0)
PU-MOX ($\text{PuO}_2\text{-UO}_2$)	0-1% PuO_2 (Pu(PWR)) content	(-4.5; -3.7)

4.4.1.1 Results and discussion

KCODE calculations of 500 inactive cycles, 3500 active cycles and 10 000 neutrons per cycle are performed for the fresh fuel of each Fuel model Composition with the aid of MCNP (Jo & Kim, 2015).

Presented in **Table 4-18** are k_{inf} values of the reference fuel from a study by (Jo & Kim, 2015) obtained using MCNP6.1 and the k_{inf} values of fuel model A, fuel model B and fuel model C obtained from MCNP 6.2 in the current study. A standard deviation (σ) of less than 12pcm on average for the k_{inf} values was observed for each of the fuels simulated. The Doppler Coefficient is calculated using **formula 2.5** given in **section 2.5.2**. **Formula 2.5** is the general Doppler coefficient formula expressed in terms of k_{inf} and change in fuel temperature (ΔT_f) (De Kruijf, 1995).

For each fuel model composition, the k_{inf} value decreases from HZP to HFP fuel temperature conditions. However, this decrease k_{inf} was expected for all fuel model composition considering that each fuel has sufficient fertile material (mostly U-238) for neutrons to be absorbed in the resonances owing to the Doppler Effect. So, the reactivity decreases with increasing temperature and with that yielding a negative Doppler coefficient for all the fuel model compositions. However, the change in reactivity for each fuel model composition is slightly less than expected. A reactivity change in the order of 1000pcm was expected between HZP and HFP (Mosteller, 2007).

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Table 4-18 :The Mosteller benchmark Doppler coefficient comparison in fresh fuel of various fuel model compositions between hot zero power (HZP-600k) and hot full power (HFP-900k) at BOC with 1400ppm soluble boron.

	$K_{inf(HZP)}$ (σ)	$K_{inf(HFP)}$ (σ)	$\Delta\rho$ (pcm) ($\rho_{HFP}-\rho_{HZP}$)	Doppler coefficient DC (pcm/k)
MCNP 6.1 reference model				
(4.5 % enriched UO₂) (Jo & Kim, 2015)	1.26615 (0.00012)	1.27648 (0.00012)	-639.148	-2.1305
MCNP 6.2				
Fuel model A: 4.5% UO₂ (Koeberg reference)	1.34441 (0.00011)	1.33331 (0.00011)	-619.242	-2.0641
Fuel model B: (5% ²³⁵U, 93% ²³⁸U & 2% ²³²Th) O₂	1.341113 (0.0012)	1.32852 (0.00010)	-706.799	-2.3560
Fuel model C: (0.8% Pu(PWR), 4.5276 % ²³⁵U & 94.6724% ²³⁸U) O₂	1.27575 (0.00011)	1.26158 (0.00012)	-880.419	-2.9347

**** DC acceptable range (-4.90; -2.40) (IAEA, 2003)**

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With reference to **Table 4-18**. The MCNP 6.1 reference fuel model and fuel model A are both of the same composition and U-235 enrichment of 4.5%, but due to the slight differences in the geometry, material specifications and configuration of the fuel pin models used to simulate this same fuel model composition, the obtained k_{inf} values vastly differ both at HZP and HFP. Despite the differences in k_{inf} values between the two models at each condition, similar changes in k_{inf} values can be observed for each model between HZP and HFP. The change in reactivity for both between HZP and HFP is in the order of 600 pcm and also similar Doppler coefficients can be observed.

The Doppler Coefficient value of fuel model A falls within the Mosteller benchmark's range given in **Table 4-17** for pure UO_2 fuels with enrichment of between 4% to 5 % for U-235.

Fuel model C, the PU-MOX fuel's Doppler coefficient is strongly negative as seen in the table. The Doppler coefficient value doesn't fall within the Mosteller benchmark's PU-MOX fuel ranges for PuO_2-UO_2 fuel with less than 1% Pu(PWR). A Plausible reason for the PU-MOX fuel (Fuel model C) not entirely falling within the Mosteller DC ranges is that the fuel model contains 0.8% Pu(PWR) as well as enriched UO_2 (4.5276 % U-235) while the Mosteller PuO_2-UO_2 fuel model contains 0-1wt% Pu(PWR) and only natural uranium in UO_2 . The higher the fissile enrichment content in a specific type of fuel, the less negative the doppler coefficient becomes (Mosteller, 2007).

4.4.1.2 Comparing the Doppler Coefficients of Fresh Fuel compositions

Fuel model C has the biggest change in reactivity between the two operating conditions, followed by fuel model B. fuel model A has the smallest change in reactivity between HZP and HFP. Only the calculated Doppler coefficients of fuel model C falls within the IAEA acceptable range.

Fuel model C is very sensitive to temperature changes as shown by the magnitude of the Doppler coefficient. PU-MOX fuels are known to have a high resonance absorption rate compared to UO_2 hence a high Doppler coefficient. This is mainly due to the resonance peak in Pu-240 at thermal energies hence a higher Doppler Coefficient for Pu-MOX fuels (Mahrus & Chang, 2018).

The thorium in Fuel model B ($LEU\ Th-UO_2$) gives a favourably larger negative Doppler Coefficient value compared to fuel model A. A justifiable explanation for the large Doppler coefficient of fuel model B has opposed to fuel model A (of UO_2 composition) is the stronger doppler effect of Th-232 at elevated temperatures which surpasses that of U-238 (Fridman & Kliem, 2011). Another explanation might be that with resonance capture the joint effect of two isotopes is more than the

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sum of the two individual effects: Due to self-shielding in the large capture resonances the effectiveness of neutron capture decreases with increasing isotope number densities, i.e. 100 % of an isotope concentration will capture substantially less neutrons than twice the number of captures at 50% of the concentration. Put differently, if you have one fuel rod with 100% U-232 and another with 100% U-238 they will capture substantially more neutrons if they were to be mixed together into two fuel rods each containing 50% U-238 and 50% Th-232. (The reason being that each fuel rod will then experience less self-shielding.) This might explain why a small addition of Th-232 in this case resulted in a substantial increase in the number of captures and in the Doppler Coefficient. This arises from the relative capture of Th-232 which happens to be larger than that of U-238 (Dobuchi, et al., 2016).

4.4.1.3 Subsection conclusion (Mosteller benchmark's doppler coefficient)

The fresh PU-MOX fuel (fuel model C) yields a negatively large Doppler coefficient than the other fuels which falls within the IAEA's set range. Fuel model A and Fuel model B also yield negative Doppler coefficients, but these do not fall within the IAEA's recommended range. Thorium-based fuels have a strongly negative doppler coefficient than UO_2 fuels and this is evident when comparing the doppler coefficients of fuel model B (of Th composition) and fuel model A (of UO_2 composition).

4.4.2 Doppler Coefficients (DCs) "Three- batch full core concept" comparison

This section aims to conceptually show the influence of Fuel model A, Fuel model B and Fuel model C on the DC of a critical PWR core at BOC. Firstly, the Doppler coefficients of the mimicked full cores containing fuel model A, fuel model B and fuel model C are analysed between HZP and HFP. Then the doppler coefficients are analysed at conditions exceeding HFP (1200K) to investigate how each fuel model compositions influences the prompt reactivity feedback of a PWR in worse situations.

Critical cores of full UO_2 , $\text{UO}_2\text{-ThO}_2$ and $\text{UO}_2\text{-PuO}_2$ are represented by fuel model A, fuel model B and fuel model C respectively. A quarter sized unit fuel pin model is used to mimic a full reactor core (Koeberg PWR) and details on this concept are discussed in section 3.5. The conceptual full cores contain fresh, once burnt and second burnt compositions of each fuel model composition. It is important to note that the once burnt and twice burnt fuels contain various species of actinides and minor actinides.

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Calculated in section 4.3 are the BOC criticality requirements of each fuel model composition obtained from the use of Gd_2O_3 in each fuel model composition and 40wt% enriched soluble boron in the moderator. As mentioned, the choice of using these two burnable absorbers is limited by the model. To avoid any bias when comparing the results of Fuel model A, fuel model B and fuel model C it was decided that the same amount of Gd_2O_3 is used in each fuel. Enriched soluble boron is used to compensate for the absence of other burnable absorbers present in an actual core to ensure that each fuel model Composition simulated reaches criticality with recommended burnable absorber limits. PU-MOX fuel generally requires more BAs than UO_2 and ThO_2 fuels.

In the study (Fridman & Kliem, 2011) full cores of similar fuel model composition as in the current study are simulated in a Westinghouse PWR core. 40 wt% enriched soluble boron together with WABAs (wet annular burnable absorbers) and IFBAs (integral fuel burnable absorbers) are used to hold down the excess reactivity at BOL. The study is a credible reference because it simulates a similar core to the reference core (Koeberg) of the current study and it simulates the full core comprised of fresh, once burnt and twice burnt of similar fuel model compositions to fuel model A and fuel model C which are UOX and PU-MOX cores respectively. The reference provides means to gauge the effectiveness of the concept used to try and mimic a full core using only an infinite unit cell fuel pin of quarter size.

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4.4.2.1 Results

Table 4-19, Table 4-20 and Table 4-21 present the Doppler coefficients of the three fuel models of LEU UO₂, LEU Th-UO₂ and LEU Pu-MOX and shows that all three fuel compositions possess the required negative DC between the selected temperature ranges. The Doppler coefficients are sufficiently negative and in accordance with the IAEA recommended DC range.

Table 4-19: Change in fuel reactivity with fuel temperature changes between CZP-Cold zero power (293.6kk) and HZP-hot zero power (600k) for fuel model A, fuel model B and fuel model C.

Fuel+0.038wt% Gd ₂ O ₃ + CBC	K _{eff} (CZP) (σ)	K _{eff} (HZP) (σ)	Δρ (pcm) (ρ _{HZP} -ρ _{CZP})	α _{DC} (pcm/k)
Fuel model A: 4.5% UO₂ (Koeberg reference)	1.0234 (0.00011)	1.0101 (0.00013)	-1284.02	-4.1907
Fuel model B: (5% ²³⁵U, 93% ²³⁸U & 2% ²³²Th) O₂	1.0251	1.0114 (0.00011)	-1323.01	-4.3179
Fuel model C: (0.8% Pu(PWR), 4.5276 % ²³⁵U & 94.6724% ²³⁸U) O₂	1.0259	1.01143 (0.00012)	-1396.2	-4.5568

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Table 4-20: Change in fuel reactivity with fuel temperature changes between HZP-hot zero power (600k) and HFP-hot full power (900k) for fuel model A, fuel model B and fuel model C.

Fuel+0.038wt% Gd ₂ O ₃ + CBC	K _{eff} (HZP) (σ)	K _{eff} (HFP) (σ)	Δρ (pcm) (ρ _{HFP} -ρ _{HZp})	α _{DC} (pcm/k)
Fuel model A: 4.5% UO₂ (Koeberg reference)	1.0101 (0.00013)	0.9999 (0.00011)	-1010.54	-3.3685
Fuel model B: (5%²³⁵U, 93%²³⁸U & 2%²³²Th) O₂	1.0114 (0.00011)	1.0007 (0.00011)	-1054.82	-3.5161
Fuel model C: (0.8% Pu(PWR), 4.5276 %²³⁵U & 94.6724%²³⁸U) O₂	1.01143 (0.00012)	1.0000 (0.00011)	-1128.93	-3.7631

**** DC acceptable range (-4.90; -2.40) (IAEA, 2003)**

Table 4-21: Change in fuel reactivity with fuel temperature changes between HFP-hot full power (900k) and 1200K for fuel model A, fuel model B and fuel model C.

