

**The influence of thorium on the temperature reactivity
coefficient in a 400 MWth pebble bed high temperature
plutonium incinerator reactor**

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**Project-based mini-dissertation (Option C) for the partial fulfilment of the
requirements for the degree Master of Engineering at the Postgraduate School
of Nuclear Science, Potchefstroom Campus of the North-West University**

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November 2012

Acknowledgements

The author wishes to thank the Research Centre Jülich and the Pebble Bed Modular Reactor (Pty) Ltd for allowing the use of their VSOP 99/05 simulation code and the associated input models for the PBMR DPP-400 reactor.

This study is dedicated to Marili Richards, for her enduring love and support.

Abstract**Title:**

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Social and environmental justice for a growing and developing global population requires significant increases in energy use. A possible means of contributing to this energy increase is to incinerate plutonium from spent fuel of pressurised light water reactors (Pu(PWR)) in high-temperature reactors such as the Pebble Bed Modular Reactor Demonstration Power Plant 400 MWth (PBMR-DPP-400). Previous studies showed that at low temperatures a 3 g Pu(PWR) loading per fuel sphere or less had a positive uniform temperature reactivity coefficient (UTC) in a PBMR DPP-400. The licensing of this fuel design is consequently unlikely. In the present study it was shown by diffusion simulations of the neutronics, using VSOP-99/05, that there is a fuel design containing thorium and plutonium that achieves a negative maximum UTC. Further, a fuel design containing 12 g Pu(PWR) loading per fuel sphere achieved a negative maximum UTC as well as the other PBMR (Ltd.) safety limits of maximum power per fuel sphere, fast fluence and maximum temperatures. It is proposed that the low average thermal neutron flux, caused by reduced moderation and increased absorption of thermal neutrons due to the higher plutonium loading, is responsible for these effects. However, to fully understand the mechanisms involved a detailed quantitative analysis of the roll of each factor is required. A 12 g Pu(PWR) loading per fuel sphere analysis shows a burn-up of 180.7 GWd/tHM which is approximately double the proposed PBMR (Ltd.) low enriched uranium fuel burn-up. The spent fuel has only a decrease of 24.5 % in the Pu content which is sub-optimal with respect to proliferation and waste disposal objectives. Incinerating Pu(PWR) in the PBMR-DPP 400 MWth is potentially licensable and economically feasible and should be considered for application by industry.

Keywords:

High Temperature Gas-cooled Reactor, HTGR, HTR, PBMR DPP-400, VSOP-99/05, reactor grade plutonium, uniform temperature reactivity coefficient (UTC)

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List of Symbols and Acronyms

α_T	Temperature Reactivity Coefficient
Am	Americium
CANDU	Canada Deuterium Uranium
CP	Coated Particle
DLOFC	Depressurised Loss of Forced Cooling
E	Neutron Energy
FTC	Fuel Temperature Reactivity Coefficient
FZJ	Research Centre Jülich
GWd	Gigawatt day
HTR-10	High Temperature Reactor 10MWth (Prototype pebble bed reactor at Tsinghua University, China)
HTR-MODUL	High Temperature Reactor Module (German design)
HTR-PM	High Temperature Reactor Pebble Bed Module (250MWth Chinese design)
HM	Heavy Metal
IAEA	International Atomic Energy Agency
k_{eff}	Effective Multiplication Factor
k_{∞}	Infinite Multiplication Factor
LEU	Low Enriched Uranium
MA	Minor Actinides
MTC	Moderator Temperature Reactivity Coefficient
Np	Neptunium
OTC	Overall Temperature Reactivity Coefficient
PBR	Pebble Bed Reactor
PBMR DPP-400	Pebble Bed Modular Reactor Demonstration Power Plant 400 MW thermal
PHWR	Pressurised Heavy Water Reactor
Pu	Plutonium
Pu_{HM}	Sum of all Pu isotopes, excluding ^{238}Pu , as defined by the VSOP HM accounting.
$\text{Pu}(\text{PWR})$	Reactor-grade plutonium from the spent fuel of LEU-fuelled PWRs
$\text{Pu}(\text{WGR})$	Weapons Grade Plutonium
PWR	Pressurised (Light) Water Reactor
TRISO	Tri-structural Isotropic
UTC	Uniform Temperature Reactivity Coefficient
U	Uranium
VSOP	Very Superior Old Programs
VSOP-99/05	VSOP-99, version 5 – Developed by FZJ

1 Introduction

1.1 Definitions of terms

1.1.1 Pebble bed high temperature reactor

A pebble bed high temperature reactor is a gas cooled, graphite moderated reactor. The 400 MWth Pebble Bed Modular Reactor Demonstration Power Plant (PBMR DPP-400) design, developed by PBMR (Pty.) Ltd., is helium cooled and is rated at 400 MW thermal.

The PBMR DPP-400 fuelling is continuous which limits the excess reactivity required to maintain criticality and it is multi-pass which enables flattening of the axial power profile. The relatively flat axial power profile allows high operating temperatures (900°C). The power conversion system of the PBMR DPP-400 is a direct helium Brayton cycle for electricity generation. The PBMR DPP-400 is able to decrease and restore load between 100% and 40% power output which enables electricity demand load following. The high temperature of the PBMR DPP-400 facilitates utilisation of the PBMR DPP-400 for supplying chemical processes heat, gasification of coal, desalination of sea water and district heating.

1.1.2 Reactor grade and weapons grade plutonium

Reactor grade plutonium is generated in conventional LEU-fuelled water-cooled nuclear reactors. Reactors with very short refuelling intervals are used to generate weapons grade plutonium. The difference between the various grades of plutonium lies in the composition fractions of the plutonium isotopes, i.e. ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu and ^{242}Pu , where weapons grade plutonium consists of mostly ^{239}Pu , which is denatured by less than 7% ^{240}Pu , while reactor grade plutonium is degraded by much higher fractions of ^{240}Pu and also by substantial fractions of ^{238}Pu , and ^{242}Pu (Serfontein, 2011: Table 23). The odd numbered isotopes are fissionable and the even ones fertile. Burning of plutonium produces similar amounts of energy per fission as uranium. Plutonium can be burnt to generate electricity and in the process be destroyed or dispositioned.

1.1.3 Fertile thorium

Thorium is a fertile nuclear fuel which means that ^{232}Th can be bred to ^{233}U which is fissile. Similar to ^{238}U , ^{232}Th captures neutrons mainly in its epithermal resonances. However, the capture resonances for ^{232}Th are fewer and thus ^{232}Th captures only about 25 % of the neutrons that an equal atom density of ^{238}U would. However using ^{238}U means that some ^{239}Pu will be generated.

1.1.4 Doppler broadening effect

Due to the Doppler broadening effect, as the temperature increases in safe reactor designs the total number of neutron captures increase and thus the resonance escape probability decreases, which decreases the thermal neutron flux, which in turn decreases the reactivity. This is because the resonances broaden and flatten (Doppler broadening) and thus the capture cross-sections at neutron energies on either side of the resonances increase. Thus, although the area below the resonance peak remains the same after broadening, due to a smaller suppression of the neutron flux in the lower and broader capture resonance peak, more neutrons are absorbed in these broadened resonances (Lamarsh & Baratta, 2001:367).

1.1.5 Temperature reactivity coefficient

The multiplication factor which is denoted by the symbol k is defined (Lamarsh & Baratta, 2001:117) as the number of fissions in one generation divided by the number of fissions in a previous generation:

$$k = \frac{\text{number of fissions in one generation}}{\text{number of fissions in the previous generations}}$$

As the number of neutrons is dependent on the number of fissions, k is also defined as the number of neutrons born in one generation divided by the number of neutrons absorbed in the previous generation.

The infinite multiplication factor k_{∞} is the multiplication factor in a reactor of infinite size. The effective multiplication factor k_{eff} is the multiplication factor of a reactor of finite size such as the PBMR-DPP 400.

Reactivity is denoted by the symbol ρ and is defined (Lamarsh & Baratta, 2001:336) as:

$$\rho = \frac{k - 1}{k}$$

The temperature reactivity coefficient is denoted by α_T and is defined (Lamarsh & Baratta, 2001:365) as:

$$\alpha_T = \frac{d\rho}{dT}$$

This can be simplified to:

$$\alpha_T = \frac{1}{k^2} \frac{dk}{dT}$$

The effective multiplication factor (k_{eff}) is applicable to this mini-dissertation as the PBMR DPP-400 is a finite reactor.

The components – the fuel, moderator, internal and external reflectors - of a reactor are not all at the same temperature. A temperature coefficient is specific to the component of a reactor where the temperature change takes place resulting in the fuel temperature reactivity coefficient (FTC), moderator temperature reactivity coefficient (MTC) and reflector temperature reactivity coefficients (RTC) which assume that the external and internal reflectors are at the same temperature. The uniform temperature reactivity coefficient is when the fuel and the moderator temperatures are varied together (UTC) (Serfontein, 2011: Section 3.4.1). The overall temperature reactivity coefficient (OTC) is the reactivity coefficient where the fuel temperature, moderator temperature and the reflector temperature are varied together. The physical interpretation of temperature coefficients should be based on the limits of the theoretical definitions.

Starting with a critical reactor, an increase in temperature in a reactor with a positive temperature reactivity coefficient will result in a supercritical state that will amplify itself until mechanical failure, external intervention or the rise in temperature creates a reduction in reactivity large enough to terminate the supercritical power excursion. Also, if the reactor has a positive temperature coefficient, a decrease in temperature will result in a sub-critical state

until the reactor shuts down or an intervention or internal mechanism restores criticality. The alternative to the unstable positive temperature coefficient is a stable negative temperature coefficient where the reactor, in the event of an increase in temperature, will decrease its reactivity and power and thus counter the temperature increase. As the fuel is where heat is generated, the FTC is more important to maintain stability, as external interventions take a relatively long time to implement. Therefore the FTC is also called the prompt temperature reactivity coefficient. In the United States of America, the Nuclear Regulatory Commission (NRC) will not license a reactor with a positive prompt temperature reactivity coefficient (Lamarsh & Baratta, 2001:367).

1.1.6 Multiplication factor analysis

The four factor formula defines the infinite multiplication factor (k_{∞}) (Lamarsh & Baratta, 2001:286) :

$$k_{\infty} = \eta_T f p \epsilon$$

- η_T is “the average number of neutrons emitted per thermal neutron absorbed in the fissile fuel nuclides”.
- The thermal utilisation fuel factor (f) is the fraction of thermal neutrons that are absorbed by the fuel nuclides. The remainder is absorbed by the moderator or absorbers in the reactor.
- The fast fission factor (ϵ) is defined as “the ratio of the total number of fission neutrons produced by both fast and thermal to the number produced by the thermal fissions alone”. As the fuel kernels are small in high temperature reactors, the number of fast fissions is low and this factor is very close to one. As a result it has a negligible effect.
- The resonance escape probability (p) is that fraction of neutrons that is absorbed, mostly in the capture resonances of fertile nuclides such as ^{238}U , ^{232}Th , ^{240}P or ^{242}Pu , while “slowing down” from fast energies to thermal energies.

An increase in η_T results in the emission of more fission neutrons. If the dominating consequence of an increase in temperature is the increase in η_T , the UTC of the reactor considered would thus be positive.

To determine the k_{eff} it is also necessary to know what neutron percentage leaks out of the reactor to the surroundings as well as what fraction is captured in the fission products. The six factor formula defines k_{eff} and is as follows (Lamarsh & Baratta, 2001:290):

$$k_{\text{eff}} = k_{\infty} P_T P_F$$

The leakage is expressed in two terms:

- the probability, P_F , that thermal neutrons will leak; and
- the probability, P_T , that fast neutrons will leak.

VSOP-99/05 only calculates the product of the thermal and fast probabilities. This product is the fraction of neutrons remaining and in this study termed as the “non-leakage” probability. The effective multiplication factor is the product of the four factor formula multiplied by the non-leakage probability.

1.2 Problem statement

There is increasing global demand for energy to achieve social and environmental justice for a growing and developing global population. Utilisation of conventional nuclear power technology exacerbates the existing and growing risk that the plutonium generated by these reactors will be utilised for nuclear weapons. There is also the possibility that the highly radioactive waste from conventional reactors containing plutonium will not be adequately managed, causing environmental contamination, which may increase the health risk for exposed individuals.

Plutonium that is not incinerated or denatured requires maximum security to reduce the weapons proliferation potential. The management of plutonium currently makes nuclear power more expensive. Development of technologies that incinerate plutonium may make nuclear power even more competitive in energy markets.

High temperature nuclear reactor technology is potentially a suitable and appropriate means to incinerate plutonium generated in conventional reactors. High temperature reactor technology can be used to incinerate this plutonium while producing electricity, process heat or electricity and process heat simultaneously.

However, as determined by Serfontein, at low temperatures a PBMR DPP-400, fuelled with 3 g or less of Pu(PWR) loading per fuel sphere only, has a positive maximum temperature reactivity coefficient (Serfontein, 2011: Section 3.4.2.2.4). This fuel cycle and reactor design is consequently non-licensable.

The safety limits of PBMR (Pty.) Ltd. for the PBMR DPP-400 are:

1. The uniform temperature coefficient (UTC) should be negative for the full range of temperatures. The temperature range is from ambient to 1650°C.
2. The maximum power per fuel sphere shall be less than 4.5 kW/fuel sphere.
3. The fast fluence ($E > 0.1\text{MeV}$) shall be less than $8.0 \times 10^{21} / \text{cm}^2$.
4. The maximum fuel temperature shall be less than 1130°C.

The problem to be solved is to determine a fuel design containing thorium and plutonium or plutonium only where the maximum UTC is negative for the full range of temperatures as well as meeting the PBMR (Pty.) Ltd. safety limits.

1.3 Aims

1.3.1 General aims

The general aims are as follows:

1. to further the scientific research of the potential for high temperature reactor technology to contribute to satisfying increasing energy demand; and,
2. to create a conceptual fuel design for the incineration of plutonium from the spent fuel of pressurised light water reactors for the PBMR DPP-400 that is potentially licensable.

1.3.2 Specific aims

The specific aims are to test the following hypotheses:

1. *There is a conceptual fuel design that contains a mixture of plutonium and thorium where the maximum UTC is negative for the full range of temperatures.*
2. *There is a conceptual fuel design that contains a specific mass of only plutonium fuel where the maximum UTC is negative for the full range of temperatures.*
3. *Sufficiently lowering the average thermal flux of plutonium based fuel designs by increasing volume fraction of the coated fuel particles will induce negative UTCs over the full range of temperatures.*
4. *There is a conceptual fuel design to economically burn reactor grade plutonium which meets all of the PBMR safety limits and is therefore conceptually licensable.*

1.4 Literature review

1.4.1 PBMR DPP-400 temperature reactivity coefficients

Using VSOP-99/05 Reitsma demonstrated that the 400 MW thermal Pebble Bed Modular Reactor Demonstration Power Plant (PBMR DPP-400), using the reference case of a heavy metal loading of 9 g uranium per fuel-sphere and an enrichment of 9.6 wt%, achieved the overall temperature reactivity coefficient of -3.66×10^{-5} ($\Delta k_{\text{eff}}/^\circ\text{C}$) (Reitsma, 2004: Table 3). This was at equilibrium temperatures.

Serfontein investigated incineration of Pu(WGR) fuel, Pu(PWR) fuel as well as Pu(PWR) and MA fuel in the PBMR DPP-400 and found that none of the designs met the PBMR (Pty.) Ltd. safety limits (Serfontein, 2011: Section 4.5).

