

Isotopic and spectral effects of Pu quality in Th-Pu fueled PWRs

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ARTICLE INFO

Article history:

Received 20 November 2017

Received in revised form 10 March 2018

Accepted 14 March 2018

Available online 5 April 2018

Keywords:

Thorium

Plutonium

Disposition

LWR

Reduced moderation

MTC

ABSTRACT

UK plutonium (Pu) management is expected to focus on the use of uranium-plutonium (U-Pu) mixed oxide (MOX) fuel. However, research has shown that thorium-plutonium (Th-Pu) may be a viable alternative, offering favourable performance characteristics. A scoping study was carried out to determine the effect of isotopic composition and spectral hardening in standard and reduced moderation Pressurised Water Reactors (PWRs and RMPWRs). Lattice calculations were performed using WIMS to investigate safety parameters (Doppler Coefficient (DC), Moderator Temperature Coefficient (MTC), Void Coefficient (VC) – in this case Fully Voided Reactivity (FVR) – and Boron Worth (BW)), maximum theoretically achievable discharge burnup, Pu consumption and transuranic (TRU) composition of spent nuclear fuel (SNF) for the two reactor types. Standard grades of Pu were compared to a predicted UK Pu vector.

MTC and FVR were found to be strongly influenced by the isotopic composition of the fuel. MTC was determined to be particularly sensitive to positive ‘peak’ contributions from fissile isotopes in the energy range 0.1–1 eV which diminish as the Pu content increases. The more extreme nature of the perturbation in FVR cases results in key differences in the contributions from fissile isotopes in the thermal energy range when compared with MTC, with no positive contributions from any isotope <500 eV.

Where the requirement for MTC to remain negative was the limiting factor, a higher maximum fissile loading, discharge burnup and Pu consumption rate were possible in the PWR than the RMPWR, although the two reactors types typically produced similar levels of U233. However, for the majority of Pu grades the total minor actinide (MA) content in SNF was shown to be significantly lower in the RMPWR. Where FVR is the limiting factor, the maximum fissile loading and discharge burnup are similar in both reactor types, while increased Pu consumption rates were possible in the PWR. In this case, lower concentrations of U233 and MAs were found to be present in the PWR. These results are for a single pass of fuel through a reactor and, while the response of fissile isotopes at given energies to temperature perturbations will not vary significantly, the maximum achievable discharge burnup, Pu consumption rate and TRU build-up would be very different in a multi-recycle scenario.

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1. Introduction

1.1. UK plutonium

The UK Pu stockpile is the largest separated Pu stockpile in the world, containing 120 tonnes of spent fuel from a variety of sources of varying initial fissile loading, discharge burnups and cooling periods (Institution of Mechanical Engineers, 2013; Nuclear Decommissioning Authority, 2010). The result is a varied isotopic mix in SNF which can be recycled using current reactor

technology. Using existing technology to reduce the volume of Pu in the stockpile could result in lower developmental costs for an accepted recycle scheme, in addition to the technical advantages associated with decreased radiotoxicity and decay heat burden of stored SNF (Lindley et al., 2015a, 2016; Ernout et al., 2015; Ashley et al., 2014; Kamei and Hakami, 2011; Schram and Klaassen, 2007; Weber et al., 1997). The management strategy for UK Pu is expected to focus on the use of MOX fuel in thermal reactors as an interim measure prior to a fast reactor fleet being commissioned (Nuclear Decommissioning Authority, 2014; Institution of Mechanical Engineers, 2013; Department of Energy and Climate Change, 2011; International Atomic Energy Agency, 2003b; The Royal Society, 1998). MOX fuel is typically manufactured using U238 as the fertile isotope mixed with plutonium

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(Haas and Hamilton, 2007). However, spent U-Pu MOX fuel still contains significant quantities of Pu – roughly 50% of the initial loading – due to Pu production from U238. For the purposes of Pu incineration, it may therefore be beneficial to use Th232 as the fertile isotope rather than U238. Although less prevalent than U-fuels, Th-Pu options have been substantially researched since the 1980s – both from R&D and operational points of view – with results showing that Th-Pu MOX may be an ideal platform for Pu incineration in Light Water Reactors (LWRs) (Insulander Bjork and Kekkonen, 2015; Insulander Bjork, 2013; Lindley and Parks 2012a,b; International Atomic Energy Agency, 2002). Th breeds U233 as the fissile component of the fuel which accounts for >90% of isotopes in the Th transmutation chain and results in a significantly lower Pu and MAs content in discharged fuel compared to standard U-Pu MOX fuels (International Atomic Energy Agency, 2005; Galperin, 1995; Shapiro et al., 1977). Th fuels show potentially lower levels of radiotoxicity and decay heat compared to standard UO₂ and U-Pu fuels after the first 100 years post discharge (Hesketh and Thomas, 2013). This, in addition to volume reduction, holds promise for UK Pu recycle schemes using Th MOX in LWRs.

1.2. Reactor type

The UK has extensive operating experience of PWRs through civil and naval nuclear programmes. The effect of moderation in PWRs has been well studied and has shown that increased, standard and reduced moderation can have specific benefits relating to Pu incineration depending on the desired objective of Pu disposition. (Shwageraus et al., 2004) determined that for once-through recycle options using low Pu loadings in Th-Pu MOX (with no MAs), reduced moderation led to unacceptably short cycle lengths due to rapid depletion of Pu239. However, at higher Pu loadings, the production of U233 and the subsequent competition between this and Pu239 led to a decrease in Pu destruction in line with the decrease in moderation. Adding MAs to the Th-Pu MOX showed that increased destruction rates were possible only by increasing moderation. However, the addition of MAs reduced the overall destruction rate by ~10% compared to Th-Pu only. (Lindley and Parks 2012a,b) considered multiple recycle options and showed that TRU material can be virtually eliminated in Th-Pu-MA fueled, standard moderation PWRs. Further studies showed that reducing moderation in multiple recycle schemes led to improved performance when compared with standard PWRs – notably a ~20% increase in achievable burnup when compared to standard PWRs with the same fissile loading (Lindley and Parks 2012a,b). This is important where the moderator reactivity coefficients limit the maximum fissile loading, and therefore total achievable burnup, in the fuel – particularly where a negative VC/FVR is required for a fully voided core. It is possible to achieve a negative FVR in RMPWRs; however, this requires targeted fuel management strategies (Lindley et al., 2014). In reality, a fully voided PWR core which has not been scrambled is deemed to be extremely unlikely and, while FVR remains of regulatory importance, it is the MTC which will likely be the limiting factor for fissile loading. It is ultimately for policy makers to decide whether once-through or multiple recycle options are preferred and, as no decision has yet been made in the UK, both standard and reduced moderation options are considered in this study. This assembly-level study will determine the sensitivity of reactivity feedback coefficients to the isotopic composition of fresh fuel and the effects of spectral hardening in PWRs and RMPWRs when loaded with the predicted UK Pu vector and other 'standard' grades of Pu. Coefficients will be calculated for each grade of Pu for various Pu loadings to determine the maximum theoretically achievable discharge burnup and Pu consumption taking account of the requirement to maintain negative

temperature coefficients of reactivity. This may be used to help inform policy makers of the maximum Pu loading for each Pu vector and, therefore, provide an indication of how quickly the stockpile can be reduced. However, these results merely provide an insight into potential options and cannot be used to make a decision without full-core analyses being undertaken.

1.3. In-core safety parameters

Many of the parameters affecting the overall reactivity of a core depend on changes in material temperature. These changes can occur as a result of transient or accident scenarios and hence it is crucial to be able to predict the outcome of variations in temperature prior to licensing reactors to operate with new fuel types. While previous studies have shown that it is possible to achieve more favourable temperature coefficients of reactivity in LWRs with Th-Pu MOX fuel than with standard UO₂ fuel (Shwageraus et al., 2004; Weaver and Herring, 2003; Lombardi et al., 1999; Galperin and Raizes, 1997), no studies have thus far considered in detail how UK Pu – subject to the effect of isotopic decay – may perform during operation. It is vital to understand the impact that the isotopic composition of UK Pu may have on reactivity feedback coefficients from a safety and control point of view, especially given that Th-fuels are associated with greater control requirements than UO₂ fuels due to the lower delayed neutron fraction of U233 and Pu239 compared to U235 (Alhaj et al., 2016; Lau et al., 2014; Fridman and Kliem, 2011; Lamarsh and Baratta, 2001). Reactors with negative temperature coefficients of reactivity are considered inherently stable whereas reactors with positive temperature coefficients of reactivity are unstable and should be avoided. Larger values indicate a greater sensitivity to changes in the parameter that has been varied.

The coefficients that will be investigated in this study are DC, MTC and FVR along with BW. DC – change in reactivity per degree change in fuel temperature – is caused by Doppler broadening of resonant absorption peaks of isotopes within the fuel. Resonant peaks typically exist in the epithermal energy range and are therefore more likely to affect harder spectrums such as reduced moderation and high Pu content cases. DC must remain negative during operation.

MTC – change in reactivity per degree change in moderator temperature – determines the ultimate behaviour of a reactor in response to changes in moderator temperature. In LWRs an increase in moderator temperature causes a reduction in density of water due to thermal expansion. This can result in either a positive or negative reactivity insertion. Several factors can influence MTC:

- Resonance escape probability: A lower density reduces the effectiveness of the moderator at slowing down neutrons through the resonance region, resulting in an increase in resonance absorption and a consequent decrease in the resonance escape probability (negative reactivity insertion). Alternatively, there may be increased fission in the epithermal/fast region (positive reactivity insertion).
- Thermal utilization: The lower density causes the thermal utilization factor to increase due to fewer parasitic absorptions of neutrons by hydrogen nuclei in the light water (positive reactivity insertion).
- Leakage: reduced density affects the non-leakage probability of neutrons across the entire energy range. As density decreases, there will be fewer neutron collisions with light water, resulting in an increase in the average neutron energy in the system. As the spectrum hardens, and the capture cross-sections of fuel nuclides decrease, more fast neutrons will be lost from the core (negative reactivity insertion).

- Interactions with absorbers: this can affect reactivity in a variety of ways depending upon whether they are present in solution or fixed. This study will consider the effect of soluble boron, but no other neutron poisons will be included.

MTC is therefore a complex trade-off of positive and negative reactivity contributions. Since the main influencing factor to MTC is typically the resonance escape probability, and each nuclide has a different resonant structure, the MTC is very sensitive to the isotopic composition of the fuel. The contribution of different isotopes must therefore be well understood and, as such, nuclide contribution to MTC will be studied in the later part of this analysis.