Fuel+0.038wt% Gd ₂ O ₃ + CBC	K _{eff} (HFP) (σ)	K _{eff} (1200k) (σ)	Δρ (pcm) (ρ _{1200k} -ρ _{HFP})	α _{DC} (pcm/k)
Fuel model A: 4.5% UO₂ (Koeberg reference)	0.9999 (0.00011)	0.9916 (0.00012)	-836.163	-2.7872
Fuel model B: (5%²³⁵U, 93%²³⁸U & 2%²³²Th) O₂	1.0007 (0.00011)	0.9924 (0.00011)	-838.97	-2.7966
Fuel model C: (0.8% Pu(PWR), 4.5276 %	1.0000 (0.00012)	0.99045 (0.00011)	-965.017	-3.2167

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^{235}U & 94.6724%

^{238}U) O_2

** DC acceptable range (-4.90; -2.40) (IAEA, 2003)

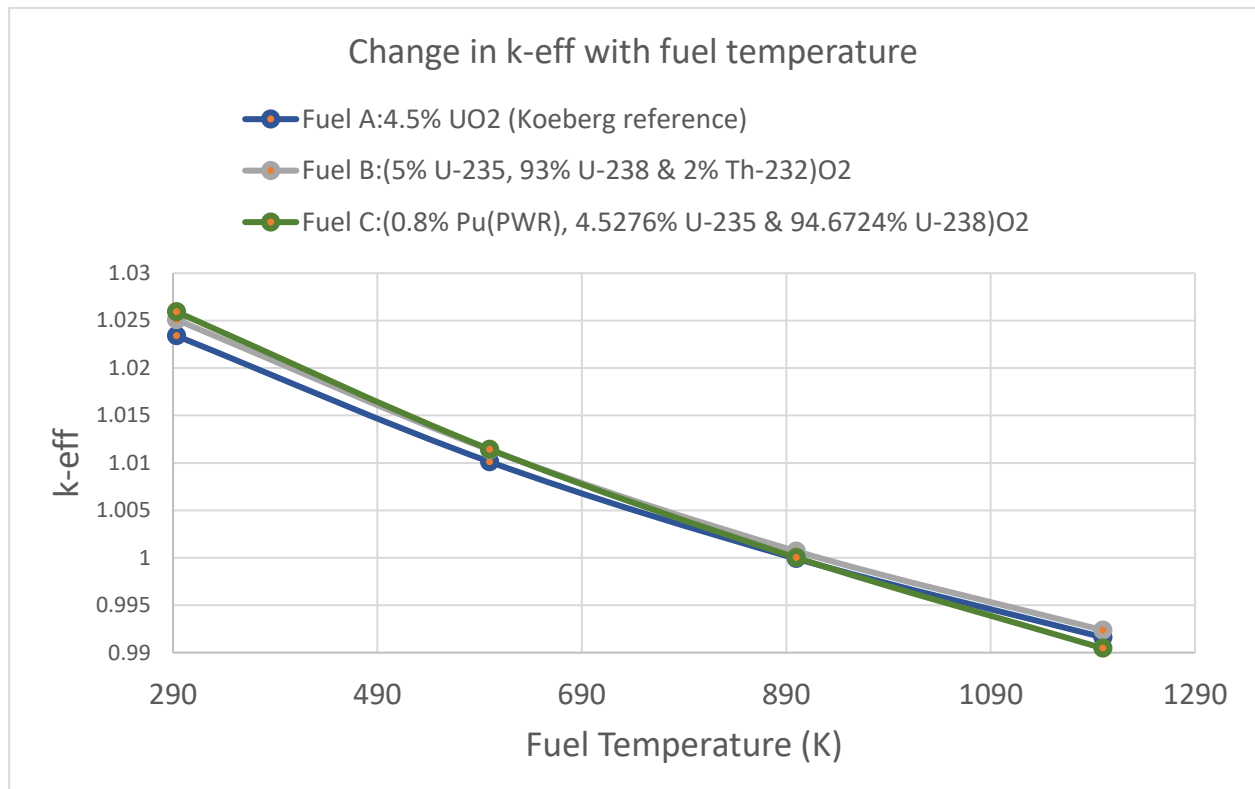


Figure 4-13: Change in k-eff of Fuel model compositions of LEU UO_2 , LEU Th- UO_2 and LEU Pu-MOX fuels with increasing fuel temperatures from CZP-293.6K to 1200K at the BOL (beginning of life) - All fuel compositions with 0.038wt% Gd_2O_3 .

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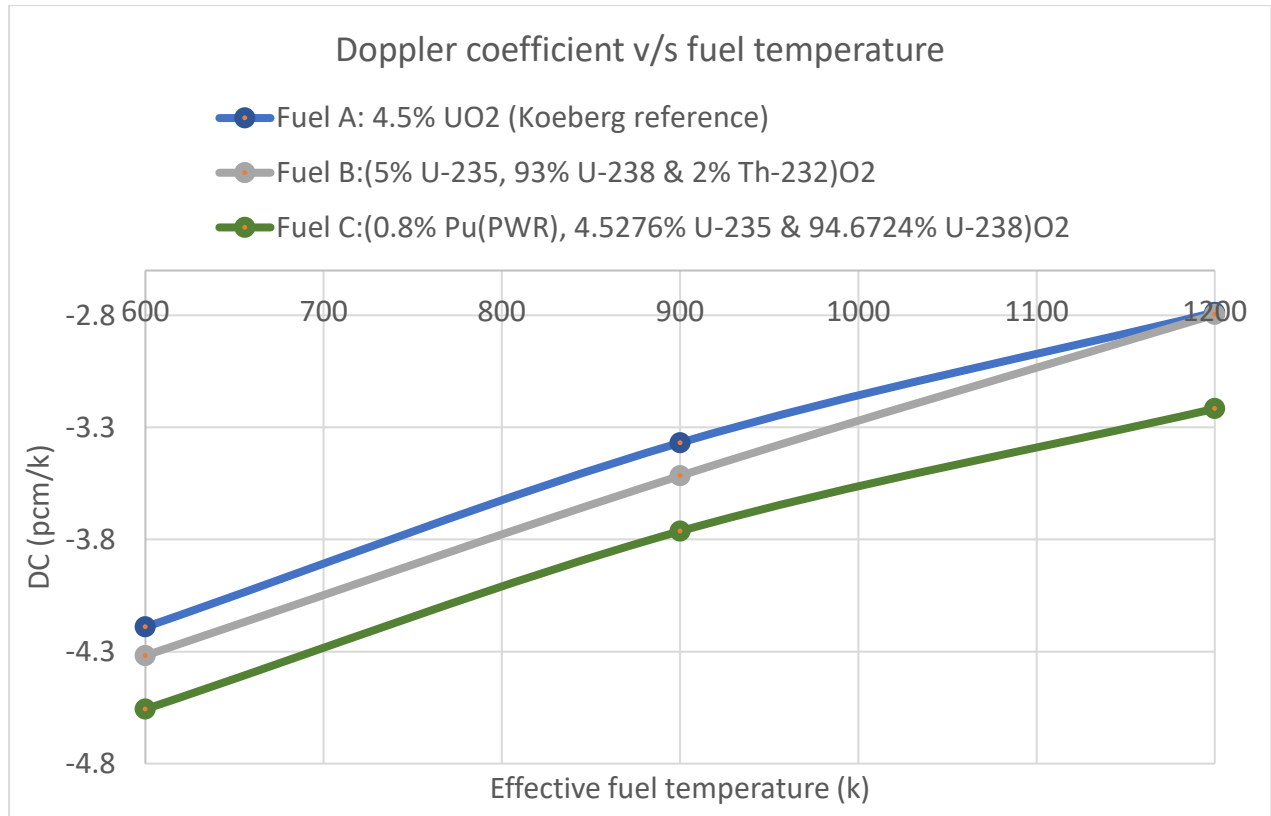


Figure 4-14: The Doppler Coefficients as a function of fuel temperature for Fuel model A (LEU UO₂), Fuel model B (LEU Th-UO₂) and Fuel model C (LEU Pu-MOX)- All fuel compositions with 0.038wt% Gd₂O₃.

Table 4-22 : The rate of change of the doppler coefficient with increasing fuel temperature.

Fuel model Composition	$\Delta DC / \Delta T$ (10^{-3})
Fuel model A: 4.5% UO₂ (Koeberg reference)	1.93
Fuel model B: (5% ²³⁵U, 93% ²³⁸U & 2% ²³²Th) O₂	2.4
Fuel model C: (0.8% Pu(PWR), 4.5276% ²³⁵U & 94.6724% ²³⁸U) O₂	1.8

The largest change in reactivity is between the lower fuel temperatures (CZP-293.6k and HZP-600k) and it is above a 1000 pcm, and the smallest change in reactivity is at higher fuel temperatures (HFP-900k and 1200K) and it is below 1000 pcm as seen in **Table 4-19** and **Table 4-21** respectively. Presented in **Figure 4-13** is the change in k_{eff} with increasing fuel temperature, and it is seen that k_{eff} decreases with increasing fuel temperature. A general trend observed in the relationship between the Doppler coefficient and fuel temperature is that the magnitude of the negative Doppler coefficient becomes substantially smaller with increasing fuel temperature as seen in **Figure 4-14** (Mahrus & Chang, 2018). Presented in **Table 4-22** is the change in doppler coefficient with fuel temperature and it can be described as the “rate of change in the Doppler coefficients with increasing temperature”. All the fuels Doppler coefficients vary in a somewhat similar fashion with increasing temperature, especially fuel model A and fuel model C as seen in **Figure 4-14**. But fuel model B's doppler coefficient seems to decrease more rapidly with increasing temperature than the doppler coefficients of its counterparts. This is merely an observation in the results obtained in this study and further investigations are needed to approve or disapprove this finding.

4.4.2.2 Technical evaluation

Given in **Table 4-23** is the total capture to fission ratio in each fuel model composition. The big contributors of capture and fission reactions are of course the fertile and fissile isotopes, while the neutron poisons play a lesser role. The once burnt and twice burnt fuels of each fuel model composition contains actinides and minor actinides. The minor actinides only contribute to capture while the actinides contribute to both capture and fission. There's Gd_2O_3 of the same percentage inserted in the fresh fuels of each fuel model composition and it contributes to capture.

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Table 4-23 : Total capture to fission ratio of each Fuel model composition at each fuel temperature condition.

Fuel model composition + 0.038wt% Gd₂O₃	Cold zero power (293.6k)	Hot Zero Power (600k)	Hot Full Power (900k)	1200k
Fuel model A: 4.5% UO₂ (Koeberg reference)	1.26	1.29	1.31	1.33
Fuel model B: (5% ²³⁵U, 93% ²³⁸U & 2% ²³²Th) O₂	1.26	1.29	1.32	1.34
Fuel model C: (0.8% Pu(PWR), 4.5276 % ²³⁵U & 94.6724% ²³⁸U) O₂	1.31	1.32	1.35	1.37

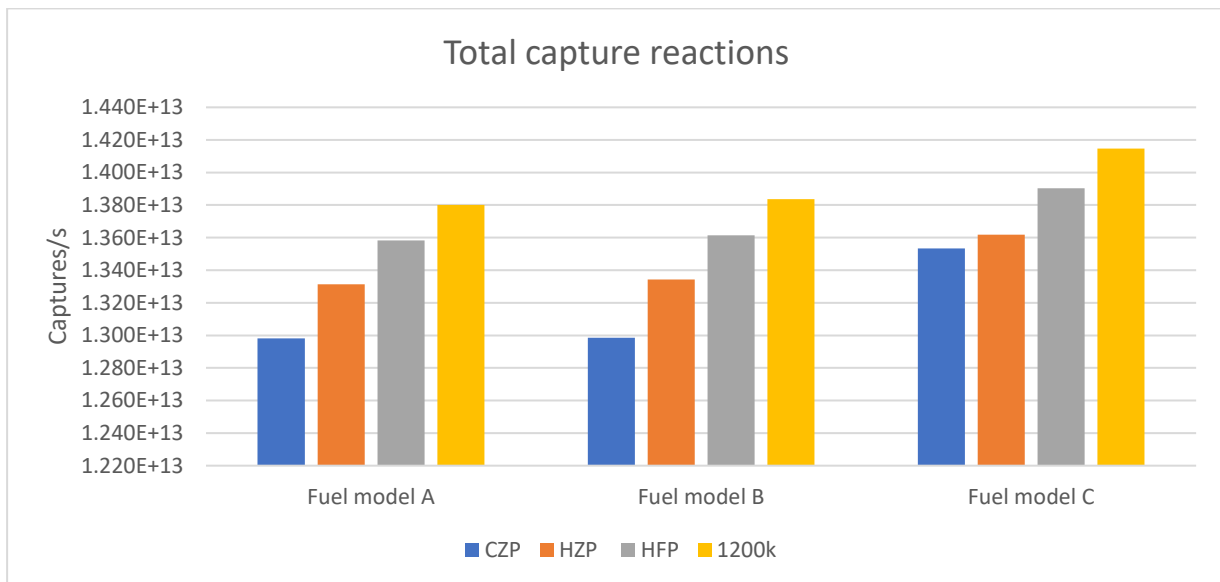


Figure 4-15: Total capture reactions in each fuel model Composition at each fuel temperature condition , CZP (293.6), HZP(600k), HFP(900k) and 1200k- Fuel model A (LEU UO₂) , Fuel model B (LEU Th-UO₂) and Fuel model C (LEU Pu-MOX), all fuel compositions with 0.038wt% Gd₂O₃.

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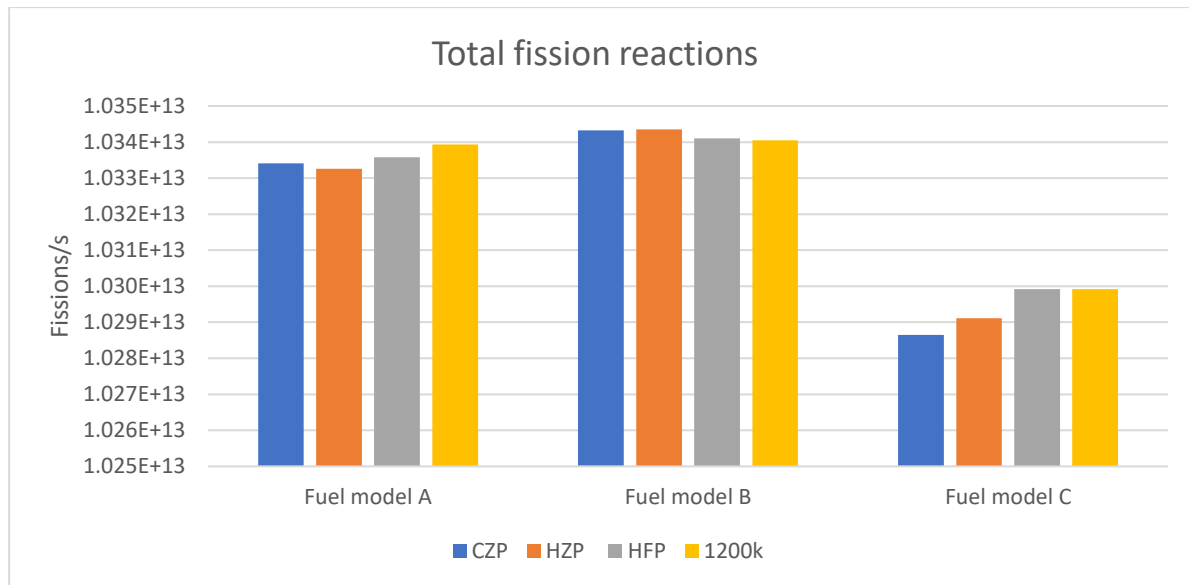


Figure 4-16:Total fission reactions in each Fuel model composition at each fuel temperature condition , CZP (293.6), HZP(600k), HFP(900k) and 1200k- Fuel model A (LEU UO₂), Fuel model B (LEU Th-UO₂) and Fuel model C (LEU Pu-MOX),), all fuel compositions with 0.038wt% Gd₂O₃.