Plutonium incineration in the PBMR DPP-400 in the absence of thorium produced a positive uniform temperature reactivity coefficient for reactor grade plutonium, weapons grade plutonium and reactor grade plutonium with minor actinides at low temperatures and high fuel burn-up (Serfontein, 2011: Section 3.4.2.2.4). Serfontein proposed redesign of the fuel, the power conversion system and the reactor geometry discussed further below:

1. The introduction of ^{232}Th (or inert materials) to the 3 g reactor-grade plutonium fuel spheres and to increase the diameter of the Pu fuel kernels from 240 μm to 500 μm .
2. The indirect Rankine steam cycle rather than the direct Brayton cycle should be utilised. This will lower the temperatures. However, it introduces the possibility of a water ingress event which requires a strong negative UTC.
3. Introduction of neutron poisons into the reflectors to counter the reduction in burn-up and thinning of the central reflector.

1.4.2 Explanations for positive maximum UTCs in Pu incinerating reactors

Serfontein (Serfontein, 2011: Section 3.3.2) reported that lowering the temperatures towards 50°C decreases the reactivity of a Pu fuelled PBMR DPP-400 reactor. A decrease in reactivity with decreasing temperature means that the overall temperature reactivity coefficient is positive at these low temperatures (Serfontein, 2011: Section 3.4.1.4.2). Serfontein emphasises that UTC is only a problem at low temperatures and high fuel burn-up and therefore would only be a problem when a reactor is shut down and restarted cold. It is only during these states that the reactor will be unstable and not acceptably controllable. However, this is not acceptable for a reactor that features inherent passive safety and may also prohibit licensing.

As the FTC in relation to the MTC is very small, investigation of the UTC may be approximated by only investigating the MTC (Serfontein, 2011: Section 3.4.2.3.1). Serfontein

references the explanation by Boer & Ougouag (cited by Serfontein, 2001: Section 3.4.1.4) that the moderator temperature coefficient at different burn-ups depends on the moderation ratio. As the fissile plutonium decreases with burn-up, so the moderation ratio increases. More clearly stated, the atomic density of the fissile material decreases so the ratio of moderator atoms to fissile atoms increases. Neutrons are better moderated by the additional moderator. Thus there are more thermalised neutrons at higher burn-ups than at lower burn-ups. This explains why the moderator temperature coefficient is different at different burn-ups and may be positive.

The capture to fission ratio of ^{239}Pu increases with increased temperature. The change of the capture to fission ratio with temperature is magnified at higher burn-ups due to the additional neutrons. The capture to fission ratio of ^{241}Pu first decreases with increased temperature and then increases with increased temperature from 0.1 eV which as per Maxwellian distribution equates to a temperature. This characteristic of ^{241}Pu is one of the reasons that there is a positive moderator temperature coefficient at low temperatures with fuel that has been burned to the extent that it has a high ^{241}Pu fraction (Serfontein, 2011: Abstract).

Serfontein's results indicated that the maximum UTC of the 3 g Pu(WGR) loading per fuel sphere was higher than the maximum UTC of the 3 g Pu(PWR) loading per fuel sphere (Serfontein, 2011: Section 3.4.2.2.4).

1.4.3 Applicability of the temperature coefficient definitions

Boer and Ougouag investigated the uniform temperature reactivity coefficient over all positions within the core and found that the lower 75 % of the core had slightly positive regions. However, the stability and power output of a reactor is determined by the sum of all its regions, rather than by a few specific region. Therefore, provided that the temperature reactivity coefficient for the whole core is negative, a positive temperature reactivity coefficient in some parts of the reactor does not affect its stability and therefore licensability (Serfontein, 2011: Section 3.4.1.4.1).

Due to the high molecular weight of graphite it takes many more scattering collisions than with hydrogen or deuterium to bring a neutron into thermal equilibrium. Therefore the temperature of the reflectors, rather than the temperature of any specific fuel sphere determines the average energy of the neutrons. Due to this decoupling of the temperature of the fuel sphere and its reactivity and a coupling to the temperature of the reflector, the feedback mechanism is slower. A slow positive reactivity coefficient allows for operator intervention.

1.4.4 Motivations for incineration of Pu with Th

Plutonium is toxic, radioactive and can be used to construct nuclear weapons. Plutonium inventories should either be controlled with high security or they should be incinerated. Plutonium that is incinerated can also be used to produce heat, electricity or both and used to generate an income rather than requiring high security and safe storage generating an expense.

Where plutonium is burnt as uranium/plutonium mixed oxide (MOX) further plutonium or "second generation" plutonium is produced. To ensure that more plutonium is consumed than produced, plutonium must be incinerated with a large plutonium to uranium ratio. Plutonium can also be incinerated as pure plutonium, plutonium with minor actinides or

plutonium with thorium (IAEA, 2003:1). ^{233}U is produced when plutonium is incinerated with thorium. However, this ^{233}U contains the highly radioactive ^{232}U . A highly radioactive material is more difficult for weapon manufacturers to steal and to utilise for weapons manufacturing. Where the fresh fuel contains small quantities of ^{238}U the spent fuel ^{233}U will be denatured by mixing it with ^{238}U so that larger quantities of the mixture will be required for a weapon.

Material usable for weapons can be reduced by incineration and denaturing. Incineration can be defined as a process that causes weapons fuel isotopes to be fissioned or to undergo other nuclear reactions, such as neutron capture with possible subsequent radioactive decay reactions, resulting in this isotope transmutating to one or more isotopes that are not suitable for weapons fuel. Denaturing of a weapons fuel isotope involves mixing of that isotope with other isotopes of the same element, in such a way that the resulting mixture is not suitable as weapons fuel. This mixing can be done by simple mixing, as is currently being done in the Megatons to Megawatts project where weapons grade highly enriched ^{235}U is down-blended to LEU by mixing it with ^{238}U (World Nuclear News, 2011). It can also be achieved by transmutation reactions, such as when weapons grade plutonium is denatured by means of neutron irradiation in a reactor. Some of the ^{239}Pu nuclides then capture a neutron in order to form ^{240}Pu , which denatures the mixture by causing pre-detonation of nuclear weapons fuelled with this mixture (Serfontein, 2011: Section 1.4.2.2). Also, weapons grade material can be rendered unsuitable for weapons fuel by the mixing of excessively radioactive isotopes. The excess radioactivity makes the manufacture of the weapon extremely hazardous. Similarly, weapons grade material can be mixed with isotopes that generate excessive heat. This heat causes the weapon to lose mechanical integrity.

Reprocessing of spent fuel from conventional nuclear reactors and incinerating the fissile plutonium to as low as possible enrichments decrease the probability of proliferation. Chemical reprocessing of nuclear waste would also increase the lifespan of the Yucca Mountain depository a 100 times and would reduce the duration of safe containment of nuclear waste from tens of thousands to only about a thousand years (Serfontein, 2011:Section 1.4.3.1).

1.4.5 Temperature reactivity coefficients of Pu incinerators with Th

The International Atomic Energy Agency (IAEA) facilitated a co-ordinated research project to investigate incineration of plutonium by using it as a driver for thorium fuel cycles using both existing reactors and proposed designs. The Chinese contributors to the IAEA co-ordinated research project found the largest magnitude of the negative fuel temperature coefficients (IAEA, 2003:130) for a 13 g weapons grade plutonium per fuel sphere loading and the largest magnitude of the negative moderator temperature coefficients (IAEA, 2003:130) for a 13 g reactor grade plutonium per fuel sphere loading, at reactor equilibrium.

Mulder and Teuchert demonstrated that it is possible to incinerate plutonium in a PBMR DPP-400 and achieve a negative temperature co-efficient (Mulder & Teuchert, 2008). The plutonium considered only contained 70 % ^{239}Pu and 30 % ^{240}Pu and therefore did not represent the compositions of Pu(PWR) or Pu(WGR) accurately. Furthermore it was carried out with VSOP-A, which does not calculate the resonance integrals for ^{240}Pu or ^{242}Pu , and does not include Americium and Curium in its transmutation chain (Serfontein, 2011, Section 6.5). The PBMR DPP-400 was loaded with two types of fuel spheres: the first fuel sphere contained a heavy metal loading of 3 g of weapons grade plutonium ($^{239}\text{Pu} + ^{240}\text{Pu}$ (70% + 30%)) and the second contained 20 g of 93% enriched uranium and thorium ($\text{U}(93\%) + ^{232}\text{Th}$

(8% + 92%)). The fuel spheres were loaded in such a way that there was a loading fraction of 50 % Pu fuel spheres and 50 % Th/U fuel spheres. Using computer code VSOP-A, Mulder and Teuchert demonstrated that the overall temperature coefficient of reactivity for the equilibrium Pu-Th/U case was -3.92×10^{-5} ($\Delta K_{\text{eff}}/^{\circ}\text{C}$) although no comment was made with respect to the overall temperature coefficient of reactivity over the full range of temperatures. (Mulder & Teuchert, 2008:2897)

The German contributors to the IAEA co-ordinated research project used a similar fuelling strategy as Mulder and Teuchert and demonstrated an overall negative temperature coefficient for both Pu(WGR) and Pu(PWR) at equilibrium temperatures. For the full range of temperatures (IAEA, 2003:43) the German contributors demonstrated that the total temperature coefficient (i.e. the sum of the fuel and moderator coefficients) was negative. Since the HTR-MODUL does not have a central reflector it contains a smaller percentage of thermal neutrons and thus the up-shifting of thermal neutrons into the resonances of Pu, which is responsible for the positive MTCs at low temperatures, can be expected to be much less of a problem. Table 1 summarises the simulation codes, reactor size, fuel strategy, fuel composition and range of temperatures analysed in the reviewed literature.

Table 1: Summary of influence of thorium on temperature reactivity coefficient research.

Researcher/s	Simulation Code	Reactor Size	Fuel Strategy (No. of Fuel Sphere Types)	Fuel Composition	Full Range of Temperature Range
Reitsma	VSOP-99/05	400 MWth	1	LEU	No
Mulder & Teuchert	VSOP-A	400 MWth	2	Pu(WGR) + U+Th	No
Serfontein	VSOP-99/05	400 MWth	1	Pu(WGR) and Pu(PWR)	Yes
IAEA Tecdoc Chinese Contributors	VSOP-94	200 MWth	1	Pu(WGR)+Th and Pu(PWR)+Th	No
IAEA Tecdoc German Contributors	VSOP-97	200 MWth	2	Pu(WGR)+Th+U and Pu(PWR)+Th+U	Yes

No literature review papers were found and the maximum number of citations of relevant papers were limited.

1.5 Mini-dissertation structure

This mini-dissertation will consist of the following chapters:

1. Introduction
2. Method to simulate PBMR DPP-400 fuel cycles
3. Discussion of Pu incinerating concepts
4. Results of fuel cycle simulations

5. Discussion and conclusions
6. Recommended further research studies

2 Method to simulate PBMR DPP-400 fuel cycles

2.1 Fuel composition

Key parameters for the fuel spheres, coated particles and Tristructural-isotropic (TRISO) particles are summarised in Table 2, Table 3 and Table 4 below. The fuel kernel with a diameter of 0.05 cm is contained within the TRISO coated particle. The design of the fuel sphere is such that there is a coated particle free inner spherical core, a shell containing the coated particles embedded in the graphite matrix, and then a fuel free outer shell. This shell fuel sphere configured fuel sphere contains a specific mass of heavy metal and of that a proportion will be Pu(PWR).

Table 2: Fuel sphere key parameters.

<u>Fuel sphere parameter</u>	Unit	Value
Outer Radius of Zones: Graphite Centre/ Fuel Matrix/ Graphite Shell	cm	1.8 / 2.5 / 3.0
Volume of Fuel Sphere	cm ³	113.097
Volume of Fuel Matrix	cm ³	41.021

The volume fraction is the ratio of the volume of coated fuel particles to the total volume of the mixture of coated fuel particles and graphite matrix in the fuel zone of a fuel sphere. Where the volume fraction is too high the coated particles can press against each other when the graphite matrix material is compressed during manufacturing, resulting in unacceptable coated particle breakage. The German high temperature programme did not systematically set out to identify volume fraction limits. The highest certain volume fraction is therefore only the highest volume fraction available from the experimental programs. As this is a theoretical study the volume fraction will not be limited (Serfontein, 2011: Section 4.4.1.1).

Table 3: Coated particles key parameters.

<u>Coated particle parameter</u>	Unit	Value
Diameter of Fuel Kernels	cm	0.05
Diameter of Coated Particle	cm	0.092
Coated Particle Volume	cm ³	4.0772E-04

Table 4: Tristructural isotropic (TRISO) coatings key parameters.

Tristructural isotropic (TRISO) coating parameter	Thickness (cm)	Density (g/cm³)
(1) Inner porous carbon	0.0095	1.05
(2) Inner pyrolytic carbon	0.0040	1.90
(3) Silicon carbide	0.0035	3.18
(4) Outer pyrolytic carbon	0.0040	1.90

Table 5 (Hosking & Newton, 2003, quoted by (Serfontein, 2011: Table 3) details the isotopic composition of reactor grade plutonium with a burn-up of 41.2 GWd/t and 3 years of cooling

Table 5: Isotopic composition of reactor grade plutonium Pu(PWR) (41.2 GWd/tHM and 3 years cooling).

Isotope	Mass Fraction (%)	Atomic Fraction (%)
²³⁸ Pu	2.59	2.78
²³⁹ Pu	53.85	53.91
²⁴⁰ Pu	23.66	23.59
²⁴¹ Pu	13.13	13.03
²⁴² Pu	6.77	6.69

²³⁸Pu is excluded from the heavy metal accounting Pu fuels in VSOP-99/05. VSOP-99/05 treats the ²³⁸Pu as a component of the matrix graphite, i.e. it distributes it homogeneously throughout the fuel sphere, instead of placing it in the fuel kernels (Serfontein, 2011: Table 25). As a result the atomic density of ²³⁸Pu must be manually changed when the volume fraction or kernel size is changed. In contrast the atomic density of the other Pu isotopes is automatically recalculated by VSOP-99/05 when the volume fraction of the coated particles or size of the fuel kernels is modified in the input.

2.2 Reactor geometry and parameters

The PBMR DPP-400 has an effective height of 11 m and an annular core with an inner diameter of 2 m and an outer diameter of 3.7 m. The standard PBMR DPP-400 design dimensions and parameters are used to demonstrate the influence of thorium on the temperature reactivity coefficient in a plutonium incineration reactor.

The major reactor geometry and parameters are detailed in Table 6 below.

Table 6: PBMR DPP-400 reactor geometry and parameters.

Reactor parameter	Unit	Value
Fuel Core Volume	m ³	83.73
Fuel Sphere Packing Fraction	-	0.61
Number of Fuel Spheres in Core	-	451,606
Core Height	m	11.625
Core Annular Inner Radius	m	1.000
Core Annular Outer Radius	m	1.850

2.3 Calculation method for temperature reactivity coefficients

VSOP-99/05 was used to determine the resonance integrals, the reactor equilibrium conditions and the temperature reactivity coefficients. The temperature reactivity coefficients were calculated for the fuel, moderator and for the combination of fuel and moderator (uniform). The component of interest, for example the fuel was changed from equilibrium temperatures to 50°C and then increased in steps of 100°C to 1850°C, while the rest of the components remained at equilibrium temperatures. The multiplication factor (k_{eff}) was determined at each temperature. The temperature reactivity coefficient was then calculated for 100°C, 200°C, etc., up to 1800°C.