VC – change in reactivity per percent voiding in the light water of the moderator/coolant – determines the behaviour of a reactor in response to loss of coolant accidents (LOCAs). In this case a near-complete loss of coolant is simulated (representing a fully voided core (FVR)). Like changes in moderator temperature, voiding also changes the coolant density. As with the MTC in LWRs, an increase in voiding (and subsequent decrease in density) results in a reduction in reactivity if the thermal absorption cross-section of nuclides in the water is not made too large through the addition of soluble boron. In this case, a reduction in density may result in an increase in reactivity due to the reduction in density of boron. Scenarios which result in a fully voided but unscrammed core are deemed unlikely, but this will still be analysed to satisfy regulatory constraints.

Since the addition of soluble boron is being considered, BW will also be analysed. BW – change in reactivity per ppm change in boron concentration – typically becomes more negative with burnup as the boron levels are highest at beginning of cycle (BOC) when the excess reactivity is most substantial.

1.4. Practical considerations

From an operational and regulatory point of view there are a number of key factors that must be addressed. The maximum achievable discharge burnup should be given as this dictates the cycle length of a given fuel type; the rate of Pu destruction should be considered as this may influence policy decisions; and information regarding TRU accumulation in SNF should be provided.

While the maximum discharge burnup is an important factor from a commercial point of view, it is also crucial to in-core safety. The current cladding limit for PWRs is ~60 Gwd/tHM (Nuclear Energy Agency, 2006). Although cladding failure can occur above this limit, research is ongoing in the field of advanced cladding technology and, therefore, higher burnups will be considered in the final part of this study.

2. Method

2.1. Model and code

Assembly-level calculations were performed for a standard 17×17 PWR lattice with reference geometry and operational parameters outlined in Table 1. The soluble boron concentration was fixed at 500 ppm for the purposes of comparison. Each assembly contains 264 fuel pins with zircaloy cladding and 25 water holes. Increasing the fuel pin diameter from 9.5 mm in the PWR to 11.0 mm in the RMPWR causes the hydrogen-to-heavy-metal (H/HM) ratio to decrease and the neutron spectrum to harden. The increased diameter has been shown to allow for a neutronically feasible design which fulfils thermal-hydraulic constraints and allows multiple reloads to take place if required (Lindley, 2014). The cladding and gap thickness have been kept constant

Table 1
Reference assembly geometry and operating conditions.

Parameter	PWR	RMPWR
Fuel pellet radius (cm)	0.4095	0.4845
Gap thickness (cm)		0.0085
Cladding thickness (cm)		0.0570
Lattice pitch (cm)		1.26
Fuel temperature (K)		900
Cladding temperature (K)		600
Moderator temperature (K)		585
Moderator density (g/cc)		0.7007
Boron concentration (ppm)		500
Rating (MW/tHM)		38

between the two models to maintain the basis of comparison. However, it may be the case that for higher burnups the cladding thickness needs to be increased in the RMPWR to account for the higher stresses caused by the increased pin diameter. In addition, an increased gap thickness or larger plenum may be needed to contain the additional fission gas release compared to the standard PWR. This will be considered in future work along with the addition of control rods and fixed burnable absorbers. The theoretical densities of PuO₂ and ThO₂ were assumed to be 11.5 g/cc and 10.0 g/cc respectively. For all fuel materials used, the assumed density is 95% of their corresponding theoretical density. A 3-batch loading scheme was assumed with fuel being replaced on average every 18 months, equating to a maximum core-time of ~4.5 years per assembly.

Simulations were carried out to compare the effect of varying the Pu vectors (Weapons grade (WG), Reactor grade (RG), MOX grade (MOX) and UK Pu) (Table 2) and %wt Pu on homogeneously mixed Th-Pu MOX fuel in both reactor types. Realistically, one may wish to consider matching the overall reactivity of the assembly rather than simply the composition and percentage loading. However, since the main objective of this study is to understand the effect of isotopic composition and spectrum changes on reactivity feedback coefficients, this was not considered as part of this work.

WIMS10A (Lindley et al., 2015b; Askew et al., 1966) was used to perform the analysis using the ENDF/B-VII data library (Chadwick et al., 2006). The model used to complete this analysis was benchmarked against published data for 5%wt RG Pu in Th-Pu MOX fuel in a 17×17 PWR lattice (International Atomic Energy Agency, 2003a). Good agreement was found between the model used in this study and the published data. The minor discrepancies are attributable to the older, unspecified codes and data libraries used by the states participating in the IAEA study. Additional validation was carried out using SERPENT and results showed good agreement with WIMS in all cases considered.

The results of this study were generated by first performing an approximate flux solution using 172 neutron energy groups with geometric approximations inherent to the code (Lindley et al., 2017), and then by performing a more detailed final solution using 47 groups and the method of characteristics. Thermal and epithermal region energies were split into a larger number of groups than

Table 2
Isotopic composition of plutonium vectors.

Grade	Nuclide composition (%wt)					
	Pu238	Pu239	Pu240	Pu241	Pu242	Am241
WG	0.012	93.800	5.800	0.350	0.022	0.000
RG	1.300	60.300	24.300	9.100	5.000	0.000
MOX	1.900	40.400	32.100	17.800	7.800	0.000
UK Pu	0.240	66.080	25.660	1.690	2.420	3.910

(Gill, 2016; Mark, 1993).

Table 3
Perturbed operating conditions.

Coefficient	Perturbation
DC	Fuel temperature +50 K
MTC	Moderator temperature +5 K
FVR	Moderator density reduced to 0.6886 g/cc
BW	Moderator density reduced to 0.0100 g/cc ¹ Boron concentration + 50 ppm

¹ Representing an extreme LOCA.

the fast region energies, as these represent the energy ranges where the majority of interactions occur in standard and reduced moderation PWRs. Burnup calculations were performed and perturbations to temperature, density and boron concentration were simulated.

2.2. Reactivity feedback coefficients

Changes in reactivity for DC, MTC, FVR and BW were calculated using Eq. (1).

$$\Delta\rho = \frac{k_{\infty}^p - k_{\infty}^n}{k_{\infty}^p \cdot k_{\infty}^n} \frac{1}{\Delta X} \quad (1)$$

where n and p refer to nominal and perturbed conditions respectively and X is the change in temperature, % voiding, or boron concentration. Nominal operational conditions are defined in Table 1 while perturbed conditions are shown in Table 3.

2.3. Isotopic contribution to reactivity feedback coefficients

Once reactivity feedback coefficients are calculated, the contributions to the overall MTC can be broken down further to determine the isotopes of key significance (Ganda and Greenspan, 2010). This can be used to predict the reactivity response of a given isotopic mixture to changes in moderator density. Contributions are analysed firstly by isotope and then by energy group (Zainuddin et al., 2016). As described by (Ganda and Greenspan, 2010), where “the major contributors to the reactivity coefficient are fissile isotopes, it is more convenient to rank the constituent contribution by the number of fission neutrons they emit per neutron absorbed in the system”. Normalizing “per absorbed neutron”, k_{∞} is broken down as per Eq. (2).

$$\begin{aligned} k_{\infty} = \eta f &= \frac{\sum_j v_j \sum_{fj} \Phi_j}{\sum_i \sum_{ai} \Phi_i} = \frac{v_1 \sum_{f1} \Phi_1}{\sum_i \sum_{ai} \Phi_i} + \frac{v_2 \sum_{f2} \Phi_2}{\sum_i \sum_{ai} \Phi_i} + \dots \\ &= \frac{v_1 \sum_{f1} \Phi_1}{\sum_{a1} \Phi_1} \cdot \frac{\sum_{a1} \Phi_1}{\sum_i \sum_{ai} \Phi_i} + \frac{v_2 \sum_{f2} \Phi_2}{\sum_{a2} \Phi_2} \cdot \frac{\sum_{a2} \Phi_2}{\sum_i \sum_{ai} \Phi_i} + \dots \end{aligned} \quad (2)$$

Defining η and f as:

$$\eta_j \equiv \frac{v_j \sum_{fj} \Phi_j}{\sum_{aj} \Phi_j}; \tilde{f}_j \equiv \frac{\sum_{aj} \Phi_j}{\sum_i \sum_{ai} \Phi_i} \quad (3)$$

k_{∞} may then be described by:

$$k_{\infty} = \eta_1 \tilde{f}_1 + \eta_2 \tilde{f}_2 + \dots \quad (4)$$

where:

η_j is the total fission neutrons produced per neutron absorbed in component j

\tilde{f}_j is the fraction of neutrons absorbed in component j from the total neutrons absorbed in all system constituents. This definition differs slightly from the standard thermal utilization definition.

Change in reactivity due to perturbations in moderator density can then be defined as per Eq. (5).

$$\begin{aligned} MTC &= \frac{k_{\infty}^p - k_{\infty}^n}{k_{\infty}^p \cdot k_{\infty}^n} \frac{1}{\Delta T} \\ &= \frac{1}{\Delta T} \left(\frac{\eta_1 \tilde{f}_1|_p - \eta_1 \tilde{f}_1|_n}{k_{\infty}^p \cdot k_{\infty}^n} \right) + \left(\frac{\eta_2 \tilde{f}_2|_p - \eta_2 \tilde{f}_2|_n}{k_{\infty}^p \cdot k_{\infty}^n} \right) + \dots \end{aligned} \quad (5)$$

An initial benchmark study was performed using this method and a limited number of results from the original paper (Ganda and Greenspan, 2010). Good agreement was achieved for the majority of nuclides contributing to the calculated coefficients. Discrepancies were found to exist when comparing contributions from U238 due to improved representation of the U238 resonances in the ENDF/B-VII.0 data library used in this study compared to the ENDF/B-V data library used in the original study (Forget et al., 2014).

2.4. Reactivity limited discharge burnup, Pu destruction and MA build-up

The maximum achievable discharge burnup for each grade was calculated using the Linear Reactivity Model (Driscoll et al., 1991) based on a 3-batch management scheme. For each of these batch burnups the MTC and FVR were recorded to provide an indication of what may be limiting factors for Pu loading. The MTC and FVR values quoted for BOC and EOC are 3-batch averages. The first batch is assumed to be shuffled when k_{∞} drops below 1.03.

Pu destruction rates have been calculated by dividing the difference in mass of Pu (kg/tHM) in fresh and spent fuel by the reactivity limited discharge burnup (GWd/tHM) for each Pu vector in each reactor type, while the accumulation of MAs is simply the total amount of MAs present in discharged fuel (kg/tHM) for each Pu vector in each reactor type.