Fuel model composition + 0.038wt% Gd ₂ O ₃	(n, gamma)	Cold zero power (293.6k)	Hot Zero Power (600k)	Hot Full Power (900k)	1200k
Fuel model A: 4.5% UO ₂ (Koeberg reference)	U-238	6.52E+12	6.81E+12	7.05E+12	7.24E+12
Fuel model B: (5% ²³⁵ U, 93% ²³⁸ U & 2% ²³² Th) O ₂	U-238	6.22E+12	6.51E+12	6.74E+12	4.54E+12
	Th-232	3.73E+11	3.95E+11	4.05E+11	4.16E+11
Fuel model C: (0.8%	U-238	6.10E+12	6.30E+12	6.53E+12	6.72E+12

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Pu(PWR), 4.5276 % ²³⁵ U & 94.6724% ²³⁸ U) O ₂	Pu-240	1.53E+12	1.52E+12	1.56E+12	1.59E+12
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The capture reactions significantly increase while the fission reactions slightly decrease with increasing fuel temperature. This is probably because captures happen mostly in the resonances of the fertile material, which increases substantially with increase in temperature, due to the Doppler Effect. Fission, on the other hand, happens mostly in the thermal energy range for the fissile isotopes. Since most of these fissions do not occur in resonances, they are not increased by the Doppler Effect. Furthermore, for some of the dominant fissile isotopes, especially U-235, their microscopic fission cross-sections decrease faster with increasing energy than $1/v$ and, therefore, an increase in the temperature of the thermal neutrons result in a decrease in the fission rate. The opposite is, however, true for the less dominant Pu-239 and Pu-241. **Table 4-23** shows that the capture-to-fission ratio of all fuels increases with increasing fuel temperature, for the reasons just explained. Supporting the results in **Table 4-23** is **Figure 4-15** and **Figure 4-16**. It can be seen that for all fuel model compositions, the rate of neutron captures increases drastically with increasing fuel temperature and sufficiently overpowering the fission rate, this further explains the negative DC feedback. Fuel model C has a higher capture rate than its counterparts at all fuel temperatures and this explains the strong negative DC of Pu-MOX fuels.

4.4.2.3 Doppler Coefficient (DC) subchapter conclusion

The Doppler coefficients of all three fuel compositions fall within the IAEA's required range for all fuel temperature conditions from 293.6K-1200K. Though k_{eff} decreases with increasing fuel temperature, the magnitude of this negative Doppler coefficients decreases with increasing temperature. The Doppler coefficient of the LEU Th- UO_2 composition (Fuel model B) becomes less negative at a much quicker rate than the other fuel compositions. This observation may require further investigation to fully decipher this behaviour. It is seen that PU-MOX fuels typically possess stronger negative doppler coefficients than other fuel compositions, this is evident by comparing the doppler coefficient results of fuel model C (of PU-MOX composition) with the results of the other fuel compositions. This supports observation on the doppler coefficients of PU-MOX fuels in other studies. The capture-fission ratio substantially increases with increasing fuel temperature and this is clear sign of increasing resonance capture with increasing fuel temperatures showing the doppler effect influence and resonance absorption.

It is important to note that the capture to fission ratio given here is not that of individual fissile isotopes normally given in text books, but it is the total fission to capture ratio of all the isotopes in the fuel model, keeping in mind that each fuel model composition has fresh, once burnt and twice burnt fuels. The fresh fuels have fertile and fissile fuel isotopes as well as burnable absorber Gd_2O_3 . On the other hand, the once burnt fuels and the twice burnt fuels have a total of over 40 actinides and over 400 minor-actinides. While the actinides do fission and also capture neutrons, the minor-actinides only capture neutrons and they outnumber the actinides by a factor of about 10.

4.5 Moderator Temperature Coefficient (MTC) “Three- batch full core concept” comparison

This section consists of two parts. The first part shows the MTC results of the fuel model compositions from 500K moderator temperature to 620K (HFP conditions). This is the typical moderator temperature range at normal power producing conditions in a PWR (Du Toit, 2017). This part aims to show and compare the MTC values of fuel model A, fuel model B and fuel model C, at operating condition (HZIP/HFP, 500-600K) then at Boiling (620K).

To fully understand factors that contribute to the MTC being negative or positive, isotopic reaction rates are used and evaluated at three moderator conditions (cold zero power (CZP-296.3K), hot full power (HFP-600k) and at boiling (620k)) and at three neutron energy groups (thermal, epithermal and fast). This is the second part of the MTC analysis, and it starts under section 4.4.4 “Technical evaluation of the MTC”. Following that the MTC is very sensitive to fuel isotopic composition it is therefore very important to understand the contribution of each important isotope on the MTC (Morrison, et al., 2018).

It is important to keep in mind that all the fuel model compositions contain 0.038wt% Gd_2O_3 in the fuel and their respective critical soluble boron concentrations in the moderator.

4.5.1 Interpretation of results

Presented in **Table 4-24** is the magnitude of the MTCs of each Fuel model Composition at the specified temperatures from room temperature/CZP (293.6K) to boiling (620K. **Figure 4-17** shows the general trend of how the MTC of the LEU UO_2 , the LEU Th- UO_2 and the LEU Pu-MOX fuels changes when the moderator temperature changes.

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Table 4-24: The MTC (Moderator Temperature Coefficient) as a function of increasing moderator temperature for fuel models A, fuel model B and fuel model C.

	MTC (pcm/K)		
Temp(K)	Fuel model A: 4.5% UO₂ (Koeberg reference) + 0.038 wt% Gd₂O₃	Fuel model B: (5% ²³⁵U, 93% ²³⁸U & 2% ²³²Th) O₂ + 0.038% Gd₂O₃	Fuel model C: (0.8% Pu(PWR), 4.5276 % ²³⁵U & 94.6724% ²³⁸U) O₂ + 0.038 wt% Gd₂O₃
500	-1.44	-2.44	-5.72
520	-9.85	-10.37	-16.84
540	-12.84	-15.92	-21.64
560	-17.74	-20.90	-27.43
580	-26.94	-29.94	-39.53
600	-45.41	-48.27	-68.04
620	-1991.19	-1953.37	-1593.50

**** MTC recommended range (-70; 0) (IAEA, 2003)**

The results show that the MTCs of all three Fuel model Compositions are negative and fall within the IAEA's required range only from room temperature 293.6K to 600K. A dramatic change in the MTCs can be observed between 600-620K as the MTC of each fuel model composition drops far below -1500 pcm/k. This is caused by a sudden very large loss of moderator density, due to the onset on increased boiling which caused the neutron spectrum to harden due to fewer neutron-hydrogen collisions resulting in increased captures in the resonances of the fertile isotopes and thus a reduction in reactivity. In a finite system it would also have resulted in increased leakage and thus decreasing reactivity (Zainuddin, et al., 2016). The design pressure of a PWR is 155 bar and from thermodynamic properties of water, this pressure can only keep the moderator from boiling only up to a water temperature of about 615K. The moderator density at 620K is only 0.98g/cm³ which is substantially low and a clear indication of boiling and a large void fraction. **Table 4-24** shows that at boiling (620K), the MTC becomes strongly negative. These MTC values seen at boiling are very comparable to fully voided reactivity coefficient (FVR) values seen in a study by (Morrison, et al., 2018). According to (Lamarsh & Baratta, 2001) in a LWR thermal

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reactor, the value of the VRC is very much related to the value of the MTC, though the VRC is measured in SI units pcm/%void, i.e a strongly negative MTC will also result in a strongly negative VRC. The point where the moderator begins to boil and eventually turns into vapor, then this vapor is referred to as the void since its density is much less than the density of the liquid moderator (Lamarsh & Baratta, 2001). Excessive boiling in a PWR can be linked to a LOCA (loss of coolant accident) condition and a strongly negative feedback would be desirable in case it happens (Fridman & Kliem, 2011). However, the MTC and FRV(VRC) should not be confused as they have totally different unit.

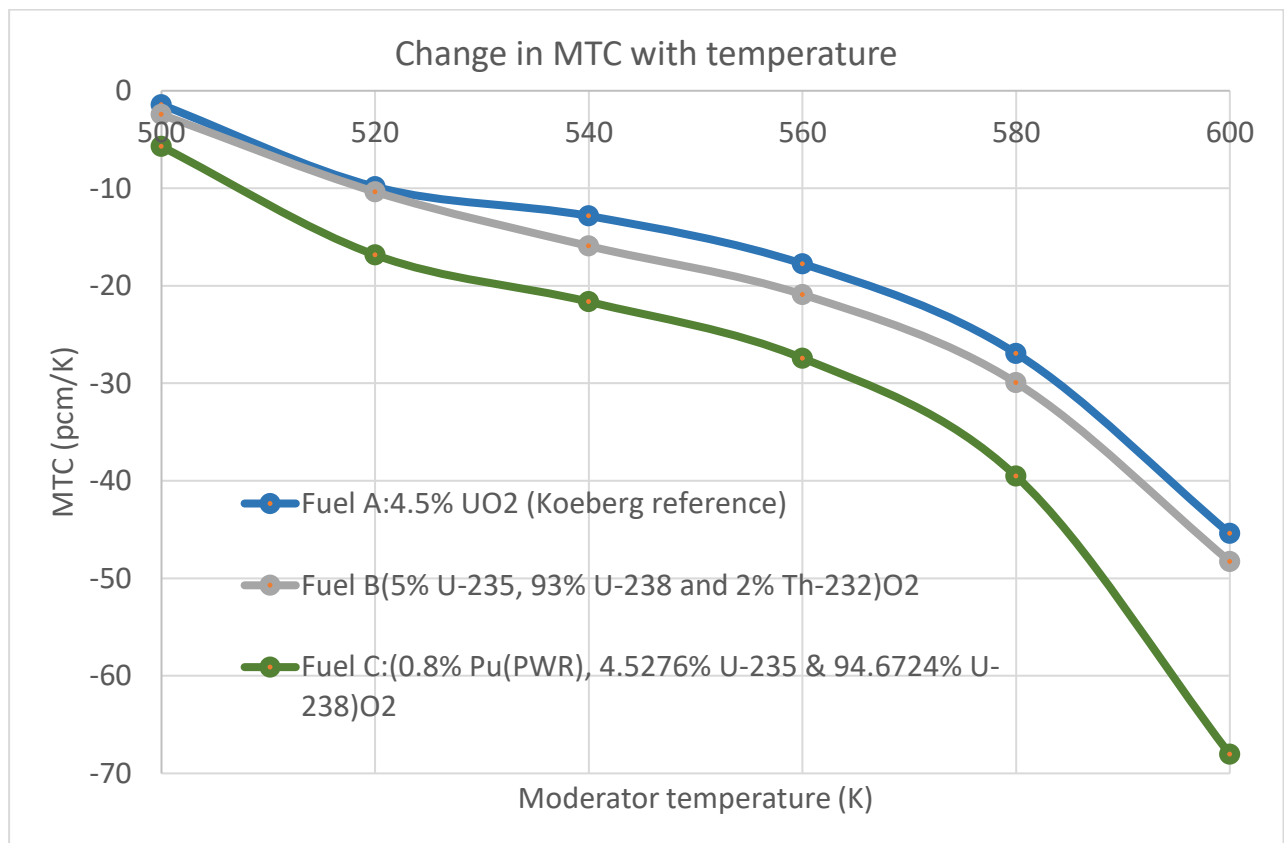


Figure 4-17: Behaviour of the MTC of Fuel model A (LEU UO₂) , Fuel model B (LEU Th-UO₂) and Fuel model C (LEU Pu-MOX) with increasing moderator temperature from room temperature to hot full power.

As seen in **Figure 4-17**, Fuel model C of Pu-MOX composition has a stronger negative MTC and this translates that it is more sensitive to moderator temperature changes in comparison with the counterpart fuels. This could partly be due to Pu-240 resonance peak near 1eV results in a strong negative MTC in Pu-MOX fuels when the spectrum hardens due to a decrease in moderation

(Mahrus & Chang, 2018). Fuel model B has the second largest strongest negative MTC and this could be owing to the large relative resonance absorption in isotope Th-232. The next section (section 4.5.2) delves into isotopes that play a part in the MTC of each fuel model composition.