Serfontein highlights that, while it is unrealistic that the material of interest is at equal temperature as there is a non-uniform temperature profile in any reactor, this approximation does not significantly affect the findings. Therefore the assumption that the materials of interest are all at a constant temperature is acceptable (Serfontein, 2011).

The Chinese contributors to the IAEA co-ordinated research project demonstrated that they achieved a negative temperature coefficient by increasing the heavy metal loading. Increasing the heavy metal loading required that the enrichment be increased in order to maintain criticality ($k_{\text{eff}} = 1.0$) (IAEA, 2003, Table 3.1.3). In this study different mass loadings of thorium and plutonium or plutonium only were investigated, however, the enrichment of the reactor grade plutonium was fixed. The rate at which the fuel spheres moved through the reactor, determined by the interval between fuel shuffling, was varied to maintain criticality.

2.3.1 Verification method

The input to VSOP-99/05 was approached in two different ways to demonstrate similar results:

- For the first input method the fuel sphere type was simulated to contain plutonium fuel and no thorium. This is the identical fuel cycle that Serfontein used in his doctorate. The accuracy of this input model was assured by showing that the results were identical to those of Serfontein.
- For the second input method the fuel type was simulated to contain a mixture of plutonium and thorium. This second approach was verified against the first by reducing the thorium to negligible levels, which produced virtually identical results to those of the first method.

Once it was established that the input models for the second fuel type produced accurate results, the ratio of thorium to plutonium was increased to investigate its influence on the uniform reactor temperature coefficient.

3 Discussion of Pu incinerating concepts

3.1 Comparison of fertile versus fissile nuclides with temperature increase

The interaction of neutrons with nuclei as temperature increases is physically different with respect to fissile isotopes (^{233}U , ^{235}U , ^{239}Pu and ^{241}Pu) and fertile isotopes (^{232}Th , ^{240}Pu , ^{242}Pu). The capture cross-sections of a fertile isotope are dominated by epithermal resonance peaks, while the fission cross-sections of a fissile isotope are dominated by non-resonance absorption at thermal energies. An increase in neutron energy does not significantly change the cross-section of the fissile isotopes. However, as the temperature of fertile isotopes increases, the broadened resonance peaks absorb a greater number of neutrons. Therefore a higher volume fraction of the fertile ^{232}Th will result in a more negative temperature reactivity coefficient. Also to achieve a high conversion ratio with thorium it is desirable to have a high volume fraction.

3.2 Transfer of heat from fuel, moderator and reflector regions of the reactor

The average energy of well thermalised neutrons is by definition equal to that of the moderator, i.e. their temperatures are identical. Therefore increasing moderator temperature will increase the energies in the Maxwellian energy spectrum of the thermalised neutron flux, which will change the microscopic cross-sections for the different reactions of these neutrons with the different nuclides in the reactor. Increasing moderator temperature also increases the average speed of the neutrons, which increases the thermal neutron flux, for a fixed number density of thermalised neutrons, which may again significantly change the reaction rates with the said nuclides.

During heat production the temperature of the fuel kernels will always be slightly higher than that of the moderator, since the heat is produced directly in the fuel and the temperatures then decreases as the heat diffuses out to the surrounding moderator, i.e. the graphite matrix. Because the coated particles are tightly embedded in the graphite matrix, the heat produced in the fuel kernels will rapidly flow into the surrounding moderator across a minimal thermal resistance and therefore the moderator temperature will quickly rise to only slightly less than the fuel kernel temperatures. This assumption that the fuel and moderator temperatures will be virtually identical provides the rational for the usefulness of the definition of the UTC, in which identical temperatures are allocated to both the fuel and moderator.

It is of interest that the reflector temperature reactivity coefficient at the equilibrium temperature for the PBMR DPP-400 U/Pu-cycle reference case is positive ($+3.2 \times 10^{-5} \Delta K_{\text{eff}}/^\circ\text{C}$) (Reitsma, 2004: Table 3). This is, however, acceptable since the transfer of heat from the fuel spheres to the reflectors is slow. An increase in power will thus cause a prompt increase in the fuel and moderator temperatures in the fuel spheres, which will cause a prompt decrease in reactivity, which will promptly counter the initial power increase. Transfer of excess heat from a power excursion to the reflectors will increase the reactivity of the reactor and may thus necessitate deeper insertion of the control rods. However, this will happen so slowly that it does not pose a safety risk. This assumption of a long time lag between increases in temperatures in the fuel spheres and in the reflectors is the basis for keeping the reflector temperatures constant, while varying the temperatures for

the fuel and moderator, as was done for the calculation of all FTCs, MTCs and UTCs in this study.

Section 1.4.3 of the literature study discusses that certain regions in a Pu fuel core may have positive UTCs, while others may have negative UTCs, depending on the burn-up, fuel composition and temperature of the different regions. The stability and power output of a reactor is, however, determined by the combined effect of all its regions and not by only one or the combination of a few regions. Therefore a positive UTC in one part of the reactor core will not cause a reactivity and power excursion, provided that the overall reactor UTC is negative. Therefore such a positive UTC in fractions of the core does not affect the core's stability and therefore licensability of the reactor.

3.3 Comparison of the MTC and the FTC

The MTC determines the temperature reactivity coefficient as a function of the graphite moderator temperature in the fuel, assuming the fertile fuel (i.e. the resonance absorbers) remains at the equilibrium temperatures. A determinate of the properties of the fuel is the composition. The FTC defines the temperature reactivity coefficient for the fertile fuels, i.e. the resonance absorbers. Since increasing Doppler broadening due to increasing fuel temperature causes only small decreases in the resonance escape probability for most Pu-based fuels, the contribution of the FTC to the UTC is much smaller than that of the MTC.

Pu fuel cycles for the PBMR DPP-400 with positive maximum UTCs can presumably be redesigned to have negative maximum UTCs by increasing the magnitude of the negative FTC or by decreasing the positive MTC .

The magnitude of the negative FTC may possibly be increased by increasing the loading of Th in the fuel spheres, thereby increasing the effect of Doppler broadening and thus decreasing the resonance escape probability.

It is possible that positive MTCs can be reduced by decreasing the average thermal flux since the positive MTC is caused by the upshift of the thermalised Maxwellian neutron flux spectrum into the thermal fission and capture resonances of ^{239}Pu and ^{241}Pu at roughly 0.3 eV (see Figure 12) due to increasing moderator temperature. It follows that decreasing the thermal neutron flux should reduce the magnitude of the MTC. Increasing the Pu loading will reduce the thermal flux and consequently this will be investigated as a mechanism for reducing the MTC.

3.4 Effect of increasing kernel size

The principal difference between the 3 g Pu(PWR) loading per fuel sphere Serfontein characterised and the Pu(PWR) loading per fuel spheres of the current study is that the fuel kernel diameter has increased from 0.24 mm to 0.50 mm.

Serfontein investigated increasing the kernel size while keeping the total mass of plutonium per fuel sphere the same. Following a proposal from literature he diluted the fuel kernels by increasing the kernel size and decreasing its density. Although this was an unrealistic arrangement, it facilitated the study. Serfontein demonstrated that increased kernel size did reduce the maximum UTC (Serfontein, 2011: Section 3.4.2.2.3).

Serfontein explained that dilution of the kernel with inert material and thereby increasing the kernel diameter would result in unacceptably high volume fractions. For this reason Serfontein only investigated the 1 g Pu(PWR) per fuel sphere.

3.5 Alternative proposal to compensate for positive maximum UTC

It is possible that a positive UTC at low temperatures will result in fracturing of the particle coating due to a rapid uncontrolled reactivity and thus power increases at low temperatures. It may be possible to control these power surges at lower temperatures by means of the control rods.

Serfontein discussed a proposal, which he rejected, to overcome the positive UTC at low temperatures. The proposal is to design electrical heaters and specify an operating procedure that the reactor must first be brought to temperature before starting up the reactor. The proposal is rejected because the marketed feature of a fuel sphere bed reactor being passively safe would be compromised by a safety critical operating procedure. It would be possible to install temperature elements and prohibit the reactor operating at low temperatures by installing the control rods. This, however, violated the principle that the PBMR DPP-400 as designed is safe without insertion of the control rods due to the “laws of nature” (Serfontein, 2011: Section 4.2.3.2).

If the heavy metal component is unacceptably high, chemical processing of the spent fuel is required. The recovered product would thereby be available for incineration utilising alternate fuel strategies or other reactor designs.

3.6 Analysis of maximum fuel temperature

The maximum fuel temperature for the Pu fuel simulations often does not occur in the main part of the reactor but in the spent fuel outlet or chute. The maximum fuel temperature is not against the central reflector at the peak of the maximum power as would be expected. The design of the PBMR DPP-400 is such that the fuel spheres pass through the reactor six times. The fuel spheres that pass through the channel immediately adjacent to the central reflector experience such high burn-ups that their fissile fuel contents are depleted and thus their values of k_{∞} are sharply reduced, which causes the fission rate in these fuel spheres to decrease towards the bottom of the core (Serfontein, 2011: Section 3.4.3.3.2).

In contrast the fuel spheres in the central channels have low burn-ups because they are exposed to neutrons that are less moderated. When the fuel spheres pass through the discharge fuel chutes at the bottom of the reactor, the narrowing of the distance between the reflectors causes these fuel spheres to be exposed to more well-thermalised neutrons. As a result the fuel spheres’ power spikes. In addition, the helium coolant at the bottom of the reactor is at its maximum temperature, and thus the cooling rate at its lowest. The combination of these factors leads to a spike in the temperature and thus the maximum temperature is reached in the outlet chutes.

The application of the indirect Rankine cycle, as opposed to the direct Brayton cycle of the PBMR DPP-400, will reduce the maximum fuel temperature. This change of the power conversion system, as proposed by Serfontein, will reduce the inlet and outlet helium temperatures and thereby reduces both the average and maximum equilibrium fuel temperatures. The lowering of the temperatures of all the reactor structures will also reduce the amount of heat stored in these structures during equilibrium operation. This will also

reduce the temperatures during loss of coolant accidents. However, lowering the temperatures of the helium limits potential utilisation of this configuration for high temperature process heat applications. The utilisation of the steam cycle can result in a water ingress event and therefore will require a strongly negative UTC (Serfontein, 2011: Section 4.4.2).

3.7 Diffusion of radioactive silver through coated particles

The diffusion of silver through the coated particle layers is determined by the maximum fuel temperature as per Figure 1 below. The maximum equilibrium fuel temperature of a reactor design is limited by the diffusion of radioactive silver through the coated particles. The design limit of the PBMR DPP-400 with LEU fuel was 1130°C. However, ^{239}Pu generates “an order of magnitude” more silver in the fission products than ^{235}U . It is therefore possible that the maximum fuel temperature needs to be significantly less than that adopted by the PBMR company (Serfontein, 2011: Section 3.3.4.3).

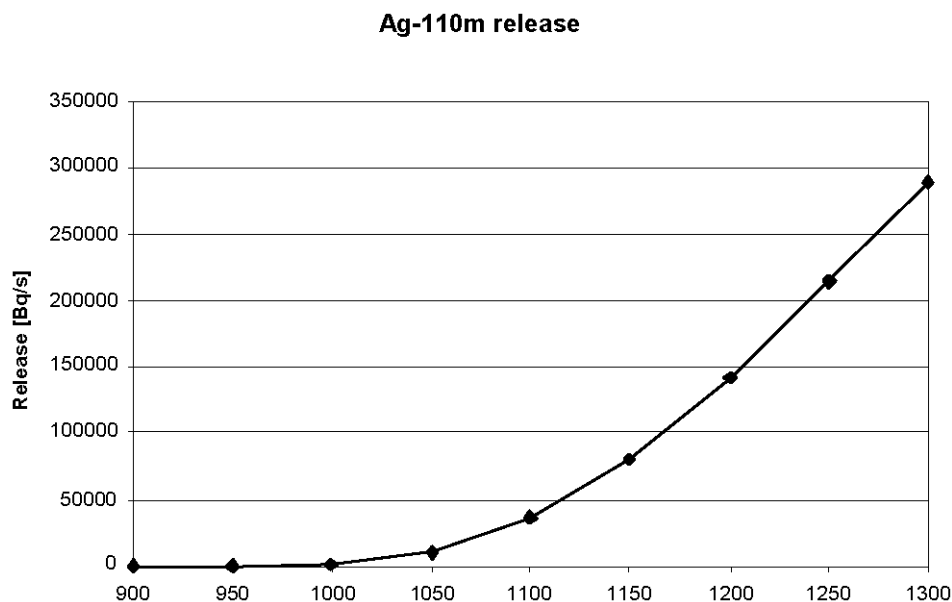


Figure 1: Release rate of radioactive $^{110\text{m}}\text{Ag}$ out of coated fuel particles, as a function of temperature (Serfontein, 2011).

Serfontein states that the PBMR DPP-400 fuelled with LEU fuel spheres was limited as the design had a direct cycle. This could result in maintenance personnel receiving high doses of radiation while maintaining the turbine. Serfontein proposes that utilising an indirect cycle would not have the same problem as the turbine would be driven by uncontaminated steam.

3.8 Significance of higher burn-up for Pu incinerating reactors

Higher burn-up of reactor grade plutonium fuel has two main benefits. Firstly more energy is produced per ton of heavy metal or manufactured fuel sphere and secondly the amount of ^{241}Pu that is sent to final disposal is reduced. The isotopic composition of the fuel spheres for final disposal determines the length of time it takes for the radioactivity to reach background levels, the radiotoxicity during that period and the potential for abusing the spent fuel for proliferation of nuclear weapons.

Serfontein demonstrated that the burn-up of 3 g Pu(PWR) loading per fuel sphere is much more (601.0 GWd/tHM) than the burn-up of 9 g U/Pu (LEU) loading per fuel sphere (90.8 GWd/tHM). The burn-up of 9 g U/Pu (LEU) loading per fuel sphere (90.8 GWd/tHM) is also higher than the burn-up of conventional light water reactors (41.2 GWd/t). It should be noted that the enrichment of Pu(PWR) is much higher than for LEU, which in turn is higher than for conventional light water reactors.

As the burn-up of 3 g Pu(PWR) loading per fuel sphere is more than the 9 g U/Pu (LEU) loading per fuel sphere and the power output of a PBMR reactor is still the same, Pu(PWR) fuel spheres spend longer in the reactor. The total residence time of a Pu(WGR) fuel sphere is six years and two and a half years for an LEU fuel sphere (Boer, et al., 2009:1051). However the axial power profile is dependent on the rate of change of the k_{∞} as the fuel spheres slowly flow down from the top to the bottom of the core. This rate of change is higher for Pu(WGR) than for LEU fuel. To achieve a flat axial power profile it is necessary to pass the fuel spheres through the core at a faster rate (i.e. the number of passes through the core over the fuel spheres' residence time should increase). Dust generation is more of a concern when the number of times that fuel spheres pass through the reactor is increased.

Serfontein highlighted that attaining very high burn-up's are a theoretical possibility and recommended that an experimental programme determine what is actually achievable whilst still maintaining the coated particle integrity.

3.9 Economic considerations of burning Pu with Th

The focus of the International Atomic Energy Agency (IAEA) co-ordinated research project was to "constrain plutonium and reduce long lived waste toxicity." (IAEA, 2003:1). The nuclear industry, however, is in the power generation business and burn-up, the measure of thermal energy produced per mass of heavy metal, is a suitable indicator of the economic viability of a reactor. With high temperature reactors fuel costs include the cost of manufacture of the fuel sphere and in the case of plutonium incinerating reactors fuel costs must include the costs of processing the waste of conventional nuclear reactors. It may be that nuclear operators may be willing to pay for a company to take its spent fuel or only plutonium away. The plutonium incinerating power generating company would receive an income from the nuclear operator for accepting its spent fuel and it would also make a return from the power generated.