3. Results and discussion

3.1. Neutron multiplication

Fig. 1 shows the k_{∞} vs. burnup curves for each of the Pu grades considered in both the PWR and RMPWR at 5%wt Pu loading. For most cases considered there is, as expected, a steady decrease in k_{∞} with burnup. However, UK Pu displays a slight increase in k_{∞} between BOC and middle of cycle (MOC) in the both the PWR and RMPWR, while RG displays a similar increase in the RMPWR. This becomes less apparent as Pu content increases.

The only significant isotope whose concentration increases early in the cycle is fissile Pu241 (Fig. 2), resulting in the slight increase in k_{∞} in RG and UK Pu shown in Fig. 1. The increase in k_{∞} is greater in UK Pu than RG as, despite the overall Pu241 concentration being higher in RG due to the isotopic composition of the fresh fuel, the difference in Pu241 concentrations between BOC and EOC is much greater in UK Pu. The RMPWR shows a more pronounced increase in k_{∞} than the PWR due to the higher concentrations of Pu241 in the RMPWR. To a lesser extent, Am242m production also influences the k_{∞} curves (Fig. 3). Again, the effect is greater in UK Pu compared to RG and in the RMPWR compared to the PWR.

Fig. 1 also shows that RG and UK Pu display significant differences in k_{∞} at different stages of burnup despite having a similar Pu composition for fresh fuel. At BOC the difference in k_{∞} is due to the presence of Am241 in UK Pu. Given that Am241 is a strong thermal neutron absorber it follows that, in both the PWR and RMPWR, UK Pu has a lower k_{∞} than RG Pu. In a thermal spectrum, the absorption cross-section of Am241 is greater than that of all

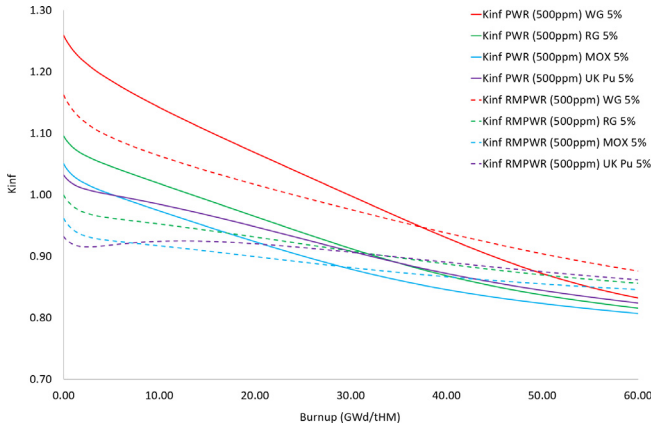


Fig. 1. Variation of k_{∞} with burnup for WG, RG, MOX and UK Pu at 5%wt Pu loading.

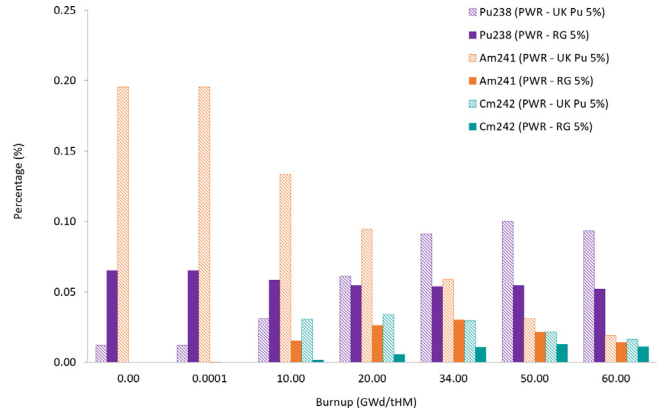


Fig. 4. Am241, Cm242 and Pu238 concentration between 0 and 60 GWd/tHM.

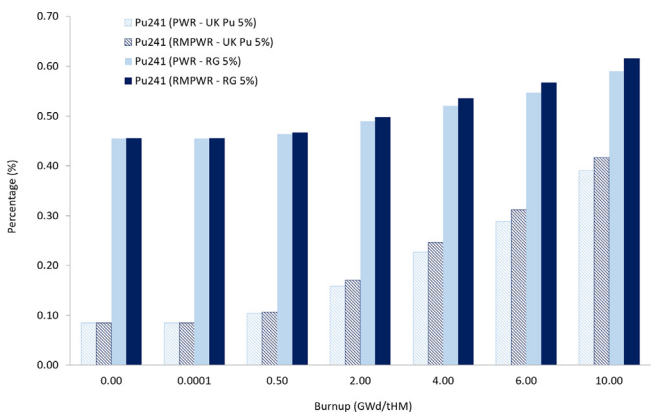


Fig. 2. Pu241 concentration in RG and UK Pu 5%wt between 0 and 10 GWd/tHM.

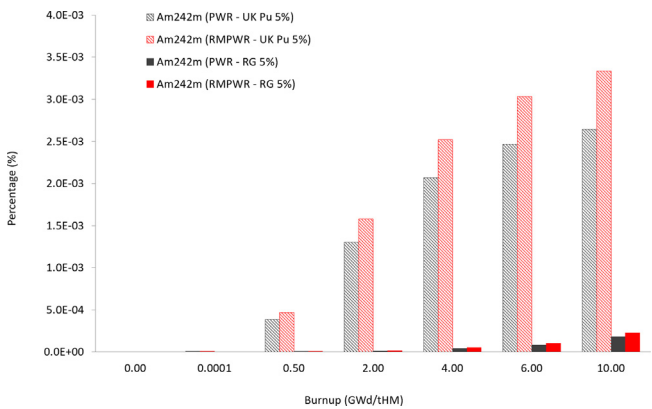


Fig. 3. Am242m concentration in RG and UK Pu 5%wt between 0 and 10 GWd/tHM.

concentration with burnup in UK Pu. Meanwhile, the Pu238 concentration in RG Pu remains relatively consistent, decreasing by only a small amount due to neutron capture. The effect is such that in UK Pu the decrease in neutron absorption in Am241 due to depletion is somewhat offset by the increase in neutron absorption by Pu238 causing the k_{∞} curve to remain lower in UK Pu than RG Pu. However, while Am241 will typically undergo neutron capture to form Am242, ~10% of captures will lead to the formation of Am242m (Fig. 5). Am242m has the largest thermal absorption cross-section of all nuclides within the fuel and so, despite accounting for only a small fraction of the total fuel composition, it can have a notable effect on reactivity. The concentration of Am242m is highest ~10 GWd/tHM (Fig. 6) with UK Pu containing greater levels of Am242m than RG Pu. Am242m production (and subsequent fission) offsets some of the reactivity penalty associated with neutron absorption in Am241 and Pu238 in UK Pu. The combined effect of depletion of Am241 and build-up of Pu238 and Am242m in UK Pu is such that by MOC the fuels display very similar k_{∞} values.

By EOC, Fig. 1 shows that k_{∞} in UK Pu is marginally higher than RG. This is caused by Am243 building up as the cycle progresses (Fig. 7). Am243 builds up due to neutron capture in Pu242 and, since fresh RG fuel contains twice as much Pu242 as UK Pu, the concentration of Am243 may be expected to be significantly higher in RG Pu. However, Am243 is also produced by successive neutron capture in Am241 which exists in significant quantities in UK Pu. Therefore, the Am243 concentration in both fuel types increases with burnup resulting in only slightly higher concentrations in RG Pu compared to UK Pu. Since MAs such as Am243 act as neutron absorbers in a thermal spectrum the result is that k_{∞} in RG Pu decreases such that the k_{∞} values for RG at EOC are lower than those of UK Pu. The difference in k_{∞} in RG and UK Pu at BOC is much more significant than the difference in k_{∞} at EOC since there

the fresh fuel constituents except Pu239. While Pu239 has a larger thermal absorption cross-section than Am241, Am241 has absorption peaks which exist at higher energies than the absorption peak of Pu239. This ultimately leads to preferential absorption in Am241 as neutrons undergo moderation; resulting in a significant impact on k_{∞} . Analysing the isotopic composition of the fuel throughout the cycle shows the Am241 concentration decreasing with burnup in UK Pu while small amounts of Am241 build-up around MOC in RG Pu (Fig. 4). As the Am241 concentration in UK Pu begins to decrease, the concentration of Cm242 increases. This, in turn, decays into Pu238 corresponding to an overall increase in Pu238

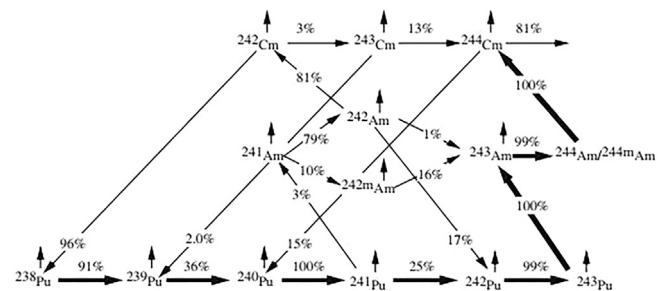


Fig. 5. Transmutation pathways in Pu and Am isotopes (Sasahara et al., 2004).

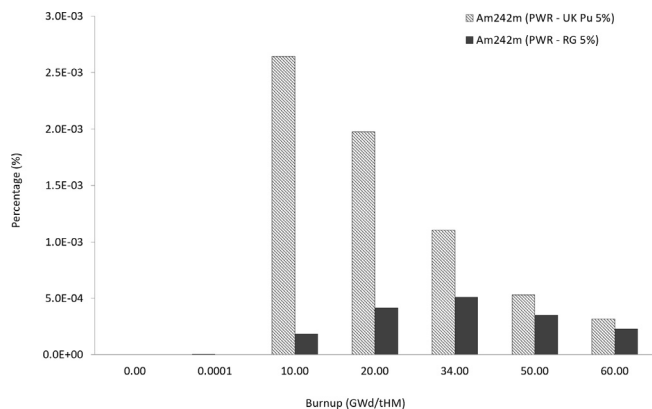


Fig. 6. Am242m concentration in RG and UK Pu 5%wt between 0 and 60 GWD/tHM.