4.5.2 Technical evaluation of the MTC

This section evaluates the shift in neutron energy spectrum brought about by the decrease in moderator density because of an increase in moderator temperature. Isotopic reaction rate tallies (reactions/cm³shake) are used to evaluate the reactions that take place only in the important isotopes of the three fuel model compositions and in the moderator. The reactions are tallied based on how changes in the moderator temperature conditions (CZP-293.6k, HFP-600k and Boiling-620k) changes the dominant reaction that occur at thermal (below 2eV), epithermal (2eV-2.5keV) and fast (2.5keV-10MeV) energies. The reactions of interest are fission and capture in the fuel, and absorption and scattering in the moderator. The dominant reactions in the moderator which have an effect on the MTC are absorption (n, alpha) by B-10 and both radiative capture and scattering by hydrogen (H-1) (Zainuddin, et al., 2016). If an isotope is not included, it means its contribution is insignificant. Note that all fuel model compositions have burnable absorber gadolinia (0.038wt% Gd₂O₃) and the moderator has soluble boron (H₃BO₃). The isotopic reaction rates in the fuel models are represented as fractional isotopic reaction rates.

- **Fractional isotopic reaction rates:** Reactions (fissions or captures) in an isotope at a particular energy spectrum are given relative to the total absorption reactions (i.e. fissions + captures+ n,alpha) in the fuel model (considering the dominant fuel isotopes + borated moderator) at all energies. The significance of this representation is to show the spectrum dependant fractional fissions or captures occurring in an isotope as a fraction of the total absorptions in the fuel and moderator model.

It should be noted that reaction tallies may contain substantial statistical errors, which may result in large relative errors, but might still give a fair enough representation of the reactions occurring in a model (Pelowitz, et al., 2014).

4.5.2.1 Thermal and resonance absorption in fuel and moderator

This section gives the tallied energy spectrum dependant fission and capture reaction rates in the dominant isotopes found in the fresh, once burnt and twice burnt fuels of the fuel model compositions at the three moderator temperature conditions (CZP-Cold Zero Power, HFP- Hot Full Power and LOCA). The aim is to compare how reaction rates change based on moderator temperature conditions and the contribution of each isotope in the fuel to bring about a negative or positive MTC at each condition.

Literature says when the moderator temperature increases and consequently the moderator density decreases, the neutron spectrum is substantially impacted and shifts to higher energies. Looking at the three moderator temperature conditions of interest, it is seen that at CZP (room temperature, 293.6 K) the neutron spectrum is concentrated at thermal energies. When the moderator temperature increases to HFP (600 K) conditions the Maxwellian tail of the thermal neutron spectrum shifts slightly higher up in the thermal window. Due to the effects already explained above, thermal absorptions decrease slightly, while absorptions at epithermal energies (Resonance absorption) increases. When the moderator starts to boil (in this case at 620 K), the change in the thermal neutron energy spectrum stops, as the temperature will not increase above boiling point. However, the decrease in the moderator density, and thus the increase in the epithermal captures, accelerate strongly.

Dominant reactions which are common in all three fuel types are fissions in isotope U-235 and captures in U-238 and this is because a large portion of the fuels is comprised of these isotopes. Considering the three-batch fuel model concept, it is important to remember actinides and minor-actinides are present in the once burnt and twice burnt fuels of all fuel model compositions. The dominant actinide is plutonium and its various isotopes, mainly Pu-239 and Pu-241, and the fission and capture contribution of these isotopes is very significant to the MTC of all fuel model compositions. Spent LWR fuel which was burnt to 42MWd/kg-U produces plutonium with a isotopic vector of approximately, 2% Pu-238, 53% Pu-239, 25% Pu-240, 15% Pu-241 and 5% Pu-242 (Bjork, 2012) (WNO, 2018). This closely correlates to the Pu vector which was used in the present study, namely 2.763% Pu-238, 53.755% Pu-239, 23.615% Pu-240, 13.108% Pu-241, and 6.758% Pu-242 (Erlank, 2017). It is assumed here that whatever the mass of the plutonium arising in the once and twice burnt fuels of all three models has the aforementioned isotopic vector. The 0.8% Pu(PWR) (reactor grade plutonium) contained in the fresh fuel of the LEU Pu-MOX fuel is of the same isotopic vector.

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Gadolinia is also present in all fuel compositions and the capture reactions in the dominant isotope Gd-155 and Gd-157 are therefore given. Gadolinia absorption decreases with increasing moderator temperature as seen in all three fuel types. This effect can add a positive reactivity and contributes to the MTC being less negative (Zainuddin, et al., 2016).

The absorption behaviour of important moderator isotopes B-10 (n, alpha) and H-1 (capture) are considered the specified energies and at specified moderator temperature conditions.

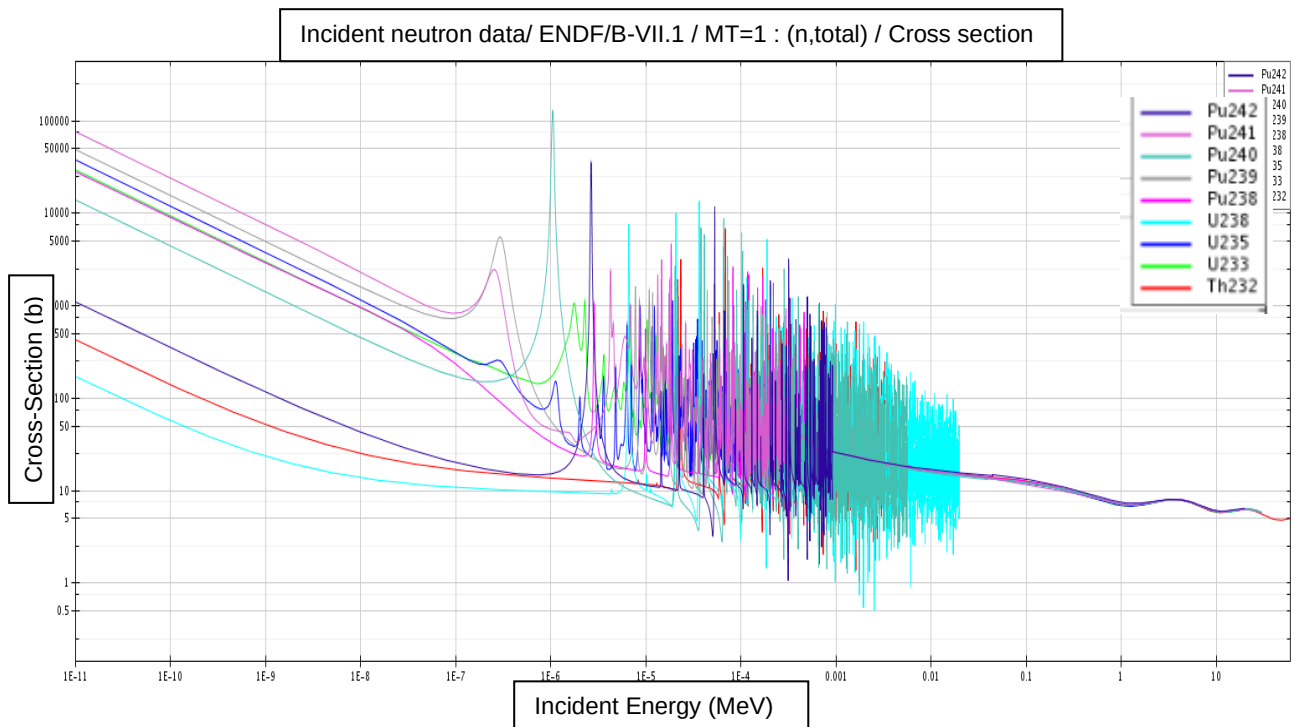


Figure 4-18: Plot showing how the total cross-section of various important isotopes found in Fuel Model A, Fuel Model B and Fuel Model C varies with incident neutron energy.

Depicted in **Figure 4-18** above is total cross-section of the important fissile and fertile isotopes that could be found in all fuel model compositions. We know that the odd isotopes are the fissile isotopes and the dominant reaction in the fissile isotopes is fission which mostly occurs at thermal energies. The even isotopes are the fertile isotopes and we know that the dominant reaction which occur in fertile isotopes is neutron capture at thermal and epithermal energies and only a small number of fissions at fast energies. This figure supports the findings of the results which show the reaction rate fractions in the important isotopes in each fuel model composition which follow in the next sections of this study.

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Figure 4-19 below depicts the absorption reactions which occur in the important isotopes in the borated moderator, the (n,alpha) reaction in B-10 (boron 10) and the captures reactions in H-1. The total cross-section of H-1 is also depicted in the figure and the aim of this is to show the difference between the total capture cross-section in H-1 and the total reaction cross-section. From what we know is that the dominant reaction between neutrons and the H-1 atom is scattering reaction because that's how water actually moderates neutrons, through H-1 scattering reactions with neutrons. This figure (see **Figure 4-19**) and helps understand the behaviour of reactions occurring in the borated moderator and how they change with increasing moderator temperature (a decrease in moderator density). This figure further supports the findings in the results which follow in the next sections of this study which depict how the isotopic reaction rate fractions in B-10 and H-1 change with a change in moderator temperature conditions from CZP to boiling.

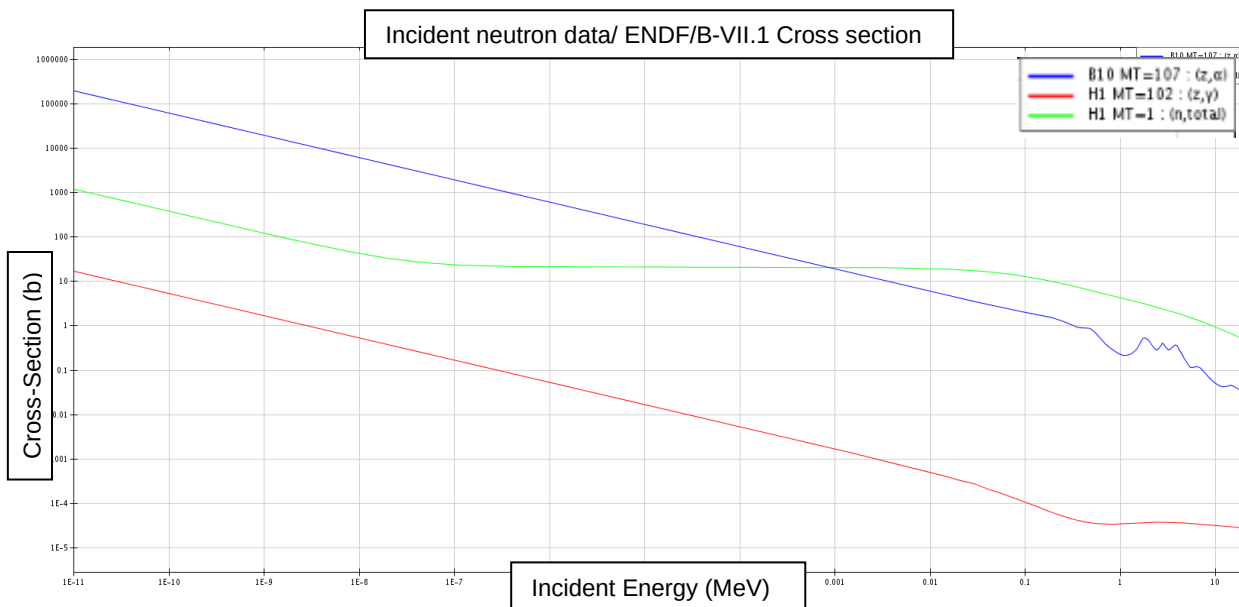


Figure 4-19: Cross-section vs incident neutron plot showing (n,alpha) in B-10 (blue), capture in H-1 (red) and the total neutron absorption cross-sections H-1(green).

4.5.2.1.1 Fuel model A: 4.5% UO_2 (Koeberg reference) + 0.038wt% Gd_2O_3 - Tallied spectrum shift reaction rates with moderator conditions

Given in **Figure 4-20**, **Figure 4-21** and **Figure 4-22** is the energy spectrum dependant fractional fission and capture reaction rates in the dominant isotopes of Fuel model A at CZP (Cold Zero Power), HFP (Hot Full Power) and at boiling (620K) respectively. These are absorptions reaction rates in the fuel and the moderator.