3.10 Proliferation consideration of incinerating Pu

With respect to achieving proliferation aims Serfontein explains how ^{241}Pu decays to ^{237}Np . ^{237}Np is a suitable weapons fuel. The mass of ^{241}Pu in the spent fuel spheres decays to ^{241}Am which decays to ^{237}Np . As ^{237}Np has a high fast fission fraction it would be more appropriate to incinerate ^{237}Np in reactors with thicker fuel rods, such as fast reactors, light water reactors or pressurised heavy water reactors, such as the CANDU. Due to, among other factors, their much thicker fuel rods, these reactor types produce a much higher fraction of fast fissions, as opposed to pebble bed reactors (Serfontein, 2011: Section 3.4.4.3). An accelerator-driven design may also be suitable for the burning of a subcritical assembly of ^{241}Pu .

4 Results of fuel cycle simulations

4.1 Influence of Th on temperature reactivity coefficients

Figure 2 and Figure 3, compare the UTC, MTC and FTC of a 0.75 g Pu(PWR) and 7.25 g Th loading per fuel sphere with the 9 g U/Pu(LEU) loading per fuel sphere and 3 g Pu(PWR) loading per fuel sphere cases respectively. To ensure that there is not a positive UTC it was required to simulate the UTC for the full range of temperatures. The figures all show a clear correlation between the UTC and the MTC. The MTC dominates the FTC and thus the trend for the MTC largely determines the trend for the UTC.

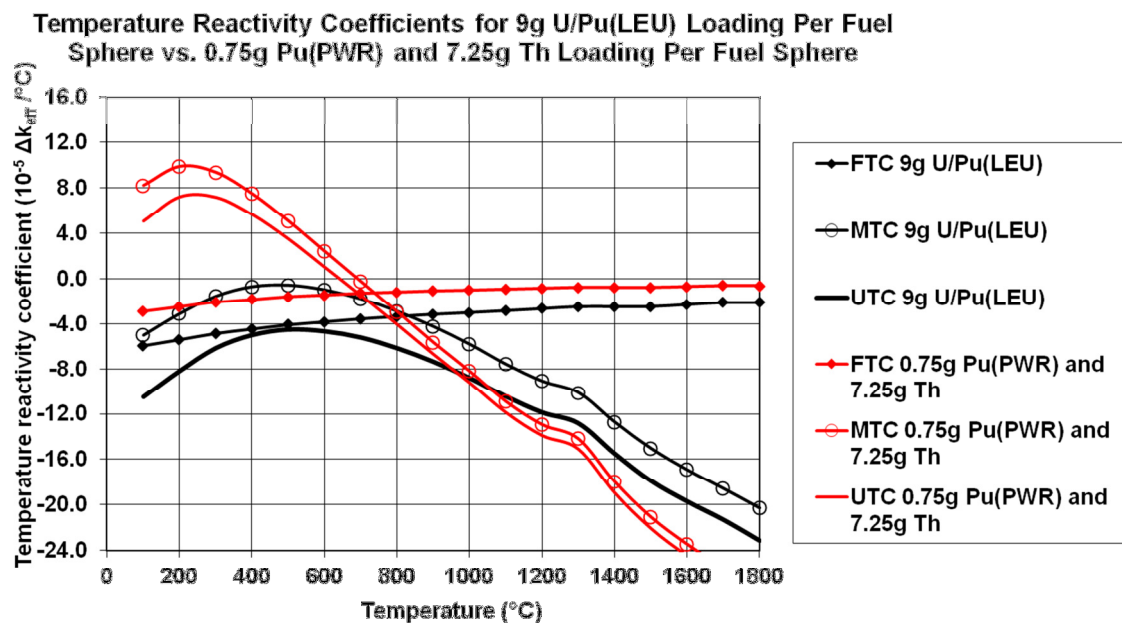


Figure 2: Comparison of UTC, FTC and MTC for 9 g U/Pu(LEU) loading per fuel sphere vs. 0.75 g Pu(PWR) and 7.25 g Th loading per fuel sphere.

The mechanisms of the FTC and the MTC are different. An increase in fuel temperature leads to Doppler broadening of the capture resonances. This increases captures and thus reduces the number of neutrons that are thermalised.

An increase in moderator temperature does not change the number of neutrons that are thermalised. Collision with the more energetic moderator nuclei causes those neutrons that were thermalised to move around at higher speeds. Consequently the thermal flux increases.

This does not necessarily produce an increase in k_{eff} . The increase in neutron speed causes the neutron energy spectrum peak to shift to higher energies. This shift can change the microscopic cross-sections that these neutrons encounter when interacting with different nuclides. This can either lead to more captures and thus a reduction in k_{eff} or to more fissions and thus to an increase in k_{eff} .

The figures show that the change in the resonance escape probability, due to increasing fuel temperature, is less significant than the changes in the neutron economy caused by change in the thermal neutron energy spectrum, due to increasing moderator temperature.

The FTC increases with temperature but remains negative for the full range of temperatures. Figure 3 below indicates that the minimum FTC for the 0.75 g Pu(PWR) and 7.25 g Th loading per fuel sphere is less than for the minimum FTC of the 3 g Pu(PWR) loading per fuel sphere. This implies that the Doppler broadening of the capture resonance changes more with respect to temperature for the plutonium and thorium mixture than for plutonium only. The similarity of the results for of the UTC, MTC and FTC to those from the literature provides confidence in the results of the current study.

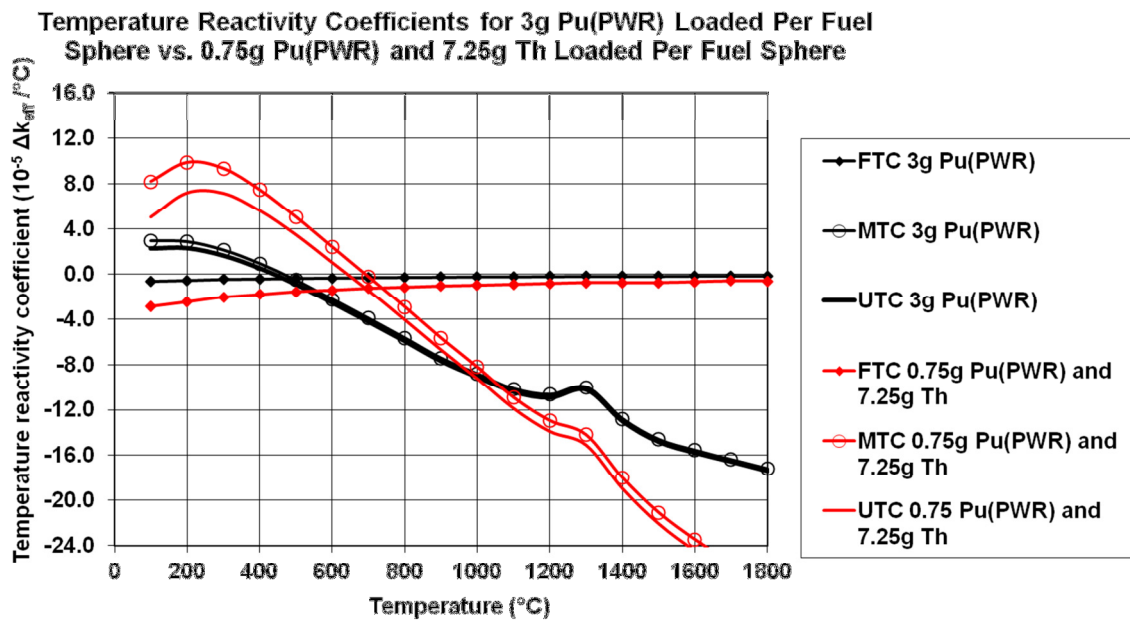


Figure 3: Comparison of UTC, FTC and MTC for 3 g Pu(PWR) loading per fuel sphere vs 0.75 g Pu(PWR) and 7.25 g Th loading per fuel sphere.

Figure 4 below shows the UTC over the full range of temperatures for different mixtures of reactor grade plutonium and thorium in an 8 g heavy metal loading per fuel sphere. It is evident that the maximum UTC occurs at low temperatures and that for all the cases the UTC is negative above 700°C.

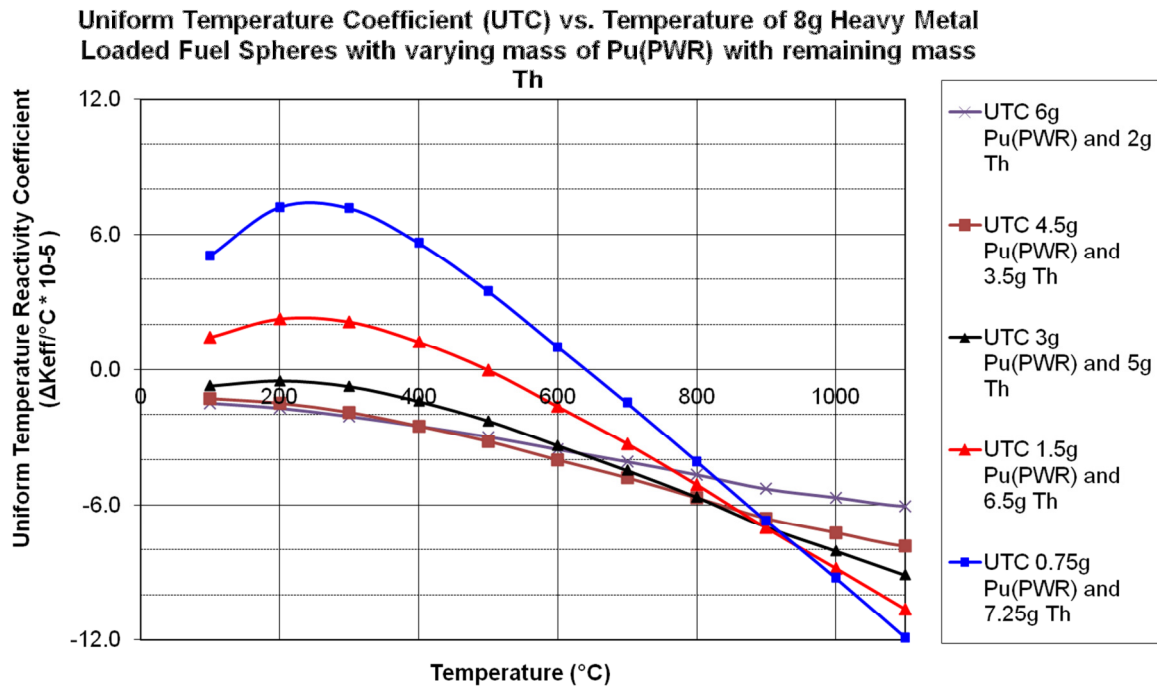


Figure 4: The UTC vs. temperature of 8 g heavy metal loading per fuel sphere with varying mass of Pu(PWR) with remaining mass Th.

It may seem that the higher concentration of thorium results in a more positive maximum UTC. Serfontein showed that decreasing the mass of Pu(PWR) per pebble without thorium resulted in a more positive maximum UTC (Serfontein, 2011, Figure 26). It thus seems likely that it is not the increasing thorium content that causes the maximum UTC to be positive. For a fixed heavy metal loading, increasing thorium content means reduced Pu(PWR) content. Thus a more accurate interpretation of this chart is that lowering the Pu(PWR) loadings leads to a more positive maximum UTC.

4.2 Analysis of the maximum UTC

Figure 5 shows maximum UTC as a function of Pu(PWR) heavy metal loading per fuel sphere for a Pu(PWR) and Th mixture with a fixed total heavy metal content of 8 g per fuel sphere. The mass of Pu(PWR) is indicated in the horizontal axis and remaining mass of the 8 g heavy metal loading is thorium. For example the first data point indicates a fuel sphere containing 0.75 g Pu(PWR) and 7.25 g Th.

The figure shows that the 0.75 g Pu(PWR) and 7.25 g Th loading per fuel sphere has a positive UTC and that the 6 g Pu(PWR) and 2 g Th loading fuel sphere has a negative UTC.

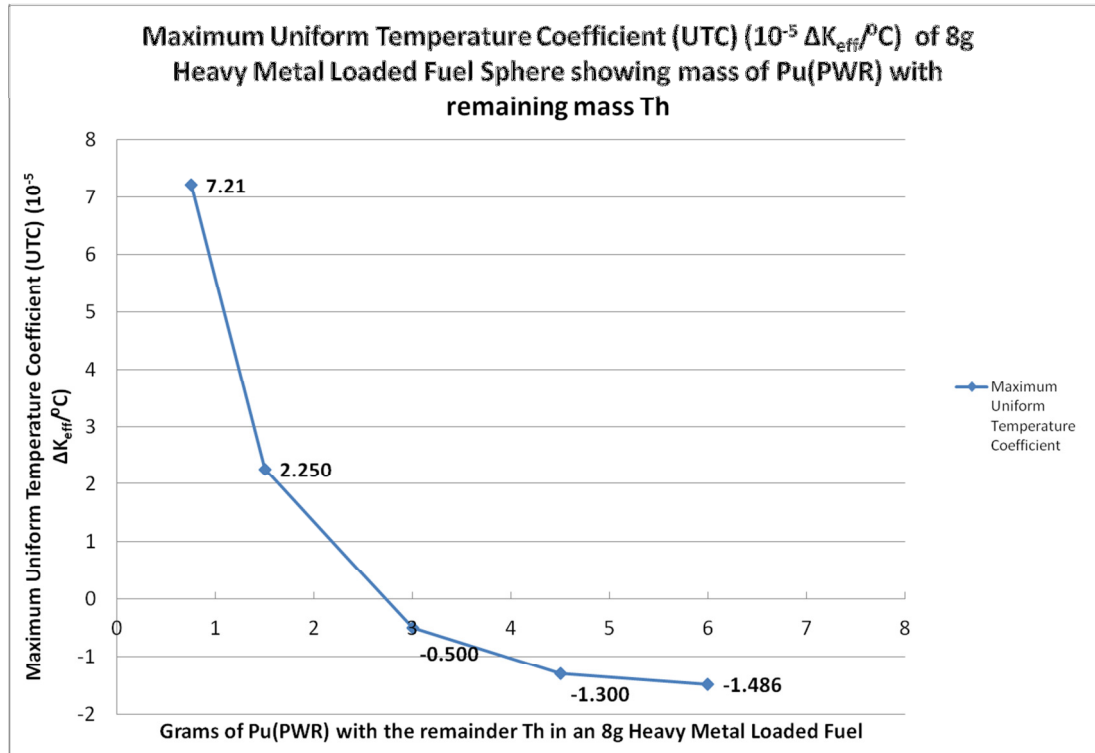


Figure 5: Maximum UTC as a function of Pu(PWR) heavy metal loading per fuel sphere for a Pu(PWR) and Th mixture with a fixed total heavy metal content of 8 g per fuel sphere.

As the mass of plutonium increases, the maximum UTC decreases sharply and eventually switches from positive to negative. As increasing mass of Pu corresponds with decreasing mass of Th, this result may seem to suggest that decreasing the amount of Th in a fuel sphere will also decrease the UTC. This would be counter-intuitive as it would be expected that as the mass of thorium increases, the increased effect of Doppler broadening in the capture resonances of thorium should increase the magnitude of the negative FTC, which should then also lead to a more negative UTC. However, when the mass of Pu(PWR) was reduced from 3 g to 1 g, for the case without thorium, Serfontein found that the maximum MTC increased dramatically. This latter very large positive MTC completely dominated the small negative FTC. It is thus clear that as the mass of Pu decreases it causes a large increase in the positive MTC which makes the negative contribution of the FTC largely irrelevant. Therefore the trend of reducing UTC in Figure 5 is probably not due to the decreasing mass of thorium but rather the mainly due to the increasing mass of plutonium.