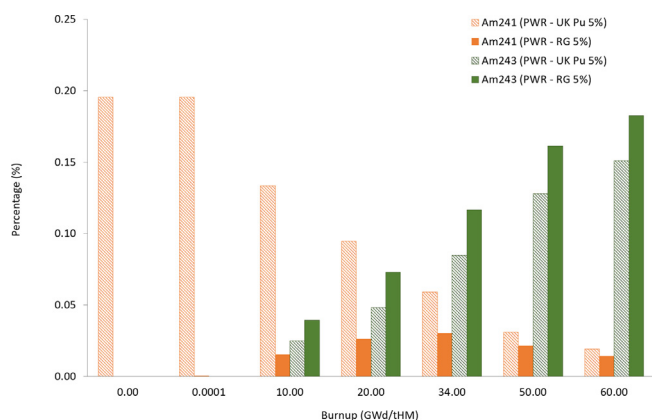


Fig. 7. Variations in Am241 and Am243 concentration in RG and UK Pu 5%wt between 0 and 60 GWD/tHM.

is significantly more Am241 in UK Pu than RG at BOC and only slightly more Am243 in RG than UK Pu at EOC.

Fig. 8 illustrates how the overall trend for k_{∞} becomes less severe as Pu content increases for all grades.

Before analysing the effect of isotopics on reactivity feedback coefficients it is useful to understand how perturbations, such as the increased fuel pin size in the RMPWR, alter the neutron energy spectrum. Fig. 9 illustrates how the spectrum changes in response to the increased fuel pin diameter and how the largest perturbation – a total loss of coolant – causes the spectrum to harden. The neutron flux is normalized per 1000 neutron productions. It is noted that the RMPWR has a lower normalized fast flux than the PWR under nominal conditions. In fact, the total (unnormalized) fast flux has increased in the RMPWR relative to the PWR but the total epithermal flux has increased by a greater amount, leading to a slight drop in the normalized fast flux. A higher overall flux is required in the RMPWR to achieve the same number of neutron productions as the PWR because fissile nuclides have a reduced fission cross in the harder spectrum of the RMPWR. At low Pu loadings, such as the 5%wt case shown, the effect of reduced moderation is such that the average neutron energy has shifted to the right. However, the fissile content is low enough that the new average neutron energy has not yet reached sufficiently high energies to correspond with the peak fission cross-sections of the key fissile nuclides.

In the fully voided case considered there is a significant increase in the high energy flux in both the PWR and RMPWR. However, the flux in the epithermal energy region has again increased by a proportionally larger amount resulting in the perceived increase in

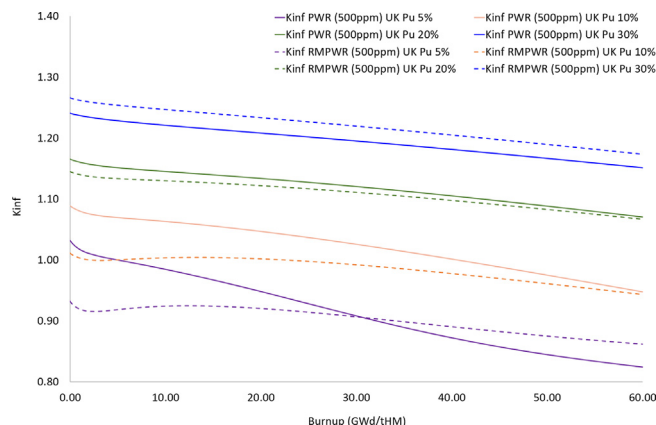


Fig. 8. Variation of k_{∞} with burnup for UK Pu at different Pu loadings.

intermediate flux/decrease in high energy flux. The increase in epithermal flux is due to the fact that fewer neutrons are absorbed before they reach intermediate energies. The increase in epithermal neutrons is more significant in the nominal vs perturbed conditions compared to the PWR vs RMPWR because the perturbation itself is more significant.

3.2. Doppler Coefficient

Results show that DC remains negative in all cases considered and, as expected, becomes less negative as more Pu is added. DC is less negative in the PWR than the RMPWR. This is due to the large absorption cross-section of Pu240 at 1 eV, which exceeds the absorption cross-sections for all other isotopes within the fuel and has a more significant effect on reactivity as the fuel temperature rises. There is increased absorption and lower reactivity as the spectrum hardens and the average neutron energy shifts towards the Pu240 absorption peak. DC typically becomes slightly more negative with burnup due to the depletion of fissile isotopes and the build-up of absorbing MAs and fission products. WG has the least negative DC, followed by UK Pu, RG and finally MOX grade Pu. This is the case for all loadings in both reactor types and is attributable to the Pu240 content of each of the Pu grades – MOX fuel containing the most and WG the least. Despite containing similar amounts of fissile and absorbing isotopes, UK Pu displays a more negative DC than RG at BOC and a less negative DC than RG at EOC. At BOC this is caused by the presence of Am241 – the strongest neutron absorber of those considered – in UK Pu (~4%wt compared to 0%wt in RG). At EOC this is caused by Am242m production in UK Pu, resulting in an overall more fissile fuel as the cycle progresses.

Fig. 10 shows how DC changes with burnup for all cases with 5% wt Pu content. This represents the most interesting case of those examined as it exhibits the greatest variation.

3.3. Boron Worth

BW displays slightly less predictable trends than DC. In all cases considered, BW is less negative in the RMPWR compared with the PWR. This is to be expected given the combined effect of the ratio of capture to absorption cross-sections of B10 monotonically decreasing with neutron energy and the reduced moderator volume (and hence boron load) in the RMPWR. Therefore, as the spectrum hardens boron becomes less effective as an absorber. Noticeable differences in BW occur when the Pu vector changes due to the effect of isotopic composition on the neutron energy spectrum. The effect is different between DC and BW as the con-

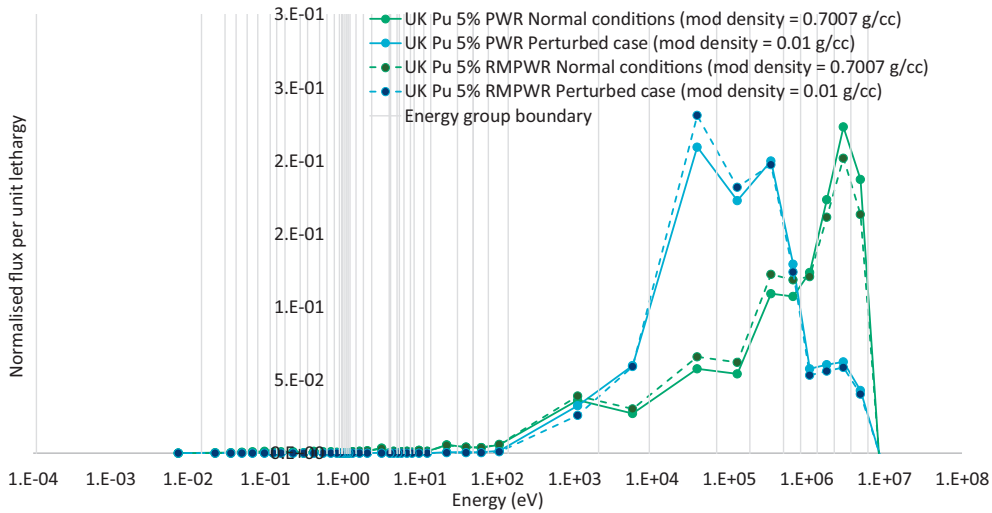


Fig. 9. Effect of varying H/HM ratio on the neutron energy spectrum of the PWR and RMPWR for 5%wt UK Pu.

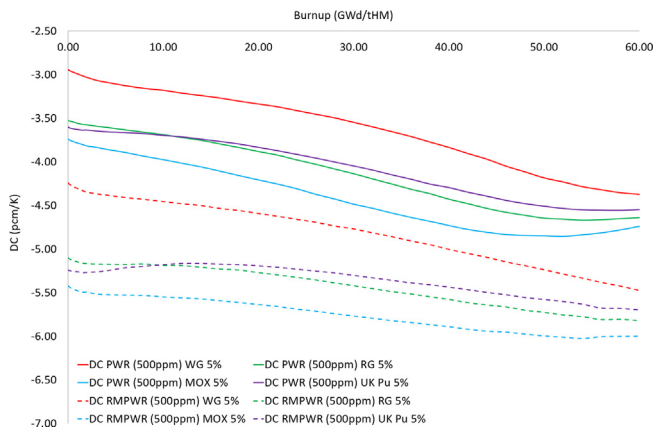


Fig. 10. Variation of DC with burnup for all grades with 5%wt Pu loading (PWR and RMPWR).

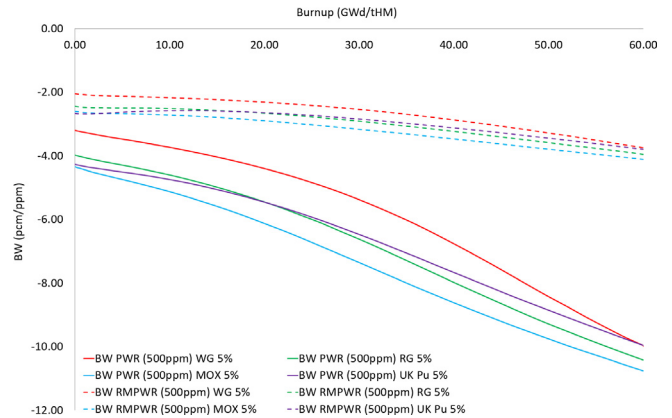


Fig. 11. Variation of BW with burnup for all grades with 5%wt Pu loading (PWR and RMPWR).

centration of boron in the moderator has a smaller effect than the isotopic contributions and their responses within the fuel itself.

At low Pu loadings, the RMPWR displays a slightly more negative BW with burnup which levels out as Pu content increases. In the PWR, however, there is a distinct decrease in BW with burnup for low Pu loading cases (Fig. 11) which levels out with increased fissile loading.