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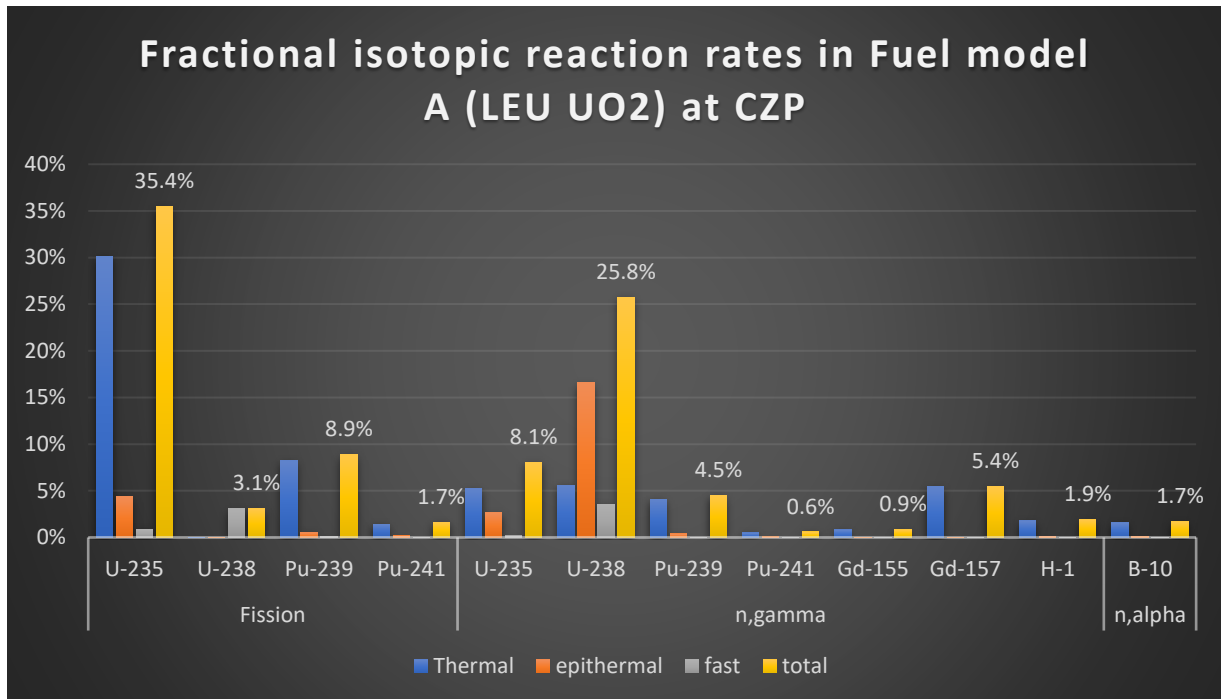


Figure 4-20 : Tallied fractional isotopic reaction rates for isotopes in Fuel model A (4.5% UO₂ (Koeberg reference)) with moderator at Cold Zero Power 293.6 K,(1.003g/cc).

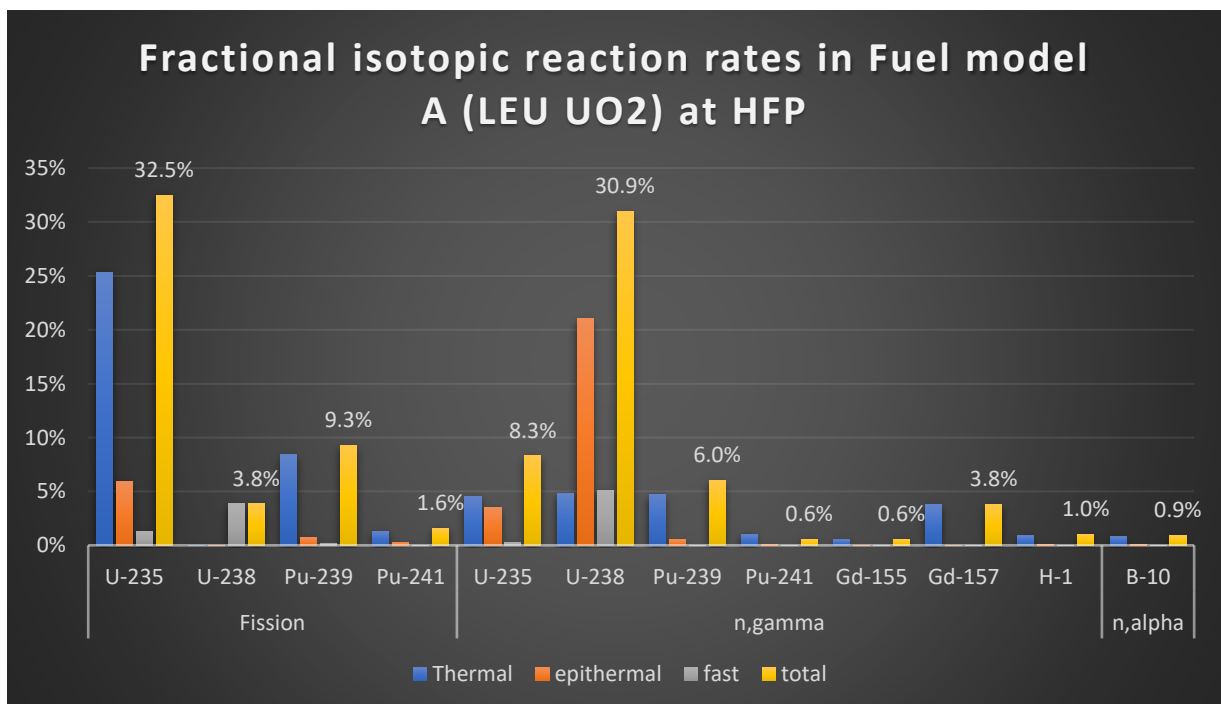


Figure 4-21: Tallied fractional isotopic reaction rates of isotopes in Fuel model A (4.5% UO₂ (Koeberg reference)) with moderator at hot full power (HFP) 600K, (0.661g/cc).

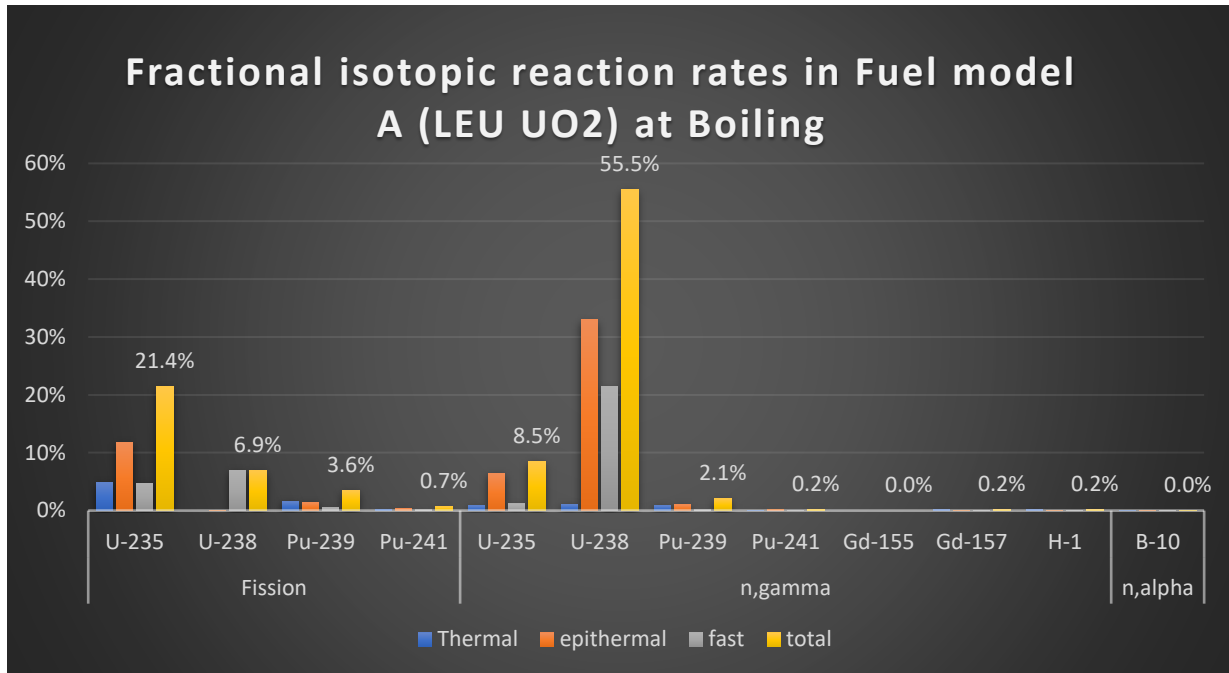


Figure 4-22: Talled fractional isotopic reaction rates of isotopes in Fuel model A (4.5% UO₂ (Koeberg reference) with moderator at boiling (620k), (0.098g/cc).

With reference to **Figure 4-20**, **Figure 4-21** and **Figure 4-22**:

The uranium isotopes, fissile U-235 and fertile U-238 play a very significant role in determining the fuel's behaviour with changes in moderator temperatures. The total fission rate fraction in U-235 decreases when the moderator temperature increases. The fissions decrease from 35.4% at CZP to 32.5% at HFP. This gives a negative contribution to the MTC. At boiling the U-235 fission rate fraction decrease to 21.4%. The largest fission rate fraction decrease in U-235 is at thermal energies. This agrees with the decreasing non-1/v factors for fission (g_f) for U-235 in Table 3.2 of (Lamarsh & Baratta, 2001). The capture rate fraction in U-235 increases with increasing moderator temperature, from 8.1% at CZP to 8.3% at HFP contributing to a negative MTC. At boiling the capture rate fraction increases to 8.5% adding a negative reactivity.

The capture rate fraction of U-238 significantly increases with increasing moderator temperature. From 25.8% at CZP to 30.9% at HFP. This increase gives a negative contribution to the MTC. The overall capture rate fraction changes by 29.7% in U-238 as the capture rate fraction increases from 25.8% at CZP to 55.5% at boiling. As seen in the figures, it is clear that the largest capture rate fraction increases in U-238 is at epithermal energies and this is because U-238 has a wide resonance width which absorbs neutrons at a much higher rate than at any other energy window

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(Dobuchi, et al., 2016). The fact that U-235 fissions substantially decrease while U-238 captures substantially increase with increasing moderator temperatures shows that a negative reactivity is added, and this contributes to the MTC being negative.

It is evident from the results that there is a significant amount of plutonium in the fuel model and this can be seen from the fission and capture contribution by the fissile Pu-239 and Pu-241. These arise from the once burnt and twice burnt fuels and were bred from the fresh fuel of the LEU UO_2 . The fission rate contribution of these isotopes is larger than the capture contribution meaning these isotopes add a positive reactivity which makes the MTC less negative.

The total fission rate fraction of Pu-239 slightly increases from 8.9% at CZP to 9.3% at HFP, adding a positive reactivity which makes the MTC less negative. At boiling the fission rate fraction of Pu-239 substantially decreases to 3.6%, which is more than half the rate at HFP and this adds a larger negative reactivity compared to the reactivity added at HFP. The Pu-239 captures behave similar to the fissions when the moderator condition changes from CZP to HFP where they increase from 4.5% to 6% respectively. A plausible reason for the increase in total fission and capture rate fraction in Pu-239 is the resonance peak around 0.4eV. The behaviour of Pu-239 agrees with the increasing non- $1/v$ factors for fission (g_f) and overall for absorption (g_a) as in Table 3.2 of (Lamarsh & Baratta, 2001)

The Pu-241 fission rate fraction slightly decreases with increasing moderator temperature from 1.7% at CZP to 1.6% at HFP then substantially to 0.7% at boiling. This observed behaviour by Pu-241 adds a small negative reactivity which contributing to a negative MTC from CZP to HFP.

The role played by both gadolinium isotope Gd-157 is quite significant considering the amount of gadolinia in the fuel (0.038wt%). The fractional thermal capture rate in the isotope substantially decreases with increasing moderator temperature and according to (Zainuddin, et al., 2016), this effect can potentially make the MTC positive depending on the Gd_2O_3 content in the fuel. Thermal captures dominate in Gd-157 the total capture rate fraction decreases from 5.4% at CZP to 3.8% at HFP then to 0.2% at boiling. Gd-155 decreases in a similar manner too but its capture rate fraction is 6 times less than that of Gd-157.

The burnable absorber B-10 fractional absorption (n,α) rate decreases similar to capture of the two gadolinium isotopes. From 1.7% at CZP to 0.9% at HFP then to 0% at boiling. This is because burnable absorbers B-10 has a high absorption (n,α) cross-section at thermal energies that is $1/v$ dependant (see **Figure 4-19**). A shift in the neutron energy spectrum caused

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by decreased moderation sees the burnable absorbers capture rate significantly decreasing because of less soluble boron per unit volume as a results of reduced moderator density.

The parasitic absorption of water also plays a significant role as in the isotope H-1 whose capture decreases from 1.9% at CZP to 0.2% at boiling. According to (Zainuddin, et al., 2016), these decrease in absorption (capture) by H-1 can add a slightly positive reactivity which contributes just a little to making the MTC less negative.

4.5.2.1.2 Fuel model B: (5% ^{235}U , 93% ^{238}U & 2% ^{232}Th) O_2 + 0.038wt% Gd_2O_3 -Tallied spectrum shift reaction with moderator conditions

Figure 4-23 , **Figure 4-24** and **Figure 4-25** is the energy spectrum dependant fractional fission and capture reaction rates in the dominant isotopes of Fuel model B at CZP (Cold Zero Power), HFP (Hot Full Power) and at boiling (620K) respectively. These are absorptions reaction rates in the fuel and the moderator.