However, a more detailed comparison with the results of Serfontein shows that the Th content in the present study indeed helped to lower the UTC. Serfontein (2011: Fig. 22, Fig. 26 and Fig. 27) found that reducing from the Pu(PWR) loading from 3 g to 1 g, increased the maximum MTC from $3 \times 10^{-5} \Delta k_{eff}/^{\circ}C$ to $16 \times 10^{-5} \Delta k_{eff}/^{\circ}C$. This trend would suggest an MTC for 0.75 g Pu(PWR) of significantly larger than $16 \times 10^{-5} \Delta k_{eff}/^{\circ}C$. However, the present results (Fig. 3) showed that the addition of the 7.25 g Th to the 0.75 g Pu(PWR) almost halved the maximum MTC to only $10 \times 10^{-5} \Delta k_{eff}/^{\circ}C$. This suggests that keeping the Pu loading fixed and increasing the Th loading (and thus increasing the total heavy metal loading to for example 16 g per fuel sphere), may reduce the positive MTC and increase the magnitude of the

negative FTC enough to obtain a negative UTC at such a low Pu loading. This possibility is thus referred to the suggestions for future study.

4.3 Influence of Th on other PBMR DPP-400 safety limits

4.3.1 Influence of Th on fast fluence

Figure 6 plots the fast fluence as a function of burn-up.

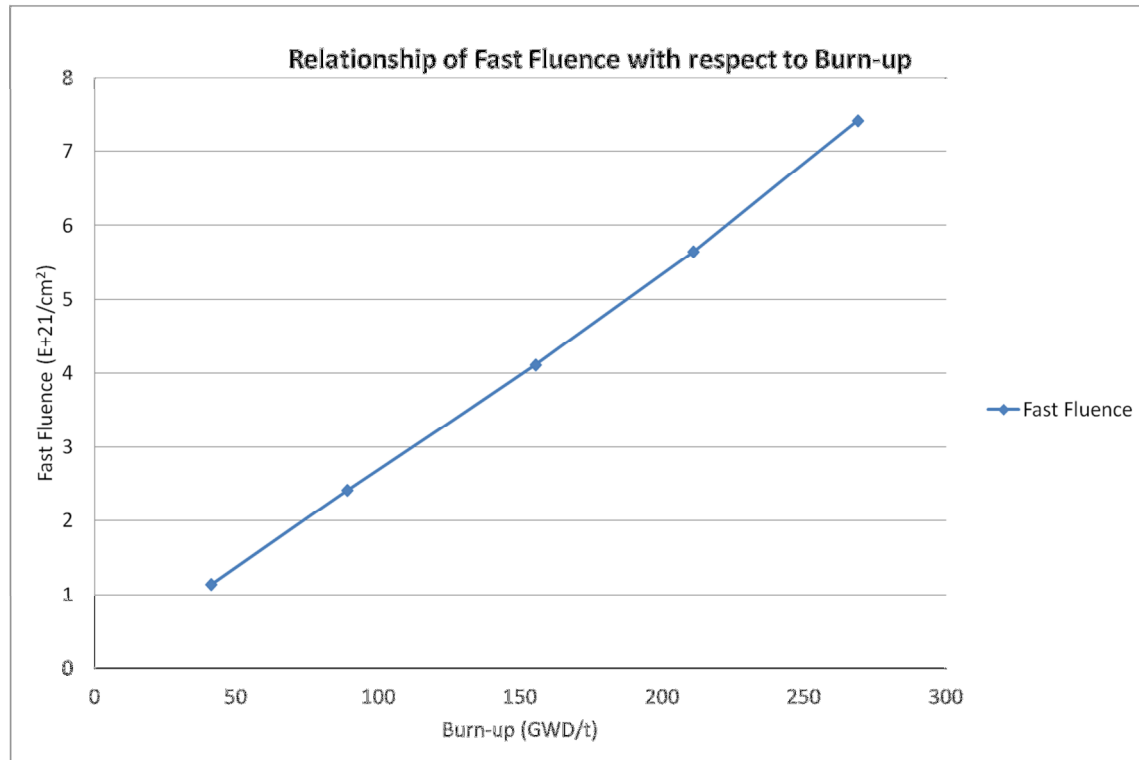


Figure 6: Relationship of fast fluence as a function of burn-up.

The higher the number of fast neutrons that pass through a material, the more likely the mechanical integrity of that material is to be compromised. This is of particular relevance to pebble bed reactors as the primary containment of radioactive fission products is the coating of the kernels. Ruptured coating layers release gaseous radioactive fission products. The PBMR safety limit for fast fluence is $8.0 \times 10^{21} / \text{cm}^2$. The longer the coated particles embedded in the fuel spheres are in the reactor, the higher the fast fluence. Generally high burn-up requires the fuel spheres to be in the reactor for longer. The figure above indicates, as expected a linear relationship between burn-up and fast fluence.

4.3.2 Influence of Th on maximum fuel temperature and maximum power per fuel sphere

The maximum fuel temperature as discussed in Section 3.7 is of high significance with plutonium incinerators as plutonium fuel generates ten times as much radioactive $^{110\text{M}}\text{Ag}$. Radioactive silver is the most difficult of the fission products to retain within the coated particles.

Figure 7 shows the maximum fuel temperature and the maximum power per ball as a function of mass of Pu(PWR) in a 8 g heavy metal fuel sphere where the remaining heavy metal is thorium.

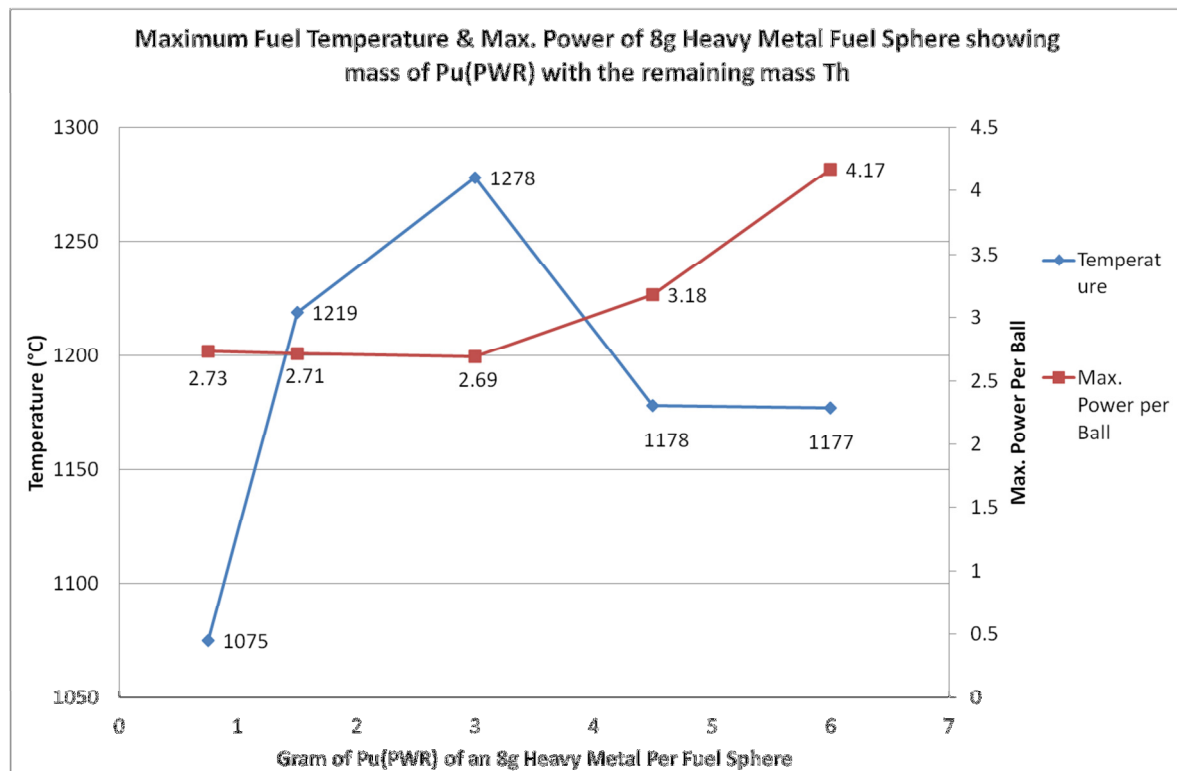


Figure 7: Maximum fuel temperature and maximum power per fuel sphere vs. 8 g heavy metal loading per fuel sphere with varying mass of Pu(PWR) with the remaining mass Th.

The maximum power per fuel sphere increases with increasing mass of plutonium in an 8 g fixed heavy metal fuel sphere.

Although this phenomenon was not quantitatively investigated in this study, it may be explained as follows. The average power per fuel sphere is always the same in a 400 MWth reactor that has the same number of fuel spheres. Thus the composition of the heavy metal loading of the fuel sphere does not affect the average power per fuel sphere. However, the maximum power per fuel sphere is dependent on the axial or radial power profile. An increased Pu loading results in a lower average thermal neutron flux in the fuel. The thermal neutron flux is lower in the centre of the core because of the high Pu loading. However, high fluxes of thermalised neutrons still diffuse from the reflectors into the fuel layers directly adjacent to the reflectors. Therefore the power peaks directly adjacent to the central reflector. The power peak causes a high maximum power per fuel sphere. Thus the trend of the maximum power per fuel sphere in an 8 g heavy metal per fuel sphere is determined by increasing the mass of plutonium, rather than by a decreasing mass of thorium.

Analysis of the position within the reactor showed that the result of 1219°C for 1.5 g Pu(PWR) and 6.5 g Th loading per fuel sphere and the result of 1278°C for 3 g Pu(PWR) and 5 g Th loading per fuel sphere occurred in the outlet fuel chute at the

bottom of the core. This is caused by a small power spike in the outlet chute, caused by increasing moderation due to the fuel stream being “squeezed by the reflector cones, so that [the flow channels] become much thinner” (Serfontein, 2011: Section 3.4.3.3.1). This effect is undesirable and in a real plutonium fuelled reactor this effect will probably be suppressed by putting neutron poisons in the graphite of the outlet chutes. Therefore the increased fuel temperatures reported here are not practically relevant and will not be investigated further.

4.4 Influence of Th on burn-up

Figure 8 below shows the burn-up of an 8 g heavy metal with varying masses of Pu(PWR) with the remaining mass Th.

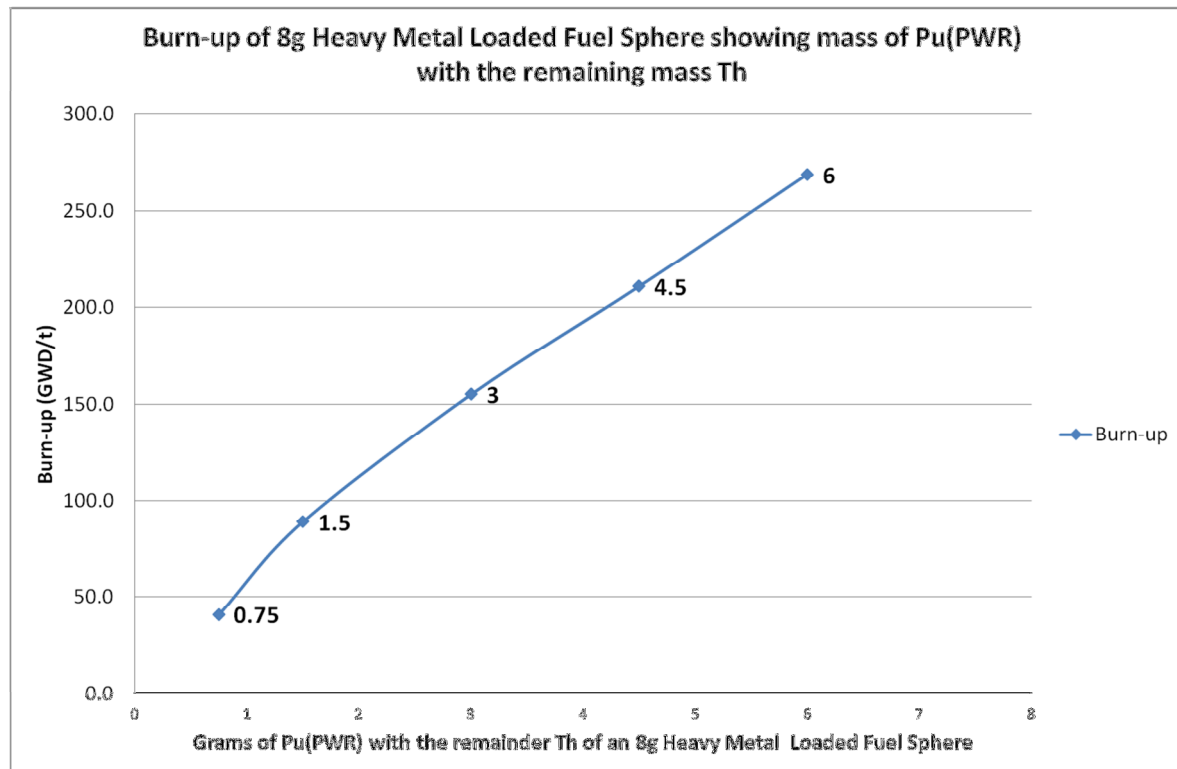


Figure 8: Burn-up of 8 g heavy metal loading per fuel sphere with varying mass of Pu(PWR) with the remaining mass Th.

It is evident that as the mass of plutonium increases and the mass of thorium decreases in an 8 g heavy metal per pebble, the burn-up increases. This can be explained as follows. The macroscopic capture cross section for the epithermal resonances of Th captures neutrons. Decreasing the mass of Th decreases the capture of neutrons which increases the resonance escape probability. This increases the k_{eff} , which means that the fuel spheres can be in the reactor for longer to achieve a k_{eff} equal to 1. The longer the fuel spheres are in the reactor the larger the burn-up.

4.5 Effect of increasing Pu loading per fuel sphere without Th

Figure 9 below shows that a fuel design with Pu(PWR) and no thorium achieves a negative UTC. A fuel design of 6 g Pu(PWR) and no Th per fuel sphere has a maximum UTC of $-0.652 \times 10^{-5} \Delta K_{\text{eff}}/^{\circ}\text{C}$ whereas a fuel design of 6 g Pu(PWR) and 2 g Th per fuel sphere has a maximum UTC of $-1.486 \times 10^{-5} \Delta K_{\text{eff}}/^{\circ}\text{C}$. This indicates that if the heavy metal is not kept

constant, but the mass of plutonium is constant and the mass thorium is increased, the maximum UTC decreases.