3.4. Moderator Temperature Coefficient

At low Pu loadings the RMPWR displays a more negative MTC than the PWR. For the 5%wt case, MTC becomes less negative with burnup for most Pu grades. For both reactor types, WG displays the least negative MTC across the entire cycle whereas UK Pu has the most negative MTC at BOC and MOX, the most negative MTC at EOC. As Pu content increases, the MTC becomes less negative overall and the PWR begins to display a more negative MTC than the RMPWR. The trend with burnup also changes from less negative to slightly more negative as the cycle progresses. In addition, the order in which the Pu grades are ranked from most to least negative MTC begins to change. At 30%wt Pu WG is the most negative in the RMPWR throughout the entire cycle and most negative in the PWR from MOC onwards. Fig. 12 shows the difference in trend

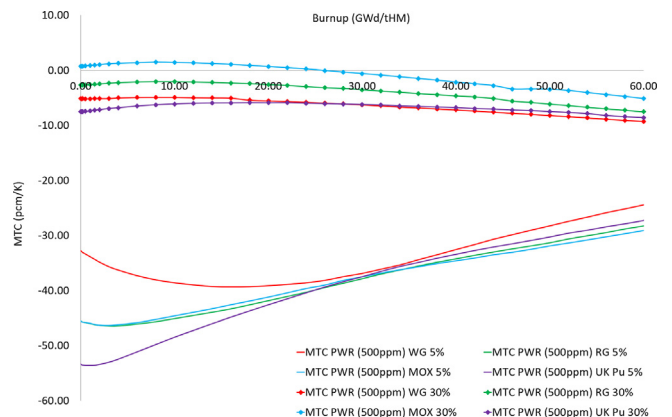


Fig. 12. Variation of MTC with burnup for all grades with 5 and 30%wt Pu loading (PWR only).

with burnup for high and low Pu loadings in the PWR and the variation in MTC by grade. Fig. 13 shows how the MTC in the PWR and RMPWR differs for a given grade at high and low Pu loadings. This illustrates that at low Pu loadings the MTC is most negative in the RMPWR but, as the Pu content increases, the PWR begins to display

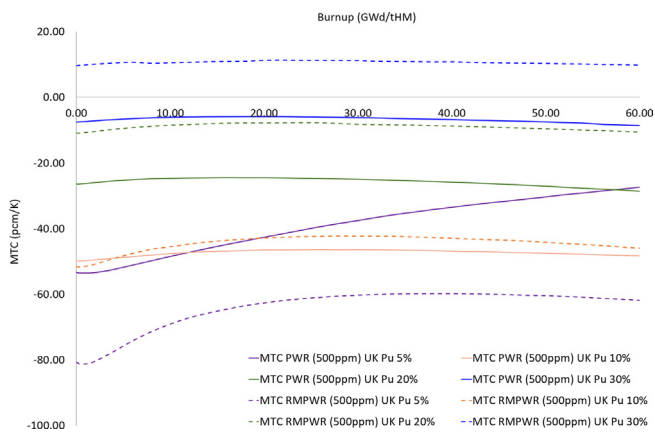


Fig. 13. Variations in MTC with burnup by Pu content for UK Pu.

a more negative MTC. It is worth noting that 10%wt Pu is the point at which the trend switches from one reactor type to the other.

The effect of spectral hardening is clearly more complex in this case than for DC and BW and therefore requires a more detailed analysis.

3.5. Fully Voided Reactivity

FVR might be expected to display similar trends to MTC, given that both result in a reduction in density of the light water moderator/coolant, but this is shown not to be the case. The perturbation for FVR is far more extreme than the perturbation for MTC and the effect is such that the spectrum in the perturbed FVR case is very different to – and much harder than – the spectrum in the perturbed MTC case. The result is that, for all cases, FVR becomes more negative with burnup, including at low Pu loadings. In addition, the order in which the Pu grades are ranked from most to least negative is different for FVR compared to MTC. The order is also inconsistent across the cases considered, as it depends heavily on Pu content. At 5%wt Pu, WG goes from least to most negative FVR during the cycle in both reactor types, while UK Pu goes from most to least negative FVR. At 10%wt Pu, WG has the least negative FVR and MOX grade Pu has the most negative FVR for both reactor types. At 20%wt, the FVR for most grades is positive, with UK Pu displaying the most positive FVR and MOX grade the least by EOC in both cases. At 30%wt loading, UK Pu has the most positive FVR while WG has the least in both cases. The difference in FVR for high and low Pu loadings in the PWR for each grade is shown in Fig. 14.

As with MTC, FVR displays a complex response to coolant density perturbations, which have been shown to be dependent on the isotopic composition of Pu and the overall Pu content in the fuel. The second part of this study considers the contribution by isotope and energy bin to MTC and FVR, to better understand the system and allow operators to predict likely responses to isotopic variations in Pu vector. It should be noted that these results do not consider the effects of burnable absorbers, which will make MTC and FVR less favourable. This will be considered in future work.

3.6. MTC by isotope

When considered by energy group and individual isotope, the fissile isotopes are shown to be dominant contributors to MTC and FVR, as expected. At BOC, Pu239 is the dominant isotope while all others play a minor role (Fig. 15). For all Pu grades considered, Pu239 has a negative contribution at thermal energies and a positive contribution at epithermal energies and above as shown by (Zainuddin et al., 2016). At lower Pu loadings, Pu239 provides a lar-

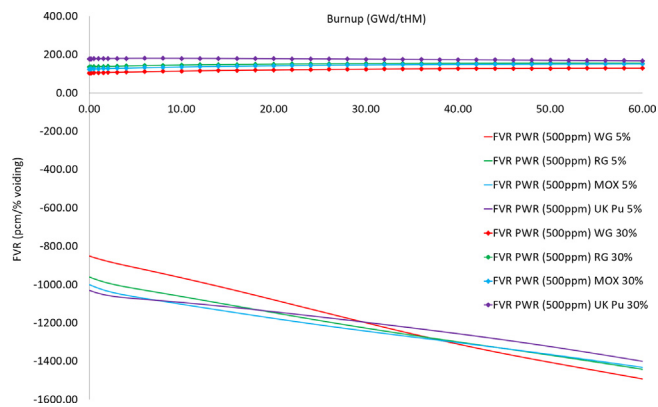


Fig. 14. Variation of FVR with burnup for all grades with 5 and 30%wt Pu loading (PWR only).

ger negative contribution in the thermal energy region in the RMPWR, while at higher loadings, there is a larger negative contribution in the PWR. As the Pu content increases, the negative contributions in the thermal region diminish and the positive contributions in the epithermal region increase causing the overall MTC to become positive at high Pu loadings.

A key feature in these isotopic contributions is a large positive contribution – present for all grades of Pu at low percentage loadings – which exists between 0.1 and 1 eV. This corresponds to the energy of the largest absorption cross-section of Pu239 and strongly influences the overall MTC. The positive peak diminishes as the Pu content of the fuel increases (Fig. 16). However, in a limited number of cases, it means that increasing the Pu content can result in a more negative MTC, as the reduction in the positive peak has a more significant effect in determining the overall MTC than the combined effects of increased positive epithermal contributions and reduced negative thermal contributions. For example, MTC at BOC in a UK Pu fueled PWR (Fig. 16) is more negative for 6%wt and 7.5%wt Pu than for 5%wt. However, for >7.5%wt the MTC becomes less negative with increased Pu loading. The effect of the positive peak may therefore be useful in terms of maximising Pu loading in the core while reducing the MTC. It should be noted that while the MTC is more negative at 6–7.5%wt Pu compared with 5%wt Pu in the example case, the FVR is significantly less negative for 6–7.5%wt Pu than for 5%wt Pu. The contributions from different isotopes to FVR should therefore be kept in mind if attempting to utilise this feature to maximise Pu content in the fuel. The largest contributions from the positive peaks between 0.1 and 1 eV exist at lower Pu loadings in the RMPWR than the PWR, because the spectrum is already harder in the RMPWR and, therefore, lower Pu loadings are required to shift the average neutron energy towards the key absorption cross-sections. However, these peaks may still be used to achieve the same reduction in MTC with increased Pu content in a limited number of cases. The reduction in magnitude of the positive contribution between 0.1 and 1 eV with increased Pu loading can be explained by the fact that the neutron energy spectrum hardens as the fissile content increases causing a shift in the average neutron energy to the right past the large Pu239 absorption peak between 0.1 and 1 eV and towards the large absorption peak of Pu240 at ~1 eV.

At EOC Pu239 is only dominant at very high Pu loadings (~20% wt and above). At lower loadings, Pu239 is almost completely depleted by EOC and therefore has a much less significant effect. Where this happens, other fissile isotopes become dominant. At 5%wt Pu loading U233 is typically the most dominant isotope at EOC displaying somewhat similar trends to Pu239 (Fig. 17).

U233 shows a similar trend to Pu239 in terms of the reduction in magnitude of the positive peaks between 0.1 and 1 eV as Pu con-

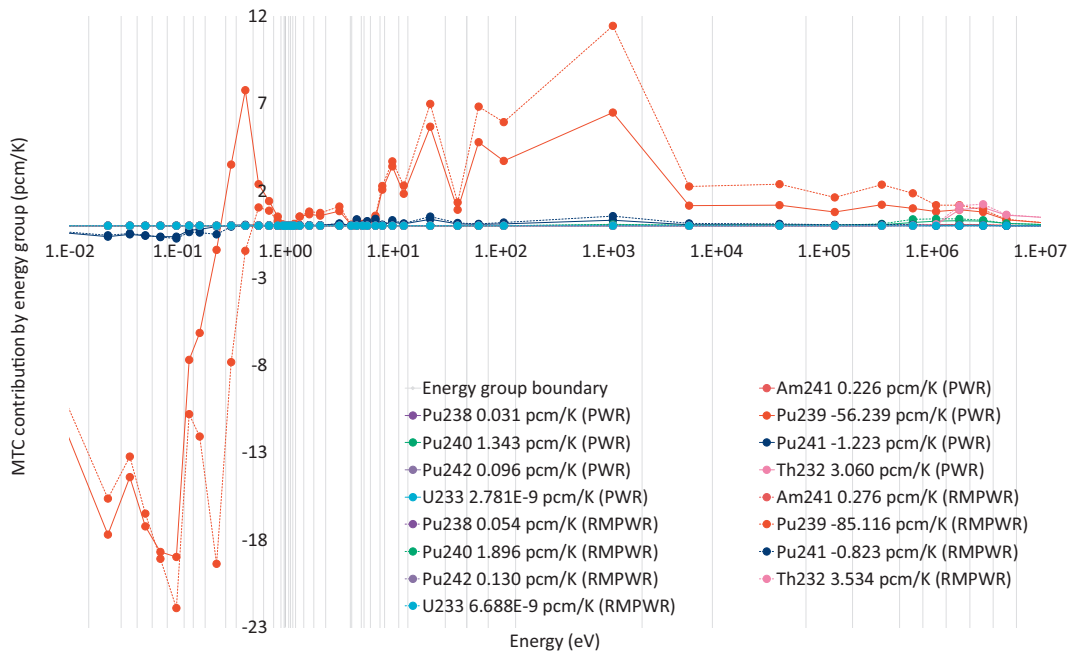


Fig. 15. MTC contributions by isotope for 5wt UK Pu in the PWR (solid lines) and the RMPWR (dashed lines) at 0 GWd/tHM. The net area represents the overall MTC in this and all similar figures.