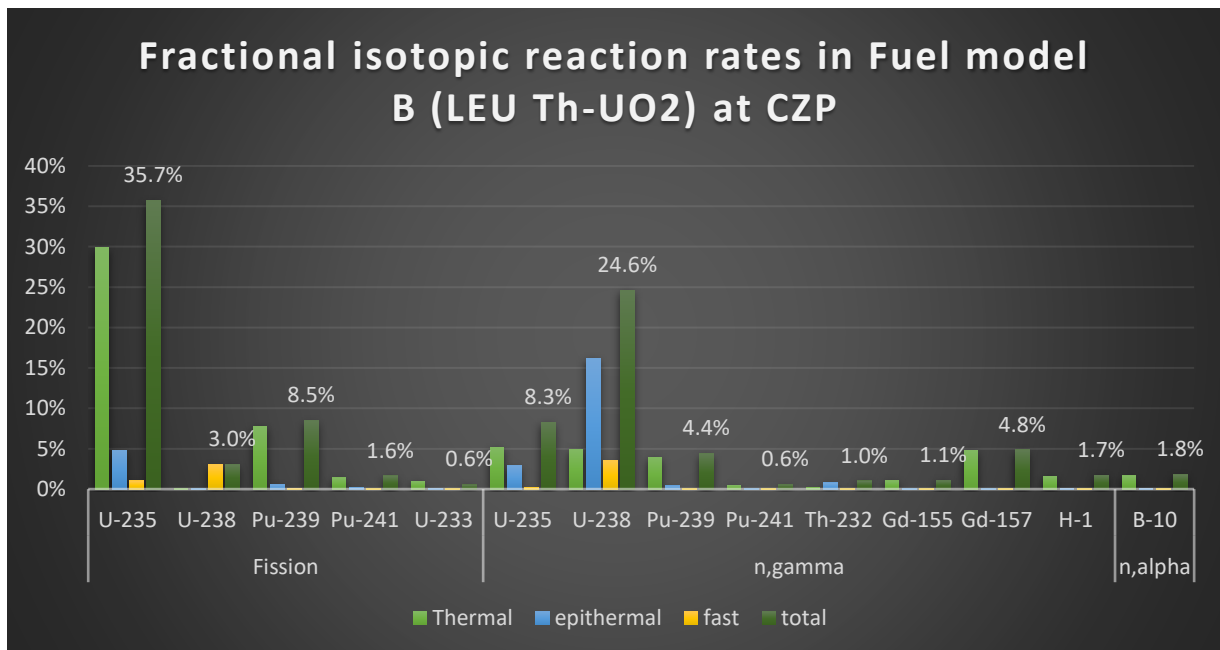


Figure 4-23: Tallied Fractional isotopic reaction rates of isotopes in Fuel model B ((5% ^{235}U , 93% ^{238}U & 2% ^{232}Th) O_2) with moderator at Cold Zero Power 293.6 (1.003g/cc).

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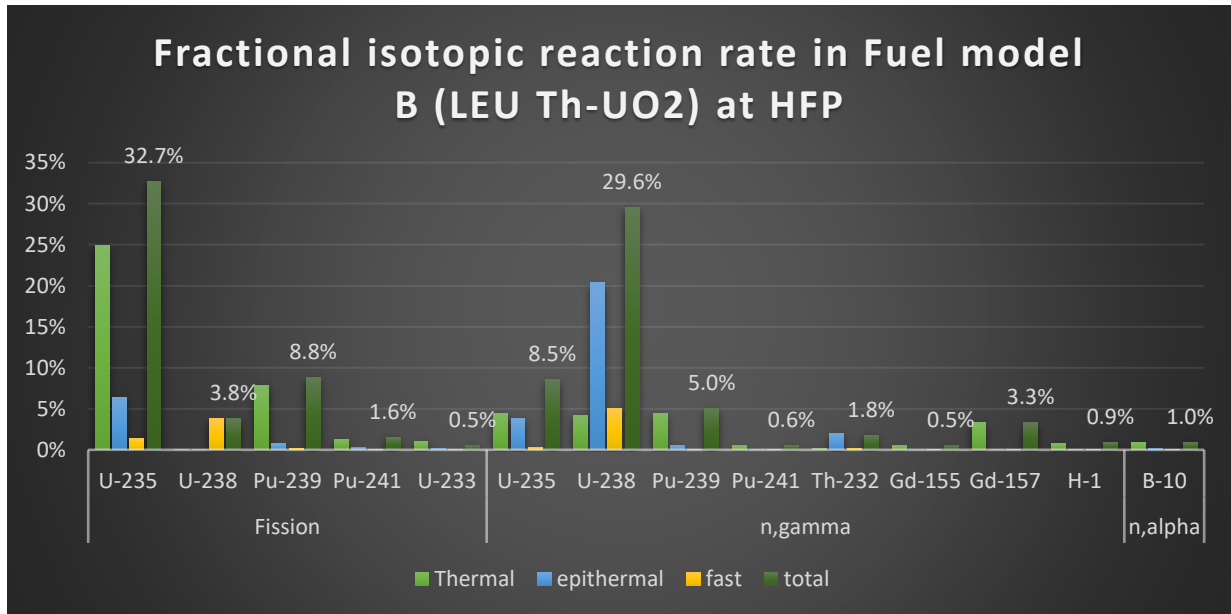


Figure 4-24: Tallied fractional isotopic reaction rates of isotopes in Fuel model B ((5% ²³⁵U, 93% ²³⁸U & 2% ²³²Th)O₂) with moderator at Hot Full Power, 600k (0.661g/cc).

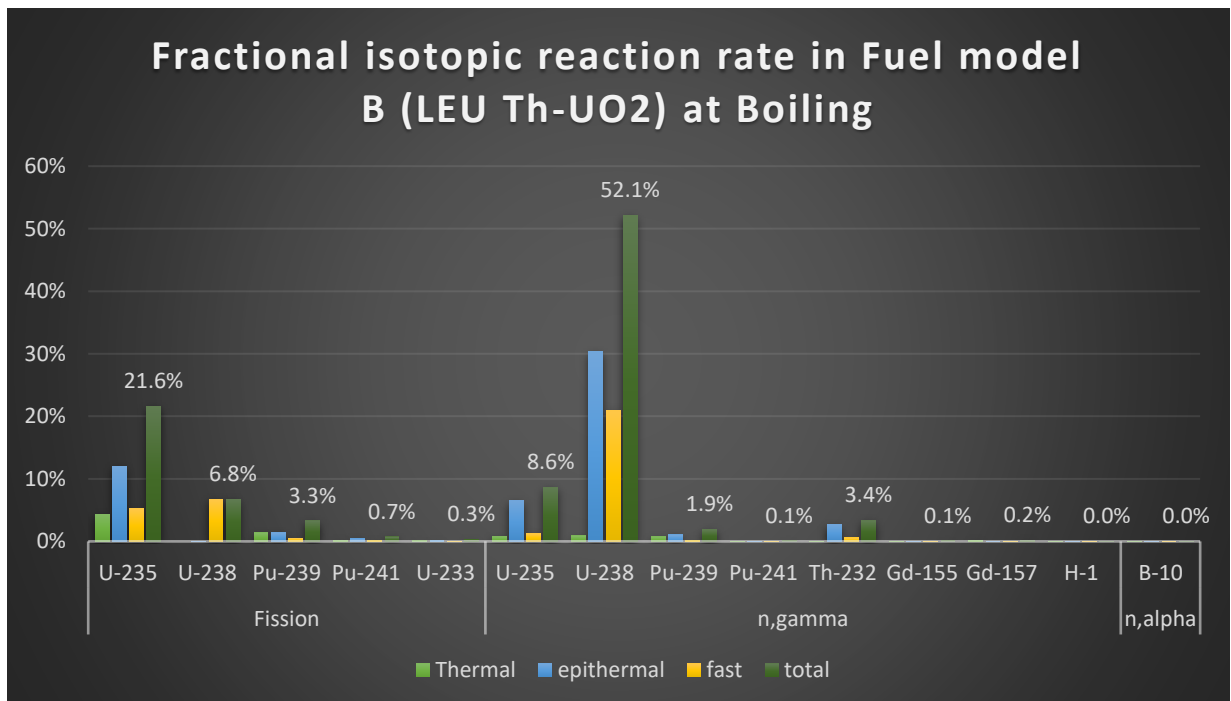


Figure 4-25: Tallied fractional isotopic reaction rates of isotopes in Fuel model B ((5% ²³⁵U, 93% ²³⁸U & 2% ²³²Th) O₂) with moderator at boiling (620k),(0.098g/cc).

The influence of thorium, uranium and plutonium fuel mixture composition on the reactivity coefficients of pressurised water reactors

With reference to **Figure 4-23** , **Figure 4-24** and **Figure 4-25**:

The total fission rate fraction in U-235 significantly decrease with increasing moderator temperature. The fission rate fraction decreases from 35.7% at CZP to 32.7% at HFP contributing to a negative MTC. The fission rate probability drastically decreases to 21.6% at Boiling. The overall decrease in fission rate fraction from CZP to boiling is 14.1%, which is 0.1% much higher than the decrease in fuel model A. This mostly owing to a decrease in thermal fissions. The capture rate fraction in U-235 slightly increases with increasing moderator temperature, from 8.3% at CZP to 8.5% at HFP and this behaviour contributes to the MTC being negative. At boiling the capture rate fraction increases to 8.6%.

The total capture rate fraction in U-238 substantially increases with increasing moderator temperature, from 24.6% at CZP to 29.6% at HFP adding a negative reactivity contributing to a negative MTC. The capture rate fraction then drastically increases to 52.1% at boiling. The overall increase from CZP to boiling is 27.5% and this increase in captures adds a negative reactivity. The largest increase contributing to the total captures is the epithermal captures. The fissions in U-238 increases from 3% at CZP to 3.8% at HFP, and this slight increase in fissions makes MTC less negative. The fission rate fraction further increases to 6.8% at boiling. It can be seen in the figures that the fission rate fractions in U-238 increases at fast energies.

While the fast fissions in U-238 increase with increasing moderator temperature, the fast captures increase too at a higher rate and therefore off-set the fission contribution.

The capture rate fraction in Th-232 at epithermal energies is quite significant considering that the fuel only contains 2% Th-232. It can be seen that the total fractional capture rate in Th-232 increases by a significant 2.4% from CZP to boiling. The capture rate fraction increases from 1% at CZP to 1.8 % at HFP, this behaviour by Th-232 adds a negative reactivity which contributes to the MTC being negative. The capture rate fraction in Th-232 then increases to 3.4% at boiling. It can be seen in the figures that the largest capture increase in Th-232 is at epithermal energies-represented by the blue bars.

Then just like in Fuel model A, fissile plutonium isotopes Pu-239 and Pu-241 make significant contributions too, to the reactivity of the fuel model . The fission rate in Pu-239 slightly increases with increasing moderator temperatures from CZP to HFP from 8.5% to 8.8% slightly adding a positive reactivity which makes the MTC less negative. The fission rate probability then substantially decreases to 3.3% at boiling. This substantial decrease in fission rate fraction adds a negative reactivity. The capture rate fraction behaves as the fission rate fraction, it increases

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from 4.4% at CZP to 5% at HFP and decreases from 5% to 1.9% at boiling. Overall, the fissile Pu-239 adds a positive reactivity which makes the MTC less negative at HFP.

Fissile Pu-241 fission rate remains somewhat constant at CZP and at HFP with a fission rate fraction of 1.6%. The fission rate fraction substantially decreases to 0.7% boiling.

According to (Zainuddin, et al., 2016), burnable absorbers can add in a positive MTC because they absorb better at thermal energies and would therefore absorb less when the spectrum shift due to a decrease in moderator density resulting from an increase in moderator temperature. As seen in the figures, the capture rate in burnable absorbers B-10 (n, alpha), Gd-155 and Gd-157 does decrease with increasing moderator temperatures. The capture rate fraction in Gd-157 decreases from 4.8% at CZP to 3.8% at HFP. In Gd-155 the capture rate fraction decreases from 1.1% at CZP to 0.5% at HFP. The absorption (n,alpha) by B-10 decrease from 1.8% at CZP to 1% at HFP.

The parasitic absorptions in H-1 decreases from 1.7% at CZP to 0.9% at HFP, then to 0% at boiling.

4.5.2.1.3 Fuel model C: (0.8% Pu(PWR), 4.5276% ²³⁵U & 94.6724% ²³⁸U) O₂ + 0.038wt% Gd₂O₃ -Tallied spectrum shift reactions with moderator conditions

Figure 4-26, Figure 4-27 and Figure 4-28 is the energy spectrum dependant fractional fission and capture reaction rates in the dominant isotopes of Fuel model C at CZP (Cold Zero Power), HFP (Hot Full Power) and at boiling (620K) respectively. These are absorptions reaction rates in the fuel and the moderator

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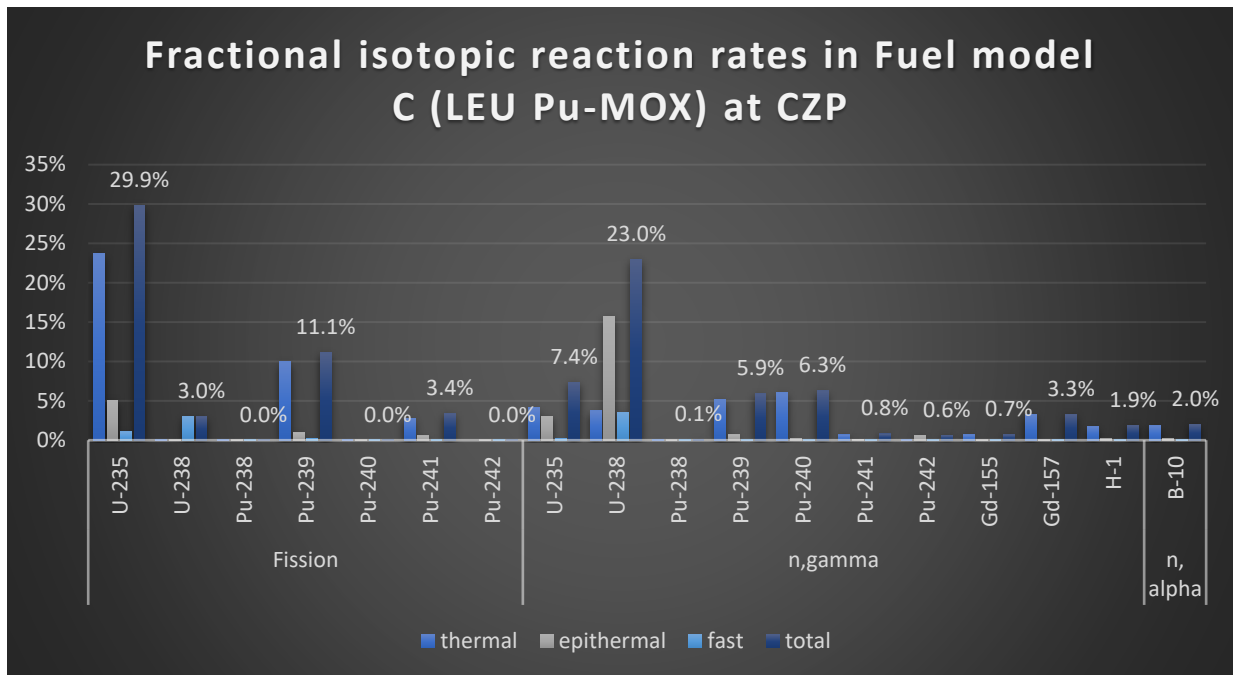


Figure 4-26: Tallied fractional isotopic reaction rates of isotopes in Fuel model C (0.8% Pu(PWR), 4.5276 % ^{235}U & 94.6724 % ^{238}U) with moderator at Cold Zero Power (293.6k), (1.003g/cc).