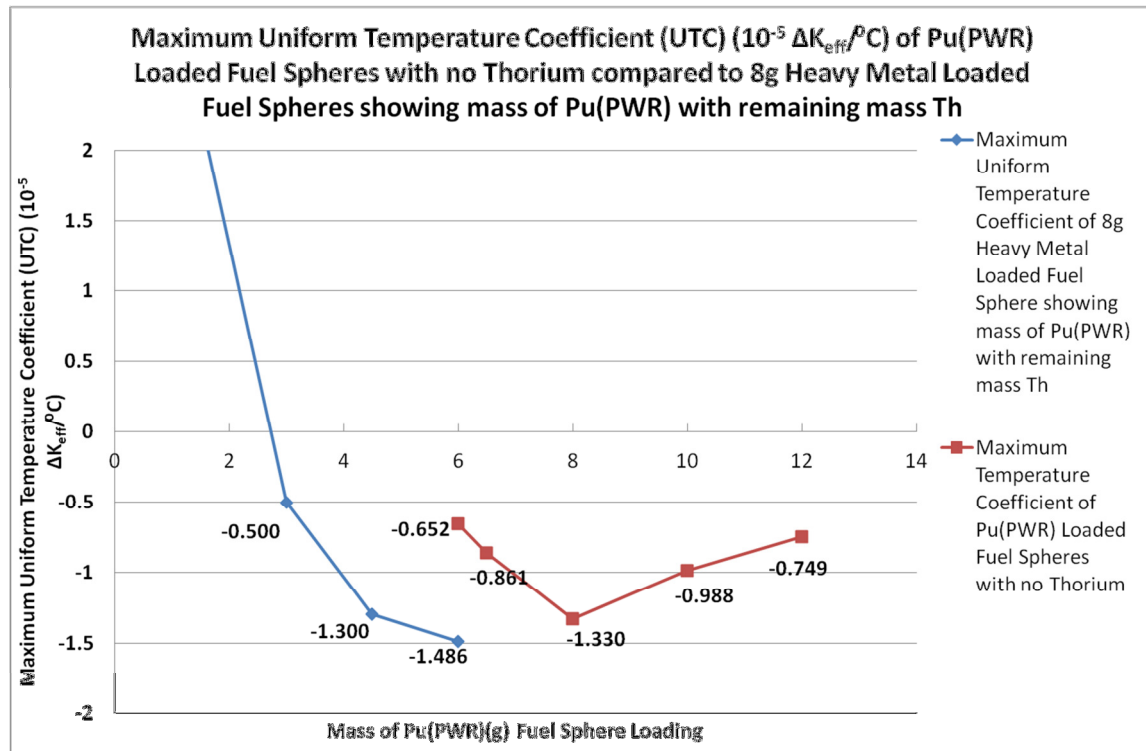


Figure 9: Maximum UTC as a function of Pu(PWR) loadings per fuel spheres for the cases with no thorium compared to the maximum UTC as a function of Pu(PWR) heavy metal loading per fuel sphere for a Pu(PWR) and Th mixture with a fixed total heavy metal content of 8 g per fuel sphere.

The result that no thorium is required to achieve a negative UTC is important. Introducing thorium generates ^{233}U . As there is no ^{238}U present in the mixture, this ^{233}U can be separated chemically. Despite the large radiological hazard from the small fraction of ^{232}U , this mainly ^{233}U mixture constitutes excellent weapons material and therefore presents a proliferation risk (Serfontein, 2011: Section 1.4.5.2)

The other significant result from the above is that the maximum UTC of the fuel spheres containing only plutonium reaches an absolute minimum of $-1.330 \times 10^{-5} \Delta K_{\text{eff}}/^\circ\text{C}$ with 8 g Pu(PWR) loading per fuel sphere. The maximum UTC increases slightly above 8 g Pu(PWR) loading per fuel sphere.

This may possibly be explained as follows. Serfontein showed that as the plutonium loading increases, the magnitude of the positive maximum MTC decreases (Serfontein, 2011: Figure 26). In the present study Figure 13 and Figure 14 below suggest that this reduction in the MTC, and thus in the UTC, is caused by a sharp reduction in the thermal neutron flux in the fuel with increasing Pu loading, due to decreasing moderation ratio. However, Figure 13 shows that as the plutonium loading increases, the thermal flux approaches zero, which would suggest that the MTC also approaches zero. From that point onward, the contribution of the MTC to the UTC will become irrelevant and the trend of the UTC will thus be

dominated by the trend of the FTC. In a well moderated fuel, thermal fissions will dominate and the negative trend of the FTC will be determined mainly by increasing neutron capture in the capture resonances of the resonance absorbers, i.e. ^{240}Pu and ^{242}Pu , due to increasing Doppler broadening with increasing temperature. However, in the virtual absence of thermal neutrons and thus of thermal fissions, fissions in the epithermal fission resonances of ^{239}Pu and ^{241}Pu may become the dominant mechanism for fissions. In such a case increasing Doppler broadening due to increasing temperatures might lead to increasing fissions in these fission resonances and thus to an increase in k_{eff} . However, the details of this phenomenon fall outside the scope of the present study and is thus referred to the suggestions for future study.

Table 7 shows the burn-up, fast fluence, maximum fuel temperature, maximum power per ball, maximum UTC and average thermal flux for fuel spheres containing a mass of only Pu(PWR).

Table 7: Burn-up, fast fluence, maximum fuel temperature, maximum power per fuel sphere, maximum UTC and average thermal flux as a function of the mass of Pu(PWR) loading per fuel sphere with no thorium.

Pu(PWR) Loading	Burn-up	Fast Fluence	Max. Fuel Temperature	Max. Power Per Fuel Sphere	Max. UTC	Avg. Thermal Flux
g	GWD/t	$\times 10^{21}/\text{cm}^2$	$^{\circ}\text{C}$	kW/Fuel Sphere	$\times 10^{-5} \Delta K_{\text{eff}}/^{\circ}\text{C}$	$\times 10^{14}/(\text{cm}^2.\text{s})$
3	448.6	9.33	1201	4.78	3.313	0.25
6	440.7	9.78	1212	4.80	-0.652	0.09
8	278.4	7.81	1232	3.01	-1.330	0.06
12	180.8	7.75	1032	2.22	-0.749	0.03

The table above shows that as the mass of Pu(PWR) per fuel sphere increases the burn-up, fast fluence and maximum power per fuel sphere decrease. There is insufficient data for a clear relationship between mass of Pu(PWR) per fuel sphere and maximum fuel temperature.

A possible explanation for the increase in burn-up of Pu is as follows. As the Pu loading increases the moderation ratio decreases as a result of the volume of graphite between adjacent fuel kernels decreasing. Therefore the probability of a neutron being absorbed in the kernels before being thermalised is increased. This causes a decrease in the resonance escape probability which causes a decrease in k_{eff} . In order to restore k_{eff} equal to 1 the old fuel spheres need to be replaced by fresh ones faster and thus the fuel spheres stay in the reactor for a shorter time and hence the burn-up decreases.

The 12 g Pu(PWR) loading per fuel sphere characteristic parameters are below the $8.0 \times 10^{21}/\text{cm}^2$, the 1130°C , the 4.5 kW per fuel sphere safety limits and has a negative maximum UTC.

The proposed PBMR 9 g U/Pu (LEU) fuel sphere has a lower burn-up of 90.8 GWd/tHM (Reitsma, 2004:Section 4.1). A 12 g Pu(PWR) per fuel sphere reactor has approximately double the burn-up at 180.7 GWd/tHM.

Table 8 shows the composition of the fuel of a 12 g Pu(PWR) fuel sphere when it is loaded and when it is discharged from the reactor. The table below provides the isotopic composition of the spent fuel for the 12 g Pu(PWR) per fuel sphere design which meets all the safety limits. The decrease in the Pu content is from 5.53 kg/GW_{thd} to 4.17 kg/GW_{thd} representing a decrease of only 24.5 %. With respect to the reference case of 3 g Pu(PWR) per fuel sphere of Serfontein the decrease in Pu is from 1.66 kg/GW_{thd} to 0.51 kg/GW_{thd} representing a much larger decrease of 69.2 % (Serfontein, 2011).

Table 8: Material balance for 12g Pu(PWR) loading per fuel sphere.

Isotope	Symbol	Unit	Loading	Unloading	Δ HM
U-234	²³⁴ U	kg/GW _{thd}	0	0.0067	0.0067
U-235	²³⁵ U	kg/GW _{thd}	0	0.0015	0.0015
U-236	²³⁶ U	kg/GW _{thd}	0	0.0008	0.0008
NP-237	²³⁷ Np	kg/GW _{thd}	0	0.001	0.001
PU-238	²³⁸ Pu	kg/GW _{thd}	0.1527	0.1611	0.0084
PU-239	²³⁹ Pu	kg/GW _{thd}	2.9734	1.8868	-1.0866
PU-240	²⁴⁰ Pu	kg/GW _{thd}	1.3064	0.9372	-0.3692
PU-241	²⁴¹ Pu	kg/GW _{thd}	0.725	0.8086	0.0836
PU-242	²⁴² Pu	kg/GW _{thd}	0.3738	0.3803	0.0065
AM-241	²⁴¹ Am	kg/GW _{thd}	0	0.2188	0.2188
AM-242M	^{242m} Am	kg/GW _{thd}	0	0.0075	0.0075
AM-243	²⁴³ Am	kg/GW _{thd}	0	0.0632	0.0632
CM-242	²⁴² Cm	kg/GW _{thd}	0	0.0015	0.0015
CM-243	²⁴³ Cm	kg/GW _{thd}	0	0.0004	0.0004
CM-244	²⁴⁴ Cm	kg/GW _{thd}	0	0.0349	0.0349
Total Pu		kg/GW _{thd}	5.5313	4.174	-1.3573
Total HM		kg/GW _{thd}	5.5313	4.5103	-1.021

The consequences of an increase in the plutonium in the spent fuel are the following:

1. The energy obtained per fuel sphere decreases and therefore the cost recovered per manufactured fuel sphere decreases.
2. Spent fuel with the above mass of plutonium is sub-optimal for direct to disposal because it still contains a large amount of plutonium and highly radioactive Minor Actinides (MA).

3. The isotopic composition of the plutonium may indicate that proliferation objectives are not optimally achieved.

The data on achievable plutonium burn-up with conventional coatings is limited. It would therefore be conservative to first construct plutonium incinerators with lower burn-up. The principal economic feasibility factor is the cost of the fuel sphere manufacture.

4.6 Relationship of thermal flux to maximum UTC

4.6.1 Factor analysis of the multiplication factor

Analysis of the UTC of these factors was not satisfactory as the data in the VSOP output for the burn-up cycle combines the influences of epithermal neutrons and thermal neutrons, while the theoretical analysis of Lamarsh and Baratta separates these.

In Figure 10 the effective multiplication factor, k_{eff} for a 3 g Pu(PWR) loading per fuel sphere is compared with the following factors, as a function of temperature:

1. the fuel utilisation;
2. the number of fission neutrons emitted per thermal neutron absorbed in the fissile isotopes;
3. the non-leakage factor, and
4. the absorption of thermal neutrons in non-fissile nuclides.

The absorption of thermal neutrons in non-fissile nuclides is not related to the resonance escape probability as this refers to the probability that fast neutrons will not be absorbed while slowing to thermal neutrons. Neutrons are mostly captured by fission products in the thermal energy regions.

The values have been normalised to show the relative change of the different variables of interest as a function of moderator temperature as opposed to absolute values. Because the fuel temperature was not changed, the change in the temperature only affected thermal neutrons.

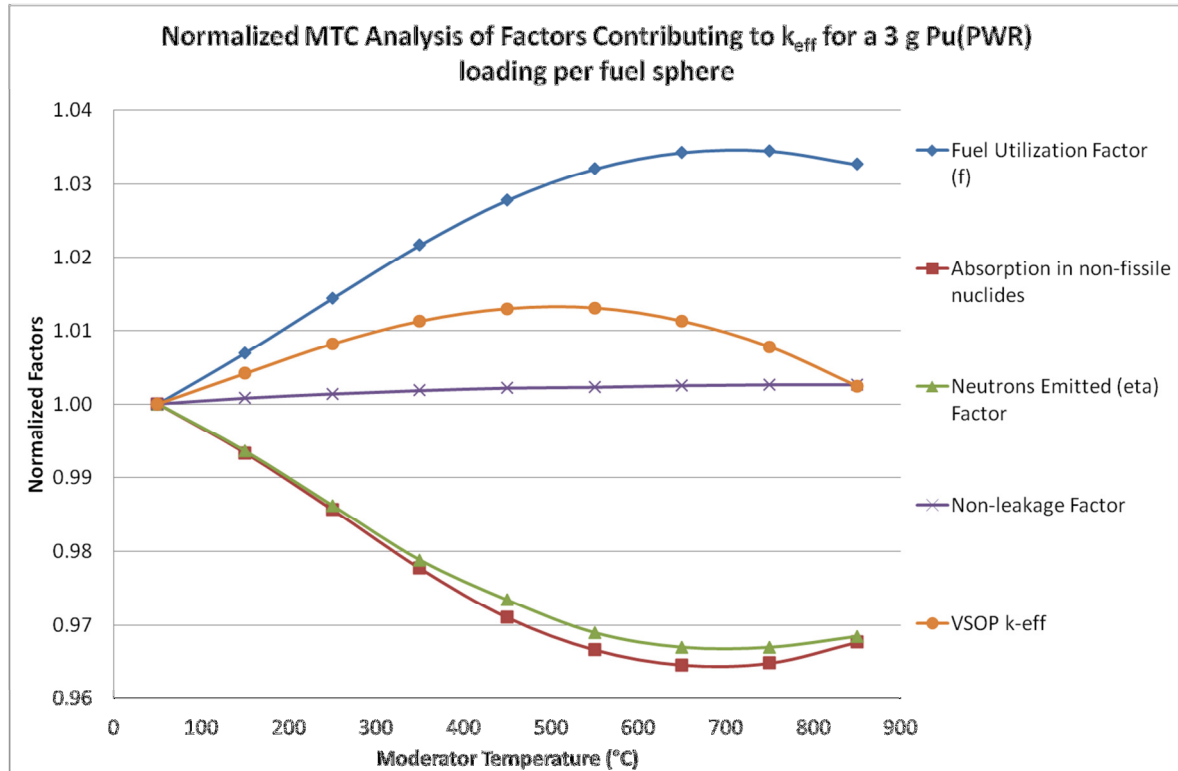


Figure 10: Normalised MTC analysis of factors contributing to the multiplication factor for a 3 g Pu(PWR) loading per fuel sphere.

From Figure 10 it is clear that the MTC is positive at low temperatures and negative at high temperatures, as k_{eff} peaks at 550 °C. The non-leakage factor marginally increases with moderator temperature. This is primarily because as the moderator temperature increases, the thermal neutron flux shifts into the fission resonances the fuel becomes less transparent and so more neutrons are absorbed in the fuel and thus fewer neutrons leave the reactor. The increase in the non-leakage factor as the moderator temperature increases was too small to be significant.

It is evident that the increase in k_{eff} at low moderator temperatures is driven by the increase in the fuel utilisation factor and the decrease in the absorption in non-fissile nuclides. These two factors dominate the opposing effect of the reduction in the η_T .

Figure 11 analyses the factors of a 6 g Pu(PWR) loading per fuel sphere as a function of increasing moderator temperature. It is clear that the maximum MTC is negative as k_{eff} only decreases with increasing temperature. It is evident that at low temperatures, similar to the 3 g Pu(PWR) loading per fuel sphere case, the fuel utilisation factor increases and the absorption in non-fissile nuclides decreases with increasing temperature, which would suggest an increase in k_{eff} . However, the effects are now much smaller than was the case for the 3 g Pu(PWR) per fuel sphere and therefore they cannot dominate the decrease in η_T .