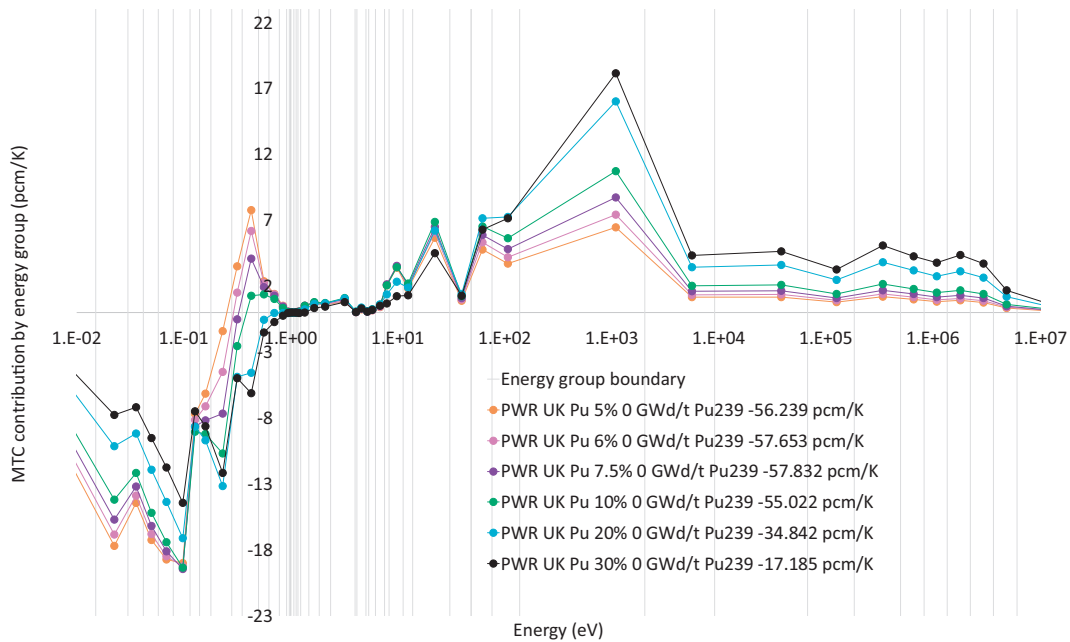


Fig. 16. MTC contributions from Pu239 for different Pu loadings of UK Pu in the PWR at 0 GWd/tHM.

tent increases. However, the total magnitude of the contribution from U233 decreases in general as the Pu content increases (Fig. 18). This is caused by competition between Pu239 and U233 in higher Pu content cases.

At 10%wt Pu loading, Pu241 is more dominant than U233 in the thermal energy region, since there is sufficient initial fissile material to warrant the Pu isotopes being of key importance at EOC. Pu239 depletes preferentially to Pu241 due to the larger peak absorption cross-section between 0.1 and 1 eV. As a result, Pu239 burns up such that by EOC in the PWR this isotope does not exist in quantities capable of having such a significant effect on MTC, resulting in Pu241 becoming more dominant at 10%wt Pu and

above. In the RMPWR, the harder spectrum causes the average neutron energy to shift further to the right which increases the likelihood of fission in other isotopes and ultimately causes fewer neutrons to be available for fission in Pu239 specifically. The result is that there is sufficient Pu239 to continue having a significant effect at EOC. Therefore, the remaining Pu241, which has a larger fission cross-section than U233 in the thermal energy region, is of greater significance (Fig. 19).

In terms of trend, Pu241 shows similar contributions at EOC (Fig. 20) to those of Pu239 at BOC (Fig. 16), albeit on a smaller scale. The main difference to note with regard to the contributions from Pu241 is that the magnitude of the negative contributions <1 eV

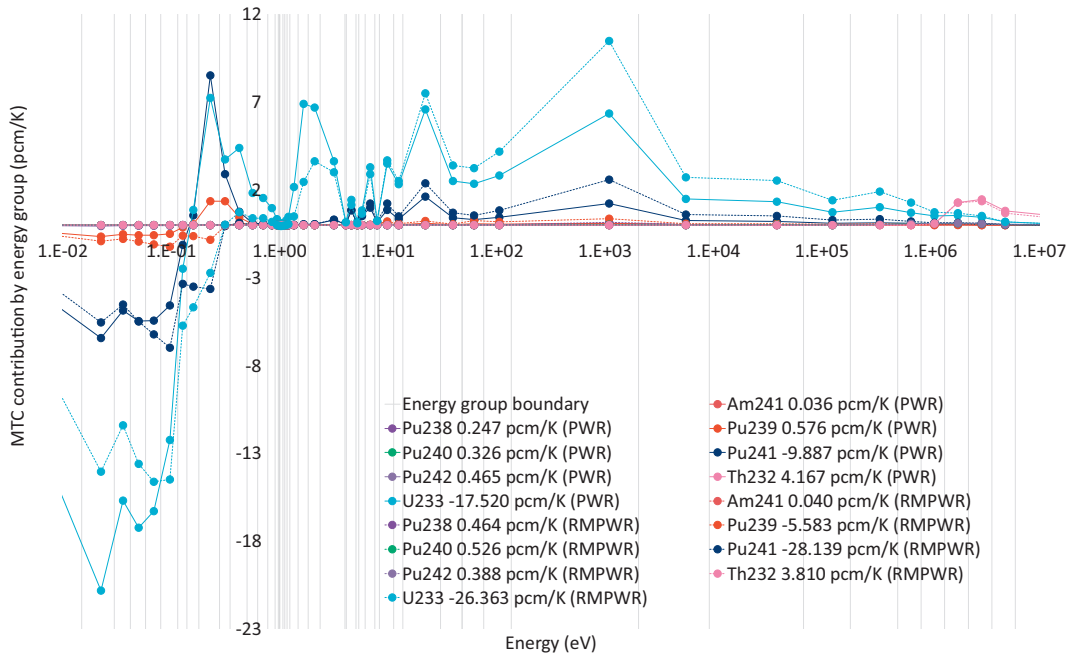


Fig. 17. MTC contributions by isotope for 5wt UK Pu in the PWR (solid lines) and the RMPWR (dashed lines) at 60 GWd/tHM.

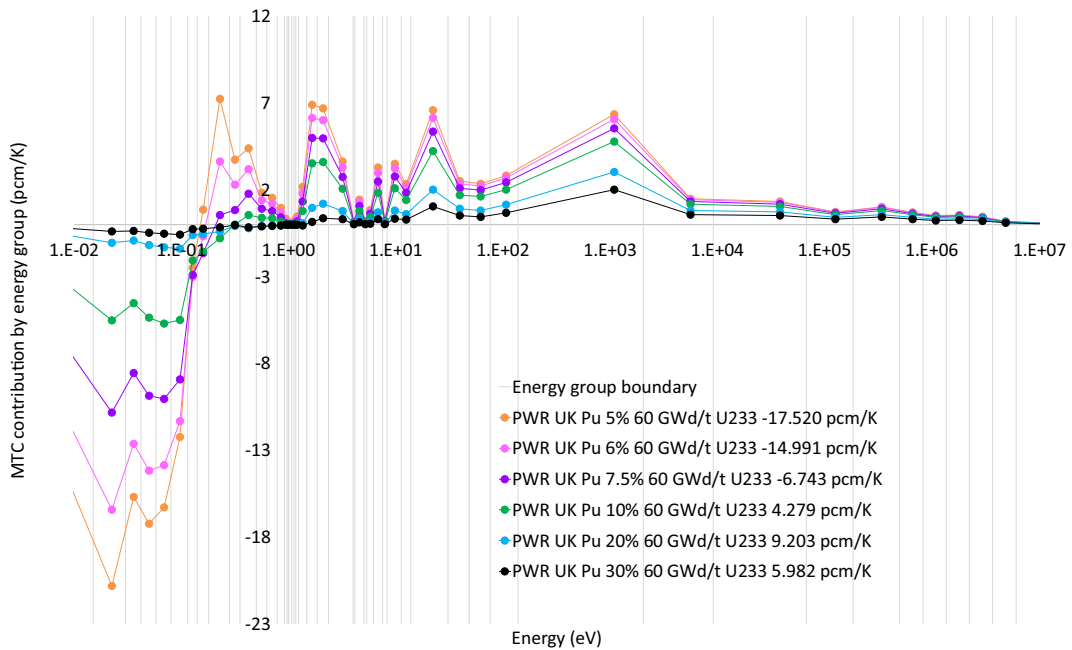


Fig. 18. MTC contributions from U233 for different Pu loadings of UK Pu in the PWR at 60 GWd/tHM.

and the positive contributions >1 eV are much less extreme. The impact of Pu241 is therefore less significant than for other isotopes.

The RMPWR displays the same trend, in terms of contributions at specific energies, as the PWR. However, as stated, a lower Pu loading is required to achieve positive peaks of a higher magnitude at 0.1–1 eV (Fig. 21), e.g. 2.5wt Pu in the RMPWR is comparable with 5wt Pu in the PWR.

The positive peaks that the fissile isotopes display between 0.1 and 1 eV can be held accountable for the change in trend in which reactor type displays the most negative MTC. At 10wt Pu loading, which represents the transition point, the positive peaks are almost completely non-existent regardless of reactor type. The lar-

ger positive contributions in the epithermal region of the RMPWR become dominant causing the RMPWR to display a less negative MTC than the PWR. Below 10wt Pu loading, the larger positive peaks in the PWR result in a less negative MTC in the PWR than the RMPWR.