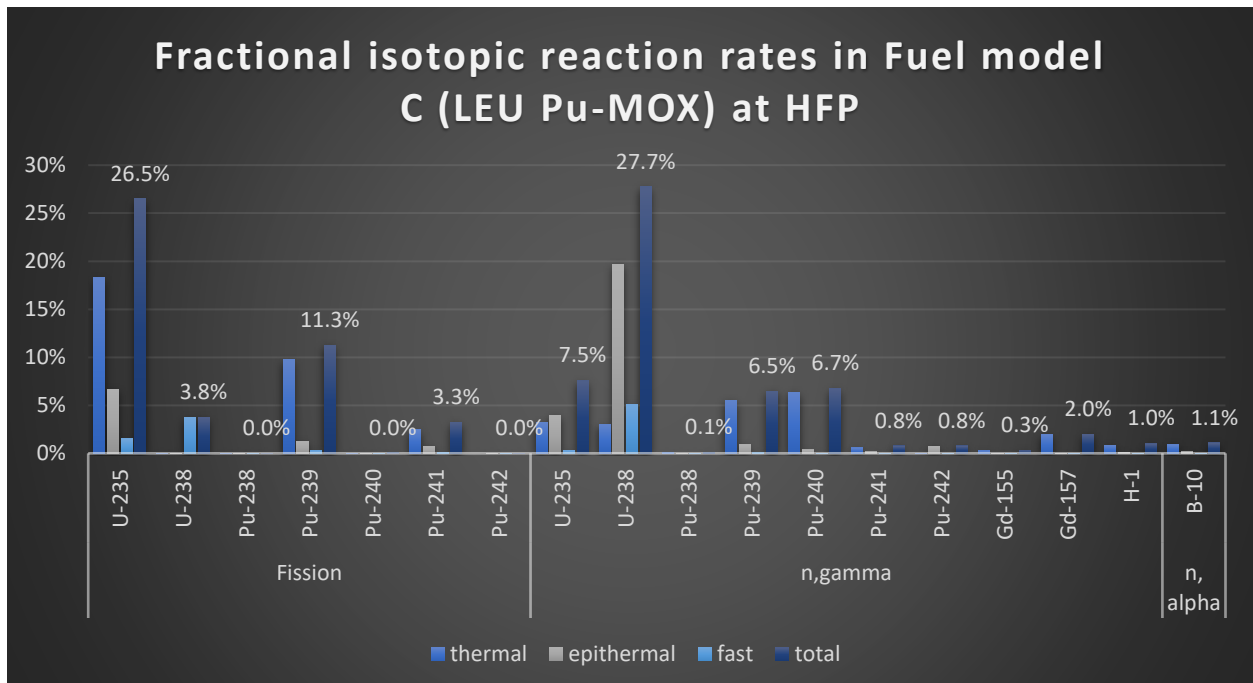


Figure 4-27: Tallied fractional isotopic reaction rates of isotopes in Fuel model C (0.8% Pu(PWR), 4.5276 % ^{235}U & 94.6724% ^{238}U) with moderator at Hot Full Power (600k), (0.661g/cc).

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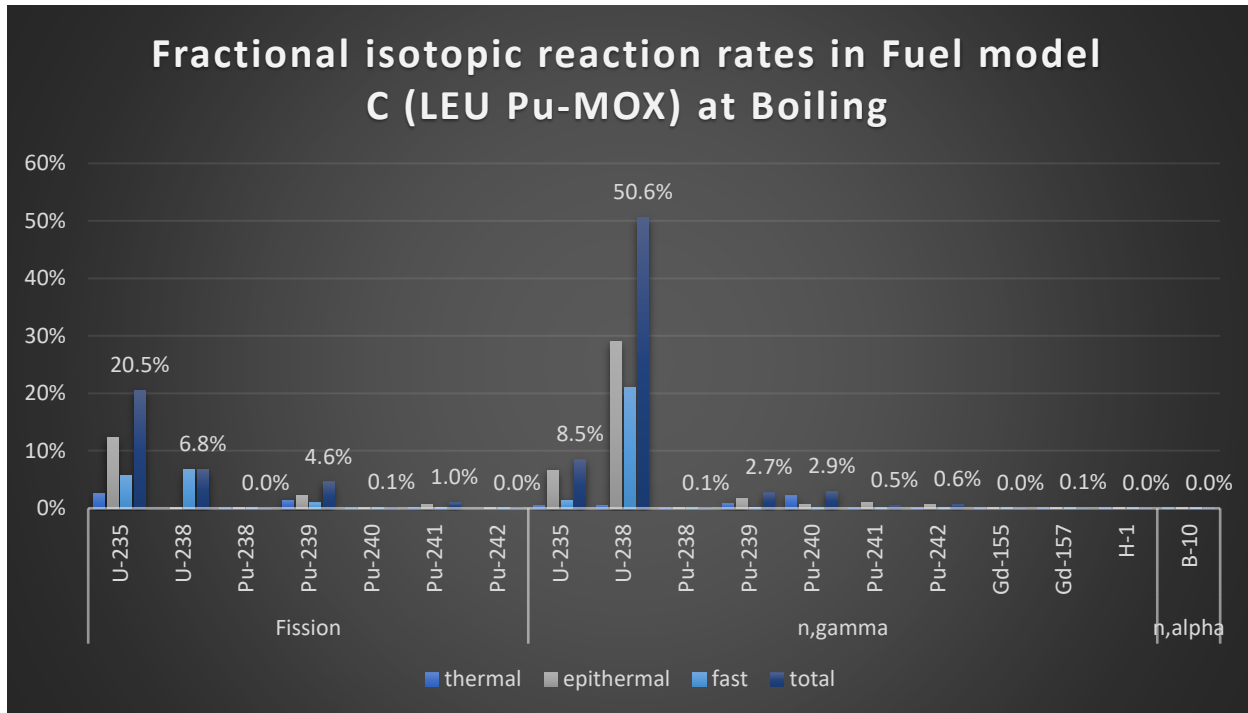


Figure 4-28: Talled fractional isotopic reaction rates of isotopes in Fuel model C(0.8% Pu(PWR), 4.5276 % ^{235}U & 94.6724% ^{238}U) with moderator at boiling - 620k (0.098g/cc).

With reference to **Figure 4-26**, **Figure 4-27** and **Figure 4-28**:

This fuel contains 0.8% Pu(PWR) of vector Pu, 2.763% Pu-238, 53.755% Pu-239, 23.615% Pu-240, 13.108% Pu-241, and 6.758% Pu-242, in addition to the plutonium arising from the once and twice burnt fuels.

Because this is predominantly a plutonium fuel model, reaction rate fractions of plutonium isotopes are much higher than in the other fuel models. In this fuel not only do the fissile plutonium isotopes (Pu-239 and Pu-241) contribute greatly to the MTC but, fertile isotopes Pu-240 makes a significant contribution too.

Fissile U-235 and Fertile U-238 behaviour in this fuel model is no different from the behaviour in the other two models. The thermal fission fraction in U-235 substantially decreases with increasing moderator temperature, while the epithermal captures in U-238 significantly increase with increasing moderator temperature. A significant difference in the behaviour of U-235 in this fuel model is that its fission rate fraction decreases at a relatively smaller rate compared to the decrease in the other fuel models. The total U-235 fission rate fractions decreases from 29.9% at CZP to 26.5% at HFP, that's a fission rate decrease of 3.4%. And at boiling the fission rate fraction is 20.50% and the overall change from CZP to boiling is only 9.4%. The capture rate fraction in

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U-235 behaves opposite to the fission rate fraction. The capture rate fraction slightly increases with increasing moderator temperature, from 7.4% at CZP to 7.5% at HFP. This effect contributes to a negative MTC. At boiling the capture rate fraction in U-235 further increases to 8.5% (1.1% increases from CZP), adding a negative reactivity. The capture rate fraction in U-235 increases at epithermal energies.

As for fertile U-238, its capture rate fraction increases from 23.0% at CZP to 27.7% at HFP, an increase of 4.7%. This increases in capture adds a negative reactivity and contributes to a negative MTC. At boiling the capture rate fraction drastically increases to 50.6%, which is an overall increase in capture rate of 27.6% which is comparable to the change of fuel model B (27.5%). It is clear from the results that the fission rate fraction in U-238 increases with increasing moderator temperatures and the increase is at fast energies. An increase in fissions adds a positive reactivity, but the overall negative contribution by U-238 off-sets the positive contribution.

Fissile Pu-239 and Pu-241 behave in a similar manner too as in the other fuel models, but their fission and capture rate contributions are at a slightly higher rate because of a larger quantity of plutonium in this fuel model. The total fission rate in Pu-239 increases slightly adding a small positive reactivity at HFP because Pu-239 has a non- $1/v$ factor for fission (g_f) which increases with increasing temperature provided that the reactor is still well moderated (Lamarsh & Baratta, 2001). Since a reactor operates at HFP, the moderator still sufficiently moderates though its density is only about 67% then. The fission rate fraction in Pu-239 increases from 11.1% at CZP to 11.3% at HFP. This effect will add a small positive reactivity thus making the MTC less negative. The total fission rate fraction in Pu-239 decreases to 4.6% at boiling and this effect adds a negative reactivity. The overall fission rate fraction change in Pu-239 from CZP to boiling is 6.5%.

Fissile Pu-241 behaves different from Pu-239. The fission rate probability of Pu-241 slightly decreases with increasing moderator temperature from 3.4% at CZP to 3.3% at HFP (a decrease of 0.1%). This effect adds a negative reactivity. At boiling the fission rate fraction is 1.02% and the overall change from CZP to boiling is 2.4%. At thermal energies, exactly below 1eV the fissile isotopes Pu-239 and Pu-241 have a fission peak as well as a capture peak which compete for neutrons (Erlank, 2017), but the fission peak is slightly stronger than the capture peak and this can be seen by the relatively higher thermal fission fraction in both fissile isotopes.

Due to a relatively larger plutonium quantity in this fuel model, fertile plutonium isotope Pu-240 comes into play and makes quite a significant contribution to the reactivity and therefore the MTC.

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It is seen in the figures (**Figure 4-26**, **Figure 4-27** and **Figure 4-28**) that the total capture rate fraction in Pu-240 substantially increases with moderator temperature from 6.3% at CZP to 6.7% at HFP. The capture contribution by Pu-240 adds a negative reactivity and therefore contributes to a negative MTC. Fertile isotope Pu-240 has its biggest radiative capture peak around 1eV (see **Figure 4-18**). At boiling the capture rate fraction of Pu-240 drops to 2.9%, an overall change(decrease) of 3.4% from CZP to boiling. This decrease in capture adds a sizeable amount of positive reactivity.

The capture rate fraction by Gd-157 is lower than in the other fuel models at all moderator temperature conditions. The capture rate fraction in Gd-157 decreases from 3.3% (CZP) to 2% (HFP). A significant large decrease can be seen at boiling where the capture rate is 0.1%. The capture rate fraction in Gd-155 is so small is it can be ignored. The (n,alpha) rate fraction in B-10 decreases from 2% at CZP to 1.1% at HFP. Then at boiling there's a relatively large decrease to 0.0% (n,alpha). Parasitic absorption in H-1 decreases from 1.9% at CZP to 1.0% at HFP then to 0.0% at boiling.

4.5.2.2 Leakage (Hydrogen, H-1)

Presented in **Table 4-25** are the tallied scattering reaction rates in each fuel model composition at different moderator temperature conditions given in units (scattering reactions/ cm³/shake). A shake in MCNP is a unit of time and it is equivalent to 1x10⁻⁸ seconds (10⁻⁸ s) (Pelowitz, et al., 2014).

Table 4-25: Tallied neutron-hydrogen scattering reactions (reactions/cm³/shake) in the moderator of various fuel model compositions at different moderator temperature conditions.