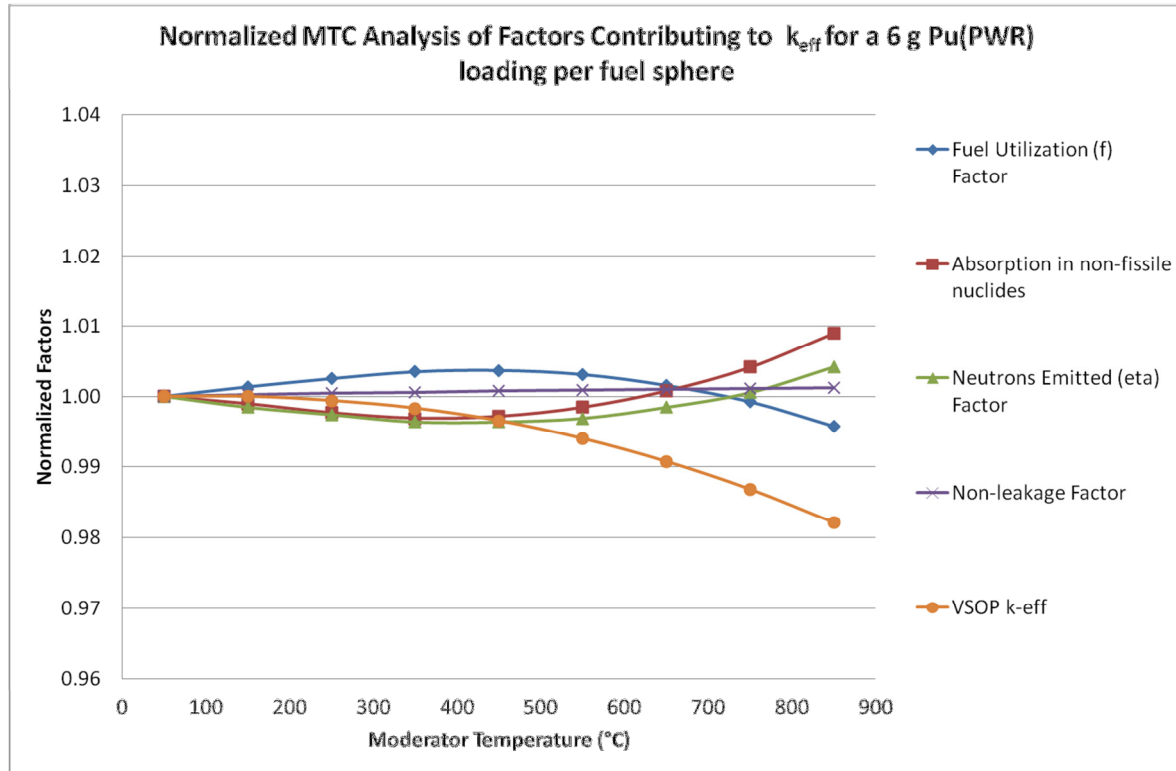


Figure 11: Normalised MTC analysis of factors contributing to the multiplication factor for a 6 g Pu(PWR) loading per fuel sphere.

The sharp increases in the microscopic fission cross-sections of both ^{239}Pu and ^{241}Pu with increasing thermal neutron energy would suggest that increasing moderator temperature should always cause more fissions and thus an increase in k_{eff} . The effectiveness of this mechanism, however, is limited by the magnitude of the thermal neutron flux. As discussed in Section 4.6.2 the thermal neutron flux is an order of magnitude less for the high Pu loading and therefore this effectiveness is sharply reduced. This is a qualitative analysis and a more rigorous quantitative analysis is required to provide certainty.

4.6.2 Comparison of Maxwellian neutron energy spectra at different temperatures to microscopic fission and capture cross-sections

Figure 12 shows the total fission and capture cross-section of ^{239}Pu and ^{241}Pu between the incident energies of 0.01 eV to 0.50 eV. The cross sections are from the ENDF/B-VII.0 library using Janis 3.4 software.

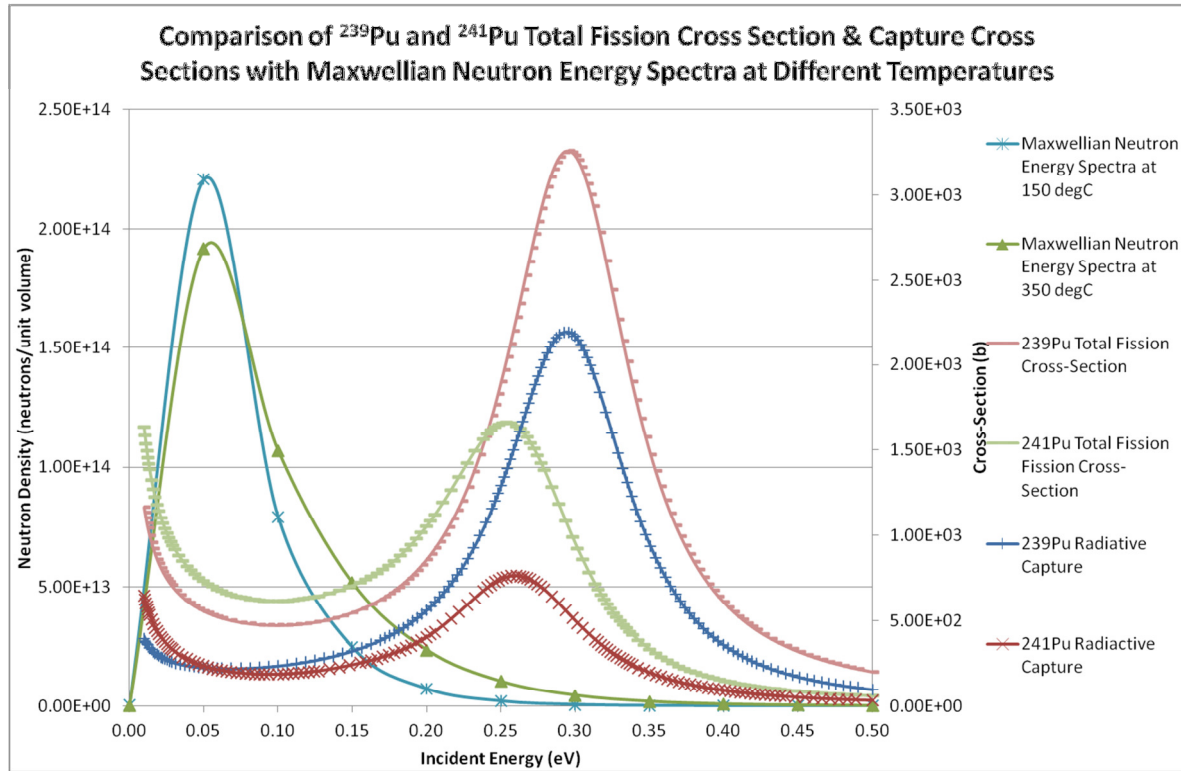


Figure 12: ^{239}Pu and ^{241}Pu total fission cross-sections and capture cross section as a function of incident neutron energy compared to Maxwellian neutron energy spectra at different temperatures.

The plots indicate that from an incident energy of 0.10 eV to 0.25 eV for ^{241}Pu and 0.30 eV for ^{239}Pu the total fission cross-sections increase with increasing energy. The figure also shows the Maxwellian neutron energy spectra for 150 °C and 350 °C. It is emphasized that the neutron density as opposed to the more relevant neutron flux is presented. Both the energy corresponding to the peak and the average energy increase with increasing temperature.

The figure above only indicates the microscopic cross-section in pure graphite, not the more relevant macroscopic cross-section. Impure or real graphite contains thermal neutron absorbers which distort the thermal neutron spectrum. The cross-sections of the different isotopes in the fuel also compete for the thermal neutrons. As a result, an increase in temperature causes an increase in absorption in ^{239}Pu and thus a decreased absorption in ^{241}Pu . The “average number of neutrons emitted per thermal neutron” absorbed in the fuel, η_T , decreases from 0.1 eV to 1 eV for ^{239}Pu (Lamarsh & Baratta, 2001:123). Therefore an increase in fissions of ^{239}Pu at higher energies may not result in an increase in k_{eff} , depending also on the change in the ratio of captures to fissions in the ^{239}Pu .

From the above discussion it is clear that increasing temperature affects many different factors. This large number of complex phenomena often act in opposing ways and it is therefore imperative that a quantitative, rather than a qualitative, analysis be performed. This is considered to be beyond the scope of this dissertation.

The average thermal flux of a 12 g Pu(PWR) and no Th loading per fuel sphere with both fuel and moderator at 150°C is $0.03 \times 10^{14} \text{ /(cm}^2\text{.s)}$. The maximum UTC for this fuel design is negative ($-0.749 \times 10^{-5} \Delta K_{\text{eff}}/^{\circ}\text{C}$). The average thermal flux of a 1.5 g Pu(PWR) and 6.5 g Th loading per fuel sphere with both fuel and moderator at 150°C is $0.24 \times 10^{14} \text{ /(cm}^2\text{.s)}$. The maximum UTC for this fuel design is positive ($+2.25 \times 10^{-5} \Delta K_{\text{eff}}/^{\circ}\text{C}$). The average thermal flux has increased by 8 times from $0.03 \times 10^{14} \text{ /(cm}^2\text{.s)}$ to $0.24 \times 10^{14} \text{ /(cm}^2\text{.s)}$.

Figure 13 below compares the average thermal flux with the maximum UTC as a function of the mass of Pu(PWR) per fuel sphere. Clear correlation between the thermal flux and of the loading of Pu(PWR) per fuel sphere can be seen from Figure 13 with the exception of the 12 g Pu(PWR) loading per fuel sphere. With more plutonium per fuel sphere, the ratio of plutonium to moderator increases. There are less thermalised neutrons. More neutrons are captured in the resonances of ^{240}Pu and ^{242}Pu as they are not moderated. Those that are moderated are absorbed soon after being moderated due to the very high Pu content, which explains the much lower average thermal flux.

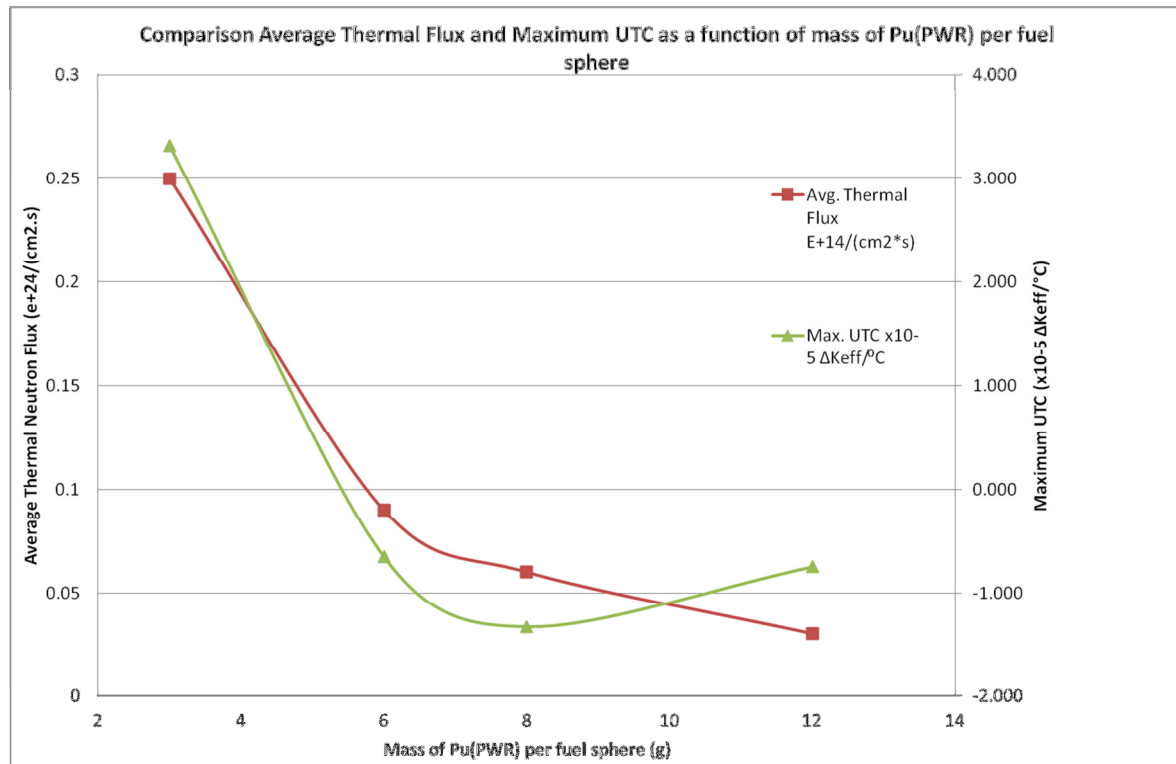


Figure 13: Comparison of average thermal flux and maximum UTC as a function of the mass of Pu(PWR) per fuel sphere.

Figure 14 plots the maximum UTC as a function of average thermal flux. The maximum UTC decreases with increasing average thermal flux up until the maximum UTC is at $-1.330 \times 10^{-5} (\Delta K_{\text{eff}}/^\circ\text{C})$.

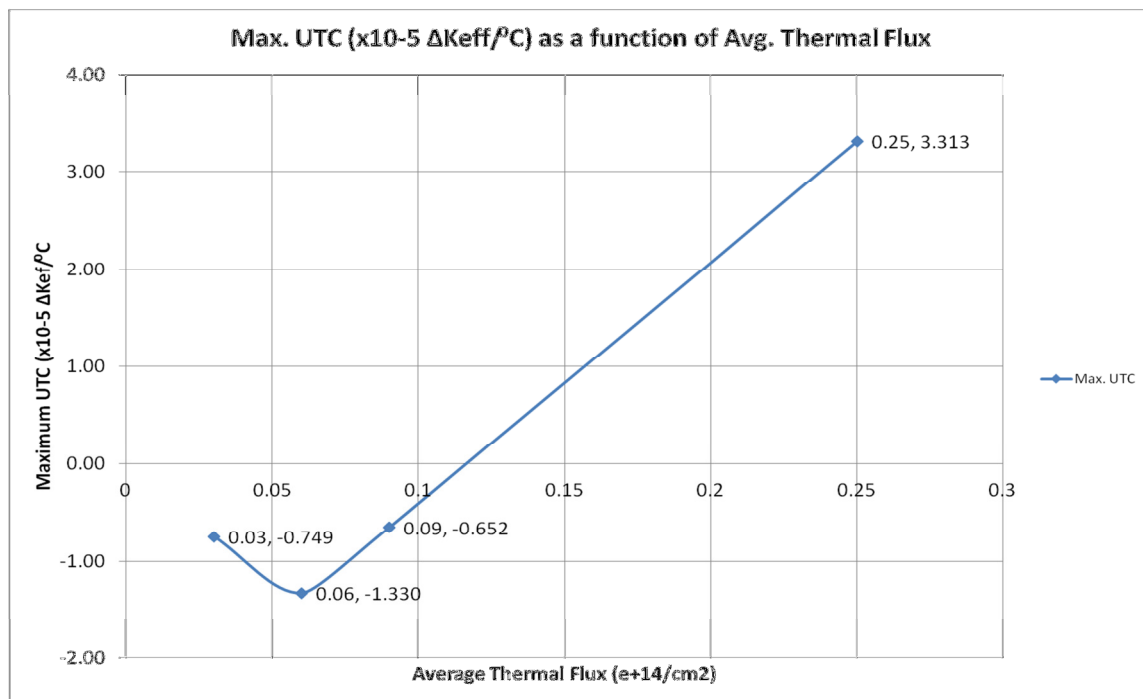


Figure 14: Maximum UTC as a function of average thermal flux.