3.7. FVR by isotope

FVR shows similar results to MTC in that there are negative contributions in the thermal energy region and positive contributions in the epithermal energy region and above. However, positive contributions towards FVR do not exist at energies <500 eV, unlike

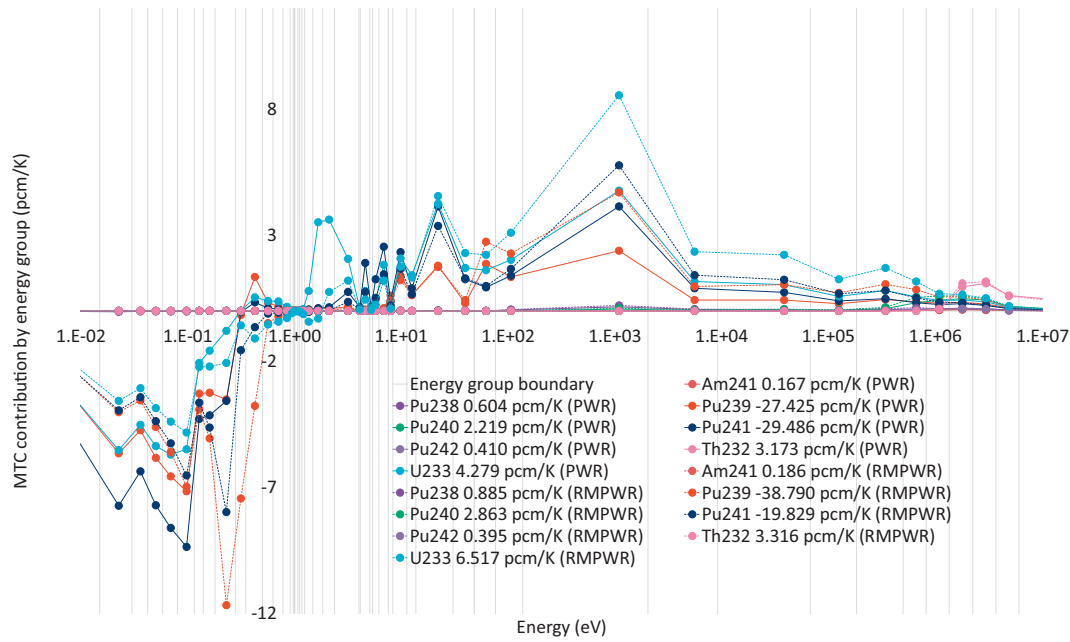


Fig. 19. MTC contributions by isotope for 10%wt UK Pu in the PWR (solid lines) and the RMPWR (dashed lines) at 60 GWd/tHM.

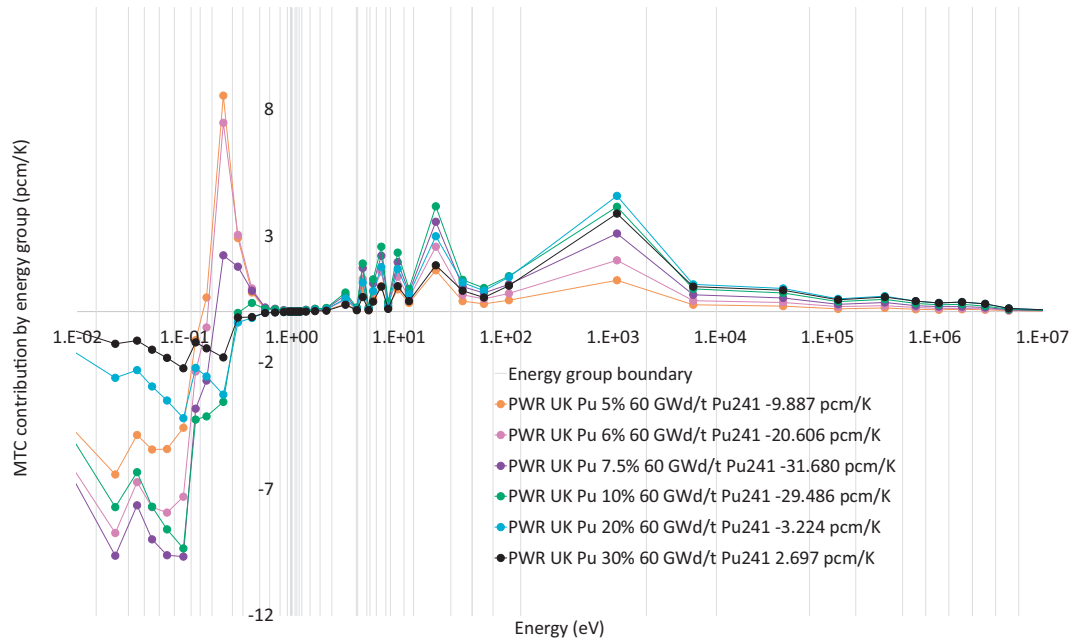


Fig. 20. MTC contributions from Pu241 for different Pu loadings of UK Pu in the PWR at 60 GWd/tHM.

with MTC. Pu239 remains the dominant isotope, assuming it exists in sufficient quantities to have an effect. Like MTC, Pu241 has a significant effect on reactivity in Pu grades which contain large amounts of this isotope (i.e. RG and MOX). In terms of overall trend, the contribution from Pu241 is very similar to that of Pu239 (Fig. 22) although the magnitude of the contributions is notably different. At EOC Pu239 is only dominant in cases where the initial fissile loading is high. If Pu239 depletes, U233 takes over as the dominant isotope. Again, U233 has a larger positive contribution in the epithermal region; however, Pu241 has a larger negative contribution in the thermal region (Fig. 23).

As the total fissile loading increases, the negative contribution from Pu239 to MTC decreases and the positive contribution increases. However, for FVR both the negative and positive contributions decrease as fissile loading increases since voiding itself causes a significant reduction in H/HM ratio, which leads to a reduced resonance escape probability as the spectrum hardens. This effect is more significant in cases with a high Pu content as the spectrum is already harder than in lower Pu content cases. FVR becomes positive as fissile content increases due to a reduction in the large negative trough between 0.1 and 1 eV (Fig. 24) (recall that at this energy there is a positive peak in the MTC cases).

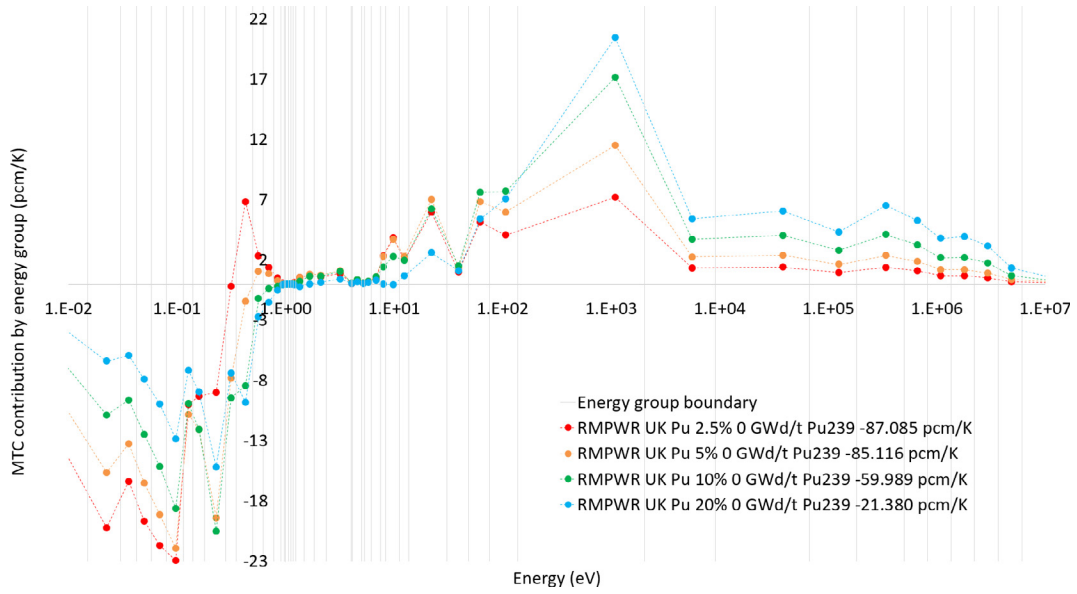


Fig. 21. MTC contributions from Pu239 for different Pu loadings of UK Pu in the RMPWR at 0 GWd/tHM.

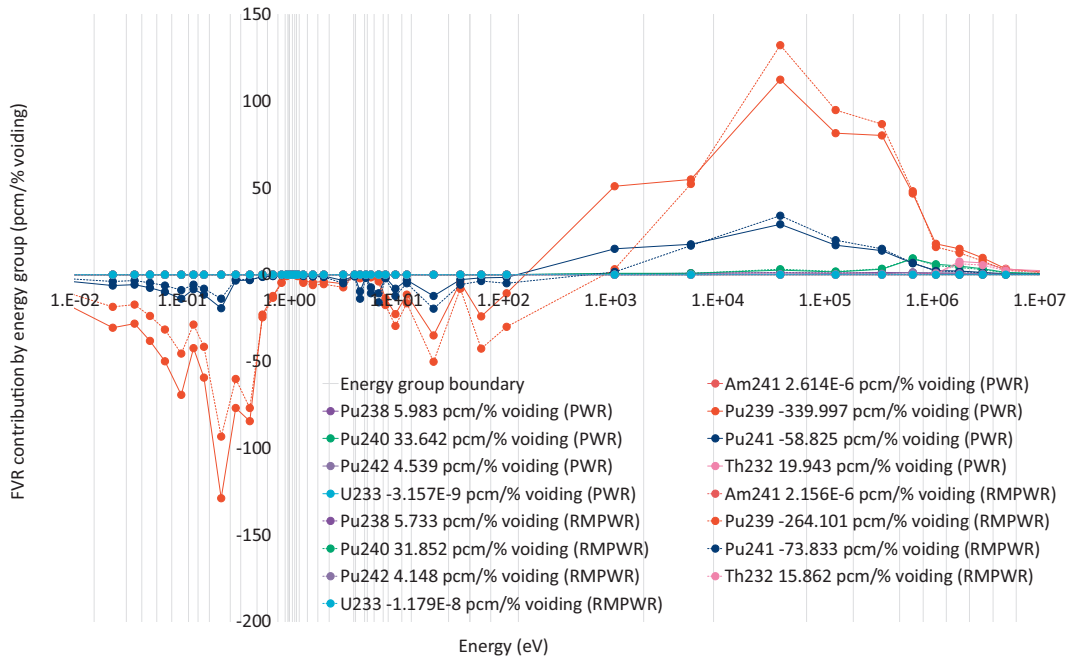


Fig. 22. FVR contributions by isotope for 10%wt RG Pu in the PWR (solid lines) and the RMPWR (dashed lines) at 0 GWd/tHM.

3.8. Practical limitations

Practical limitations such as reactivity limited discharge burnup and Pu destruction rates must be considered in addition to understanding the response of different isotopes to variations in fuel composition and moderation. Initial results (Fig. 1) indicate that $k_{\infty} < 1.03$ by EOC in a number of cases and $k_{\infty} > 1.03$ in others. Tables 4–7 show the reactivity limited discharge burnup, MTC, and FVR for each grade of Pu using different percentage loadings. Neither MTC nor FVR vary significantly between the Pu grades in either reactor type. However, the discharge burnup is considerably different for each of the Pu vectors studied.