	CZP	HZP	Boiling
Fuel model A (LEU UO ₂)	23.3613	19.83849	8.42876
Fuel model B (LEU Th-UO ₂)	22.64968	19.25185	8.17568
Fuel model C (LEU Pu-MOX)	21.31004	18.14855	7.8504

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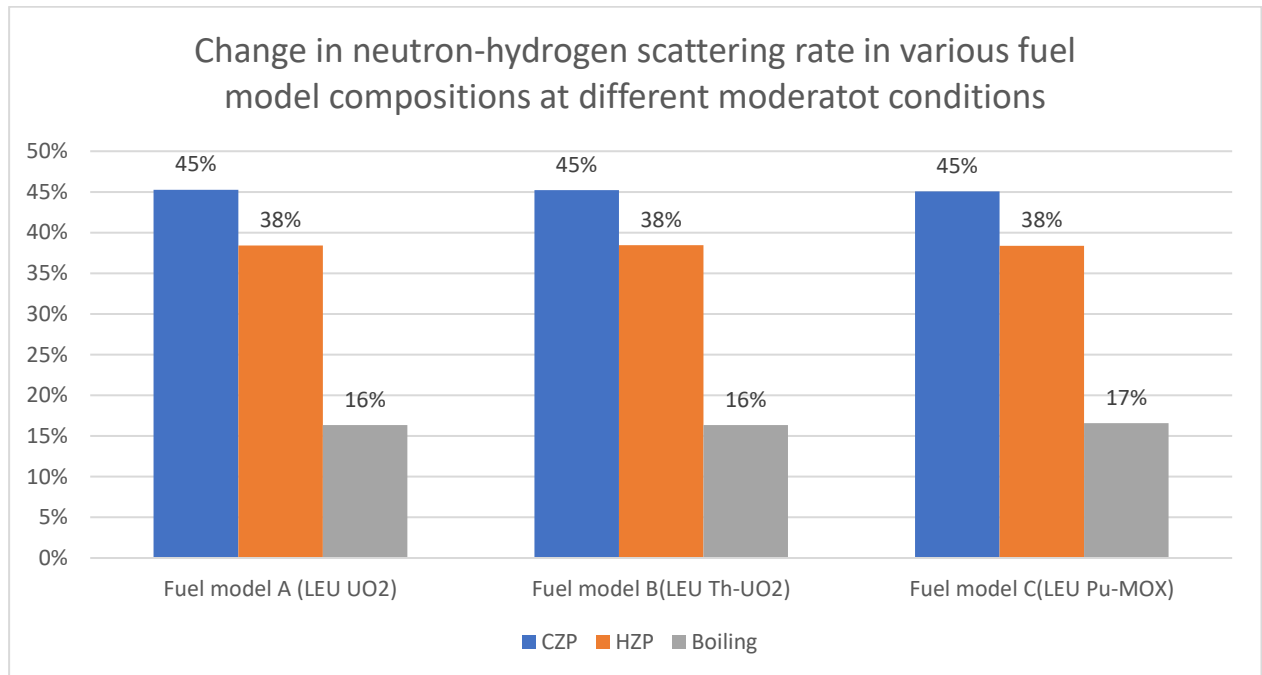


Figure 4-29: The relative percentage rate of neutron-hydrogen scattering collisions with change in moderator conditions in Fuel model A (LEU UO₂), Fuel model B (LEU Th-UO₂) and Fuel model C (LEU Pu-MOX).

All three fuel model compositions exhibit similar scattering behaviour in H-1 at all moderator temperature conditions as seen in energies and at all moderator temperature conditions as seen in **Figure 4-29**. This figure presents the relative percentage rate of neutron-hydrogen scattering rate at one moderator temperature condition relative to the total of the scattering at all moderator temperature conditions. for example, scattering rate at CZP relative to the subtotal scattering at all moderator temperature conditions (CZP +HFP + LOCA).

Generally, **Figure 4-29** indicates that as the moderator density decreases with increasing moderator temperature scattering collisions decrease and this effect hardens the neutron energy spectrum (Stacey, 2007). Overall this effect decreases the number of thermal neutrons and consequently decreases important thermal fissions and therefore inserting a negative reactivity. With the moderator insufficient to slow down neutrons at this point, this decreases the number of thermal neutrons moving from one fuel pin to another, decreasing the number of thermal fissions and increasing resonance captures. Both a decrease in thermal fissions, and neutron leakages add a negative reactivity and should results in a negative MTC.

4.5.3 MTC subsection conclusion

The MTC of all fuel compositions is negative at all moderator temperature conditions and all the MTC values are within the IAEA's recommended ranges except at boiling (620K) where they're on average about 20-30 times less negative. The MTC becomes substantially strongly negative with increasing moderator temperature. This is mainly due to the large effect of the moderator density on the reactivity of a water moderated thermal reactor (Lamarsh & Baratta, 2001).

The dominant fuel isotopes, U-235 and U-238 behave similar in all fuel compositions. Thermal fissions in U-235 decrease (and therefore total fissions) and epithermal captures (and therefore total captures) in U-238 increase with increasing moderator temperatures. This joint behaviour by the uranium isotopes adds a negative reactivity and contributes to the MTC being negative. In Fuel model C (The LEU Pu-MOX fuel) the fission rate fraction in U-235 doesn't decrease at a similar rate with increasing moderator temperature as in the other fuel models. The U-235 fissions in this model decrease at a much smaller rate in comparison – by 9% on average from CZP to HFP compared to 14% in fuel model A and Fuel model B. The nature of this observed behaviour needs to be investigated further.

The plutonium fissile isotopes behave similar in all fuel model compositions. Fissile Pu-239 adds a positive reactivity which makes the MTC less negative because of the increasing fission rate fraction in the isotope from CZP to HFP in all fuel model compositions. The capture rate fraction in Pu-239 increases too when the fission rate fraction increases, but the fission overpower the captures resulting in a net positive reactivity contribution. A substantial decrease in the fissions in Pu-239 is seen at boiling and this adds a negative reactivity. Pu-241 behaves similar in all fuel models where the fission and capture rate fractions decrease with increasing moderator temperature. The overall contribution by Pu-241 is a negative reactivity which contributes to the MTC being negative.

The burnable absorbers, Gd-155 and Gd-157 have a capture dominant absorption cross-section which is $1/v$ dependant (see **Figure 2-2**) and from the results it can be seen that the capture rate fraction in both Gd-155 and Gd-157 substantially decreases with increasing moderator temperature. And the absorption cross-sections in burnable absorber B-10 (n, alpha absorption) and H-1 (capture) which are also $1/v$ dependant (see **Figure 4-19**) decrease with increasing moderator temperature. According to (Zainuddin, et al., 2016), this behaviour by the burnable absorbers and H-1 adds a little positive reactivity making the MTC less negative.

5 Conclusion and recommendations

5.1 Conclusions

This study was based on a research gap found in a study where various fuel composition mixtures of uranium, thorium and plutonium were designed to increase the fuel cycle length of the Koeberg PWR. The fuels were successfully designed, but their reactivity coefficients which are important safety parameters in any reactor were unknown and therefore needed to be investigated. The aim was to implement a safety analysis study of the new designed fuels in terms of investigating their reactivity coefficients and see how they compare or differ from that of the LEU (4.5% enrichment) UO_2 fuel used to fuel the actual Koeberg PWR.

An extensive literature review was conducted which served as a cornerstone for carrying out this study. Research methods were formulated from the literature gathered and were used as guidelines. An infinite unit cell fuel pin model was then developed to simulate a conceptual PWR core with a three-batch refuelling scheme of fresh, once burnt and twice burnt fuel in MCNP. In the model three fuel mixture compositions of LEU UO_2 , LEU Th- UO_2 and LEU Pu-MOX were simulated and their characteristics were investigated.

From this study, the effects different burnable absorbers have on the initial reactivity of different fuel compositions could be understood. Gadolinia drastically decreases the initial reactivity of the fuels, but the biggest change in reactivity was seen in the LEU UO_2 fuel and the least change in reactivity in the LEU Pu-MOX fuel. The soluble boron concentration added to reach criticality was proportional to the fissile content of the fuels. The LEU Pu-MOX fuel has a higher fissile content which amounts to 5.0625% because of the fissile plutonium isotopes Pu-239 and Pu-241 with an abundance of 53.755% and 13.108% in the fresh fuel respectively.

The results clearly show how sensitive safety parameters of a reactor are to changes in temperature. Though the Doppler coefficient is the most important according to regulatory bodies, the MTC proves to be as equally important based on the fact that it is the most sensitive to temperature changes and more than likely it is a parameter that would limit heavy metal fissile loading in reactors (Morrison, et al., 2018).

It was discovered in this study that if there's no sufficient cooling in a reactor core, a less than 20K increase in moderator temperature to a reactor that is already at HFP (600K) could result to excessive boiling and eventually a fully voided core. At boiling the VRC/FRV becomes relevant and not the MTC. At moderator temperatures between 600K-620K significant boiling started to occur resulting in a total void fraction of 85%.

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The Doppler coefficients of plutonium fuel are more favourable owing to a large individual capture to fission ratio of fissile plutonium material and owing to the large capture peak in the fertile plutonium isotope Pu-240 at 1eV. A justifiable explanation for the large Doppler coefficient of LEU Th- UO_2 as opposes to LEU UO_2 fuel is the stronger doppler effect of Th-232 at elevated temperatures which surpasses that of U-238. At this stage it is unclear as to why the MTC of the LEU Pu-MOX fuel and the LEU Th- UO_2 fuels are strongly negative, but the same isotopes contributing to a negative Doppler coefficient mainly Pu-240 in LEU Pu-MOX and Th-232 in LEU Th- UO_2 are seen coming into play when the moderator temperature increases add a negative reactivity.

It can be seen that adding a small amount of Th-232 and a small amount of Pu(PWR) plutonium not only potentially improves the economy of nuclear power plant, but further improves the inherent safety of a reactor making it commercially attractive. It can be concluded that plutonium fuel of composition 0.8% Pu(PWR), 4.5276% U-235 and 94.6724% (LEU Pu-MOX), is the best fuel for increasing the fuel cycle length of a PWR and Though a further extensive feasibility analysis of the fuel model is required.

Overall the general aims, and objectives of the study were met, and the research problem addressed.

5.2 Recommendations

Recommendations for further developments with regards to the methods and recommendations for further studies.

Methods and models

The methods used in this study included assumptions and estimations in some instances as and as the current model was very limiting. Further extensive research involving more sophisticated research methods and tools is needed firstly to validate the fuel compositions drawn from the study by (Erlank, 2017), which are conceptual by nature and secondly to eliminate the assumptions made in this study which were influenced by the model itself. And with those eliminations, a more comprehensive study can be achieved.

In this study a portion of a fuel pin is used to represent a whole reactor core which is of complex geometry and contains many materials and structural components compared to a fuel pin. To simulate a full core with a three-batch refuelling scheme of fresh, once burnt and twice burnt fuel more accurately, at least a model resembling at least a $\frac{1}{4}$ of a full core is required.

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Further the model was very limiting, and estimations and assumptions had to be made. To accurately simulate Gd_2O_3 at least a portion of a fuel assembly is required considering that only 12 out of 264 fuel pins contain this integral burnable absorber. Furthermore, Gd_2O_3 is not the only burnable absorber used in the fuel assembly, but a few of the fuel pins in the assembly would contain ZrB_2 (zirconium diboride), which is a thin layer coated on the surface of the fuel pellet. And in a fuel assembly there're 24 guide tubes for the insertion of another type of burnable absorber B4C in the form control rods.

Further studies

A few related areas of the study could not be covered due to tight time constraints and therefore it will be ideal to develop the study further and investigate these areas with the current model or using more advanced models.

The reactivity coefficients could not be analysed as a function of burn-up to see the effects thereof. Further research could investigate the temperature reactivity coefficients as a function of burn-up to see the changes and effects at the middle of cycle and end of cycle and then compare these with the beginning of cycle.

More fuel mixture compositions of uranium, plutonium and thorium could be investigated. Firstly, the current MOX fuel model compositions only contained small amounts of thorium and Pu(PWR) reactor grade plutonium. Fuel compositions with more heavy metal loadings of thorium and Pu(PWR) could be simulated to see their influences on the reactivity coefficients and analyse the reactivity coefficients as a function of thorium, Pu(PWR) and fissile U-235 loading and how they compare with the currently investigated fuel model compositions. With the Pu(PWR) loading and the U-235 enrichments, it is recommended to stay within the allowable limits for a PWR. Secondly, more diverse fuel composition mixtures could be investigated, such as those incorporating uranium, plutonium and thorium all at once, and those incorporating weapon grade plutonium.

Analyse the doppler coefficients under severe operational conditions such as temperatures near the melting point of the fuels, to investigate the influences of things like voiding that takes place when the fuel melts and therefore the influences on the doppler coefficients. The MTC could also be investigated as a function of soluble boron to analyse the effects thereof and track the points where soluble boron concentration makes the MTC positive.

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From the burnup results certain information pertaining the actinides and minor-actinides inventory at the end of the fuel cycle of each fuel mixture composition was obtained and gathered. With this information a full proliferation risk study of the current fuel compositions could be carried out.

This recommended further study may require at least a fuel assembly model to completely investigate. A beginning of life feasibility analysis and full safety analysis of the fuel compositions could be carried out to investigate aspects such as the control rod worth, the shutdown margin and the prompt and delayed neutron fractions of the fuel compositions. Also using special plugins within the MCNP code, axial and radial hot channel factors of the fuels could also be investigated to complete the feasibility and safety analysis.

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