From this point the maximum UTC increases with increasing average thermal flux. Although there are only four data points the relationship appears to be linear. A proposed explanation for the direct relationship, not the linearity, follows.

The maximum UTC reflects the contributions of both the MTC and FTC. An “upshift” of the Maxwellian thermal neutron flux spectrum into the fission resonances of ²³⁹Pu and ²⁴¹Pu with increase in moderator temperature causes a positive MTC. The decrease of the resonance escape probability due to resonances of ²⁴⁰Pu and ²⁴²Pu due to the Doppler broadening effect causes a negative FTC. As the average thermal neutron flux approaches negligible values the decrease of the resonance escape probability dominates the “upshift” of the thermal neutron flux.

5 Discussion and conclusions

This investigation led to a qualitative understanding of possible mechanisms that operate when increasing the ratio of plutonium to thorium in a fuel sphere. These proposed explanations, which will be presented in more detail below, were also found to be applicable to plutonium fuel spheres only.

The safety limits of PBMR (Pty.) Ltd. for the PBMR DPP-400 that were adopted above and that will guide the discussion regarding the achievement of the stated aims are:

1. The UTC shall be negative for the full range of temperatures. The temperature range is from ambient to 1650°C
2. The maximum power per fuel sphere shall be less than 4.5 kW per fuel sphere.
3. The fast fluence ($E > 0.1\text{MeV}$) shall be less than $8.0 \times 10^{21} / \text{cm}^2$.
4. The maximum fuel temperature shall be less than 1130°C.

5.1 Temperature reactivity coefficients

5.1.1 Influence of Th on the FTC, MTC, UTC and maximum UTC

With respect to Section 4.1 the similarities of the plotted graphs of the UTC, MTC and FTC to those from literature results provide confidence that the results of the current study are valid. They confirmed that the maximum UTC occurs at temperatures of less than 700°C.

The UTC as per literature shows a clear correlation to the MTC as the change in the magnitude of the FTC as a function of fuel temperature is much smaller than the corresponding change for the MTC as a function of moderator temperature. This change in the MTC is driven by the interaction of the change in the thermal neutron energy spectrum with neutron temperature and the changes in microscopic fission and capture cross-sections for the Pu isotopes with neutron energy. This means that the change in the resonance escape probability, due to increasing fuel temperature, is less significant than the changes in the neutron economy caused by change in the thermal neutron energy spectrum, due to increasing moderator temperature. Simplistically, an increase in fuel temperature reduces the number of neutrons that get thermalised whereas an increase in the moderator temperature does not change the number of these neutrons, but only the neutron speed and consequently flux.

Similar to literature results the FTC remained negative for the range of temperatures, but its magnitude decreased with increasing temperature. The FTC is more negative for fuels containing Pu(PWR) and Th mixtures than for pure Pu. This probably was because of the contribution of the epithermal capture resonances of Th.

5.1.2 Maximum UTC of Th and Pu and Pu only fuel

Referring to Sections 4.2 and 4.5 it was demonstrated that certain fuel designs containing thorium and plutonium and plutonium only fuel designs had negative maximum UTCs.

For a fixed heavy metal loading, increasing thorium content means reduced Pu(PWR) content. Lowering the Pu(PWR) loadings lead to a larger, positive, maximum UTC, despite the opposing trend in the FTC, due to increased Th loading, which is much smaller in magnitude. For 8 g heavy metal fuel spheres, fuel containing a mass more than 3 g Pu(PWR) per fuel sphere with the remaining mass of heavy metals thorium, the maximum UTC is negative.

However, for a fixed mass of plutonium, addition of thorium does reduce the maximum UTC. The 3 g Pu(PWR) and the 5 g Th loading per fuel sphere is of interest because it has a negative maximum UTC while the 3 g Pu(PWR) loading per fuel sphere has a positive maximum UTC. This clearly demonstrates that the addition of Th while maintaining a fixed mass loading of Pu(PWR) per fuel sphere achieves a negative maximum UTC. The only safety limit that was exceeded was the maximum fuel temperature however maximum fuel temperatures can be reduced by modifying the PBMR DPP-400 design such that there are poisons in the fuel chute.

From literature a 3 g Pu(PWR) only fuel sphere has a positive maximum UTC (Serfontein, 2011). However, at high masses of plutonium such as higher than 6 g Pu(PWR) the maximum UTC is negative. This is an important finding as introducing thorium generates ^{233}U . As there is initially no ^{238}U this ^{233}U may potentially be chemically separated and apart from the radiological hazard to would be proliferators, would make excellent weapons fuel. (Serfontein, 2011, Section 1.4.5.2).

5.2 Other PBMR DPP-400 safety limits

5.2.1 Fast fluence

The higher the fast fluence the more likely it is that a coated particle will be ruptured. The primary containment of fission products is the coatings around of the fuel kernels. The longer a fuel sphere is in a reactor, the more fast neutrons the fuel sphere and the coated particles within the fuel sphere are exposed too. As the thermal power of the reactor is fixed, an increase in burn-up results in a longer time within the reactor. A higher burn-up has a higher fast fluence. Figure 6 shows this result.

The data on achievable plutonium burn-up with conventional coatings is limited. It would therefore be conservative to first construct plutonium incinerators with lower burn-up.

5.2.2 Maximum power per fuel sphere and maximum fuel temperature

The trend of the maximum power per fuel sphere in an 8 g heavy metal per fuel sphere was determined mainly by the mass of plutonium, not the mass of thorium as shown in Figure 7. An increased Pu loading results in a lower moderation ratio and thus lower resonance escape probability and a faster absorption of neutrons once they are thermalised. The combination of these effects resulted in a lower thermal neutron flux. However these negative influences are not at work in the reflectors and therefore the thermal flux in the reflectors and thus the influx of neutrons from the reflectors into the fuel layers directly adjacent to the reflectors are not similarly reduced by increasing Pu loading. Therefore it follows logically that increased Pu loading will lead to a sustained high thermal neutron flux and thus high power in the fuel spheres directly adjacent to the reflectors, as opposed to a much reduced thermal flux and power in fuels spheres deeper into the fuel core. This will lead to a sharp peak in the radial power profile directly adjacent to the central reflector, as was demonstrated by (Serfontein, 2011: Section 3.4.3). This will then also lead to higher equilibrium fuel temperatures in these power peaks.

The mechanism for the maximum fuel temperatures varies according to where in the reactor the maximum fuel temperature occurs. It was shown from the literature that the maximum fuel temperature tends to in the fuel chute. If this occurs in a reactor designed to burn plutonium this can be eliminated by putting neutron poisons in the chute.

5.3 Dependency of Maximum UTC to average thermal flux

5.3.1 Relationship of low average flux to a negative UTC

A six factor analysis of the UTC was not satisfactory as it did not distinguish between thermal and epithermal neutrons and the definitions used by VSOP did not always match the theoretical definitions from the literature. Similar analysis in Section 4.6.1 of the MTC showed that the increasing fuel utilisation factor and the decreasing absorption in non-fissile nuclides increased k_{eff} whereas the decreasing η_T decreased k_{eff} . With high average thermal neutron fluxes this fuel utilization effect was dominant. However, at low to negligible neutron fluxes the effect of the fuel utilization is of no consequence. In summary, negative maximum MTCs and UTCs are obtained for low average thermal fluxes and *vice versa*.

Analysis of the results as per Figure 14 of this study indicated that the average thermal neutron flux is significantly lower for fuel designs that contain a negative UTC. Detailed consideration of the Maxwellian neutron temperature spectra suggests that a quantitative as opposed to a qualitative analysis is required to identify the major driver mechanisms behind the observed results. Such an analysis was therefore referred to the suggestions for future study. This analysis should include macroscopic cross-sections as well as thermal neutron absorbers in real graphite.

5.4 Economic considerations of a licensable fuel design concept

5.4.1 Licensability of Pu only fuel designs

The fuel design of 12 g Pu(PWR) loading per fuel sphere characteristic parameters meets the maximum UTC safety limits and the remaining safety limits of maximum power per fuel sphere, fast fluence and maximum fuel temperatures (see Table 7). However, it is also noted that the safety limits, specifically the maximum fuel temperature and negative temperature coefficient, may need to be more stringent for plutonium incineration and a Rankine cycle. The reasons for this are that plutonium generates ten times as much radioactive silver per fission (see Section 3.7) and a water ingress accident requires a greater magnitude for the negative temperature coefficient in order to counter the resulting reactivity and thus power surges. .

5.4.2 Optimisation of economic versus disposal and proliferation objectives

A principal feasibility factor is the cost of the fuel sphere manufacture. The proposed PBMR 9 g U/Pu (LEU) loading fuel sphere has a lower burn-up of 90.8 GWd/tHM (Reitsma, 2004: Section 4.1). A 12 g Pu(PWR) loading per fuel sphere with no Th produced approximately double the burn-up at 180.7 GWd/tHM.

A low burn-up has the following disadvantages:

1. The energy obtained per fuel sphere decreases and therefore the cost recovered per manufactured fuel sphere decreases.
2. Spent fuel with a high mass of plutonium is sub-optimal for direct to disposal because it still contains toxic plutonium and highly radioactive Minor Actinides (MA).
3. The isotopic composition of the plutonium may indicate that proliferation objectives are not optimally achieved.

The spent fuel of 12 g Pu(PWR) loading per fuel sphere analysis (see Section 4.5) showed that 24.5 % of the loaded Pu was incinerated, as opposed to 69.2 % for the 3 g pure

Pu(PWR) loading. Thus the 12 g Pu(PWR) fuel design is sub-optimal with respect to proliferation and waste disposal objectives.

5.5 Final conclusions

Within the constraints of this study the general and specific aims were achieved and the following hypotheses were shown to be true:

1. *There is a conceptual fuel design that contains a mixture of plutonium and thorium where the maximum UTC is negative for the full range of temperatures.*
2. *There is a conceptual fuel design that contains a specific mass of only plutonium fuel where the maximum UTC is negative for the full range of temperatures.*
3. *Sufficiently lowering the average thermal flux of plutonium base fuel designs by increasing volume fraction of the coated fuel particles will induce negative UTCs over the full range of temperatures.*
4. *There is a conceptual fuel design to economically burn reactor grade plutonium which meets all of the PBMR safety limits and is therefore conceptually licensable.*

In summary there is a fuel design containing thorium and plutonium that achieves a negative maximum UTC. Further it has been shown that a fuel design containing only plutonium (12g Pu(PWR) loading per fuel sphere) achieves a negative maximum UTC. The parameters of this fuel design meet the other safety limits of maximum power per fuel sphere, fast fluence and maximum fuel temperature. As a result this fuel design is potentially licensable.

The 12g Pu(PWR) loading per fuel sphere design has double the burn-up of the proposed PBMR fuel design and may possibly be shown to be commercially viable. Utilising a fuel design consisting of a high plutonium heavy metal loading has the disadvantage that the mass of plutonium in the spent fuel increases compromising proliferation and disposal objectives.

6 Recommended further research studies

The following follow-on studies are proposed.

6.1 Quantitative analysis to investigate the UTC

There are several complex factors that determine the dependence of k_{eff} on temperature. This simple qualitative analysis of the summary data from VSOP performed in this study could not identify a single dominant driver for these dependencies. A more detailed quantitative analysis to fully understand the mechanisms involved is thus recommended. This analysis should reconstruct the summary parameters from the more detailed output of VSOP, in order to bring the definitions of the VSOP parameters in line with the theoretical definitions from the literature, by amongst others distinguishing between absorptions in the epithermal and thermal energy regions. Attention should also be paid to the macroscopic, as opposed to only the microscopic, cross-sections and realistic graphite nuclide compositions. The theoretical Maxwellian neutron flux spectra should also be replaced with the real spectra, as they are distorted by absorption cross-section peaks for the different nuclides.

6.2 Potential of Th to achieve a negative UTC

For the simulations where the Pu(PWR) content in a Pu and Th mixture with a fixed heavy metal content of 8 g per fuel sphere it was determined that the plutonium loading rather than the remaining thorium dominated the potential to achieve a negative UTC. The potential of Th to achieve negative UTC's was sidelined because of the generation of ^{233}U without ^{238}U . However it was demonstrated that increasing the mass of Th while keeping the mass of plutonium constant did significantly decrease the maximum UTC. Further studies could explore this potential of higher heavy metal loadings with a large Th fraction where the mass of Pu(PWR) remains at 3 g per fuel sphere, while also considering the generation of ^{233}U .

6.3 Further investigation of only Pu(PWR) loaded fuel spheres

As per Figure 14 it was identified that the maximum UTC decreases with increasing average thermal flux before increasing with average thermal flux. This would indicate the FTC dominates at low thermal fluxes. Determination of the FTC for the Pu(PWR) loaded fuel spheres would possibly provide the ability to validate this proposition.

The analysis could also include investigating the change in the capture to fission ratio of ^{239}Pu and capture to fission ratio of ^{241}Pu with respect to temperature and the effect this has on the MTC.

6.4 Evaluation of shell fuel sphere versus standard fuel sphere configurations

In further studies it is recommended that the present shell fuel sphere (i.e. where the central graphite sphere is surrounded by a fuel matrix shell, which is then again surrounded by the outer graphite shell) be replaced with a standard fuel sphere (i.e. a central fuel matrix sphere surrounded by the outer graphite shell). The shell fuel sphere configuration helps to reduce the temperatures of the inner coated fuel particles as these are closer to the fuel sphere surface and are thus more easily cooled by the helium coolant gas.

The negative issues associated with the shell fuel sphere configuration are as follows. Firstly, a shell fuel sphere configuration may result in a higher mechanical breakage of fuel spheres than a standard fuel sphere. Secondly the higher volume fraction of the coated

particles required to achieve the specified heavy metal loading can result in failure of the kernel coatings during the pressing of the fuel spheres. If the maximum fuel sphere temperatures can be reduced to acceptable levels, by for instance adding neutron poisons to the fuel exit chutes, the standard fuel sphere configuration would be more appropriate for a practical reactor.

6.5 Refinement of fuel and reactor designs to meet all reactor and proliferation requirements

6.5.1 Compliance of fuel and reactor design to criteria

The objective of this study has been to focus on the maximum UTC. It is recommended that further analysis of this fuel is performed to investigate whether other criteria of the reactor such as a Depressurised Loss of Forced Cooling (DLOFC) are met.

6.5.2 Proliferation resistance of spent fuel composition

An investigation is required to determine whether the isotopic composition of the spent fuel or chemically separated components of the spent fuel are suitable for the construction of usable nuclear weapons. This would determine if this reactor and fuelling strategy is suitable for reducing the global inventory of proliferation resistant plutonium. The suitability of the spent fuel for incineration in an alternative medium-term reactor design should be assessed.

6.6 Comparison of the 400 MWth PBMR DPP-400, 200MWth HTR-MODUL and HTR-PM

The South African government announced in 2010 that it would not build the PBMR DPP-400. The Chinese are constructing the HTR-PM, which is a twin 200 MWth each producing 105 MWe (World Nuclear Association, 2012). As the smaller reactor currently has more relevance, it is recommended that further work on plutonium incineration in pebble bed reactors should focus in this area.

The contributors to the Chinese research project investigated a fuel design containing plutonium with thorium. They demonstrated with VSOP-94 that a negative temperature coefficient could be achieved at the equilibrium temperature utilising the HTR-MODUL. This needs to be further investigated with VSOP-99/05 to show that a negative UTC exists for the range of temperatures. A further step would be to simulate Pu incineration in the currently operating Chinese HTR-PM reactor, which would open up the opportunity to test the simulated fuel cycles in a working research reactor

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