These results show that low Pu loadings (typically <10%wt for non-WG Pu) correspond to uneconomically low discharge burnups

and short cycle lengths. This is more noticeable in the RMPWR as per (Shwageraus et al., 2004). For higher Pu loadings, a greater cycle length may be theoretically achievable; however, in most cases the discharge burnup exceeds current cladding technology limits. MTC remains negative for all cases except RG, MOX and UK Pu at 30%wt Pu loading in the RMPWR and does not fall below -60 pcm/K, which is within the typically accepted lower limit for MTC in PWRs. FVR is positive for >30%wt Pu for all grades in the PWR and >20%wt Pu for all grades in the RMPWR. If a negative MTC is required, the maximum Pu loading, and therefore discharge burnup, is higher in the PWR than the RMPWR. Despite the difference in Pu loading, roughly the same amount of Pu is consumed when MOX fuel is loaded into the PWR and RMPWR; however, other grades display noticeable differences (Table 8). In the PWR

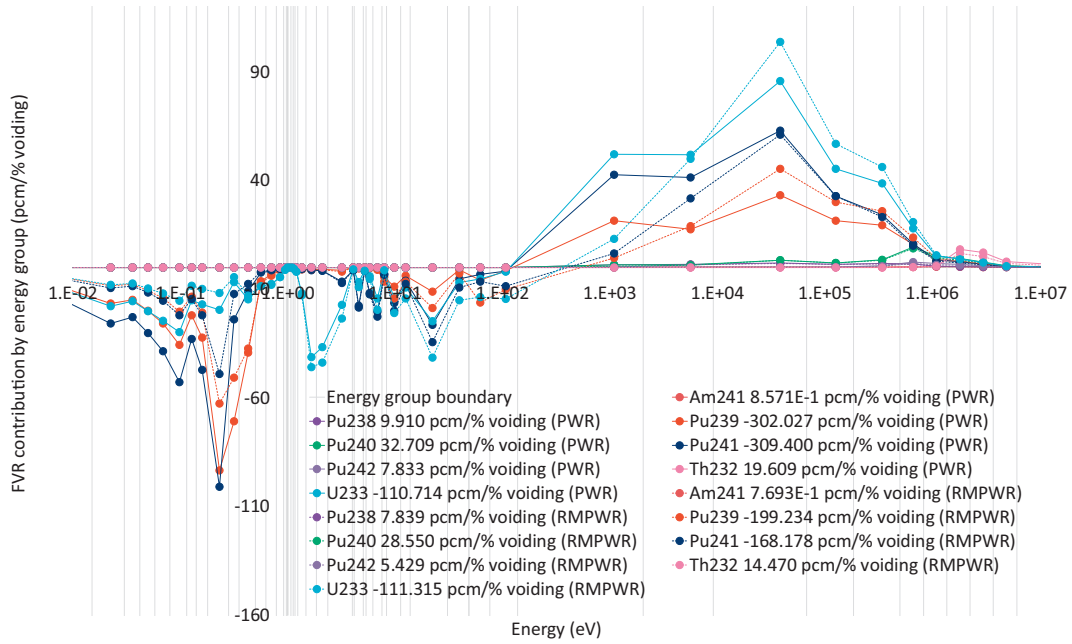


Fig. 23. FVR contributions by isotope for 10%wt RG Pu in the PWR (solid lines) and the RMPWR (dashed lines) at 60 GWd/tHM.

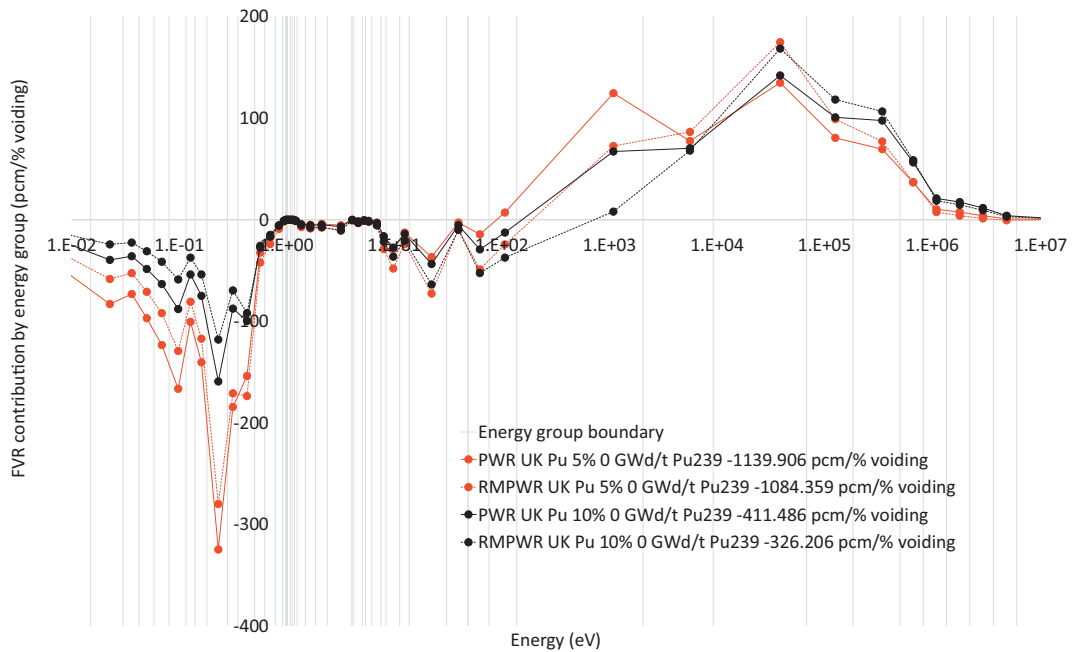


Fig. 24. FVR contributions from Pu239 for different Pu loadings of UK Pu in the PWR (solid lines) and the RMPWR (dashed lines) at 0 GWd/tHM.

Table 4
Reactivity-limited 3-batch discharge burnup (GWd/tHM) for WG Pu.

Pu (%wt)	Point in cycle	Burnup (GWd/tHM)		MTC (pcm/K)		FVR (pcm/% voiding)	
		PWR	RMPWR	PWR	RMPWR	PWR	RMPWR
5	BOC	0.00	0.00	-38.70	-58.68	-1070.49	-867.39
	EOC	38.22	25.49	-36.93	-58.85	-1144.48	-886.81
10	BOC	0.00	0.00	-43.51	-43.46	-487.60	-312.67
	EOC	95.15	85.03	-45.31	-46.93	-609.03	-359.34
20	BOC	0.00	0.00	-34.81	-20.02	-71.30	18.94
	EOC	194.29	191.76	-43.67	-27.41	-183.01	-24.03
30	BOC	0.00	0.00	-22.93	-3.59	96.08	141.16
	EOC	286.71	293.41	-36.15	-13.06	11.05	112.09

Table 5
Reactivity-limited 3-batch discharge burnup (GWd/tHM) for RG Pu.

Pu (%wt)	Point in cycle	Burnup (GWd/tHM)		MTC (pcm/K)		FVR (pcm/% voiding)	
		PWR	RMPWR	PWR	RMPWR	PWR	RMPWR
5	BOC	0.00	-	-46.07	-	-1028.13	-
	EOC	11.70	-	-45.58	-	-1044.33	-
10	BOC	0.00	0.00	-44.65	-40.22	-409.15	-272.34
	EOC	52.94	30.99	-46.16	-40.78	-454.09	-282.11
20	BOC	0.00	0.00	-29.12	-11.24	-10.14	47.18
	EOC	125.04	123.52	-34.48	-14.88	-47.35	33.47
30	BOC	0.00	0.00	-13.92	5.94	143.37	163.19
	EOC	194.01	208.33	-20.84	1.54	122.41	158.90

Table 6
Reactivity-limited 3-batch discharge burnup (GWd/tHM) for MOX Pu.

Pu (%wt)	Point in cycle	Burnup (GWd/tHM)		MTC (pcm/K)		FVR (pcm/% voiding)	
		PWR	RMPWR	PWR	RMPWR	PWR	RMPWR
5	BOC	0.00	–	–45.86	–	–1016.56	–
	EOC	1.57	–	–45.96	–	–1020.96	–
10	BOC	0.00	0.00	–42.43	–36.94	–392.41	–288.92
	EOC	33.01	10.31	–43.74	–37.00	–416.57	–290.65
20	BOC	0.00	0.00	–24.40	–6.46	–0.18	41.03
	EOC	91.65	91.37	–28.71	–9.31	–19.84	36.06
30	BOC	0.00	0.00	–8.02	10.19	149.38	159.08
	EOC	147.33	163.56	–13.28	7.34	143.10	162.97

Table 7
Reactivity-limited 3-batch discharge burnup (GWd/tHM) for UK Pu.

Pu (%wt)	Point in cycle	Burnup (GWd/tHM)		MTC (pcm/K)		FVR (pcm/% voiding)	
		PWR	RMPWR	PWR	RMPWR	PWR	RMPWR
5	BOC	0.00	–	–53.45	–	–1030.69	–
	EOC	0.14	–	–53.47	–	–1031.10	–
10	BOC	0.00	–	–46.72	–	–386.23	–
	EOC	41.98	–	–46.81	–	–414.03	–
20	BOC	0.00	0.00	–29.03	–10.98	3.05	58.08
	EOC	121.57	121.87	–33.08	–13.89	–30.46	43.56
30	BOC	0.00	0.00	–13.59	6.22	152.50	170.41
	EOC	196.15	213.90	–19.65	2.02	130.57	163.57

~30 kg/GW_y more WG is destroyed than in the RMPWR whereas for RG and UK Pu this figure changes to ~10 kg/GW_y. The U233 content in SNF is similar for all grades except WG which results in ~10 kg/tHM more U233 in RMPWR SNF than PWR SNF. However, the total MA content is ~10 kg/tHM lower in the RMPWR for all grades except WG which is ~10 kg/tHM higher in the PWR. Where a negative FVR is required, the maximum Pu loading is ~10% lower in the PWR than when the requirement to maintain a negative MTC is the limiting factor. The maximum Pu loading is the same in the RMPWR regardless of whether MTC or FVR are taken as the limiting case (Table 9). Reducing the Pu content

Table 8
Practical limitations where MTC is required to remain negative throughout the cycle.

Constraint	PWR				RMPWR			
	WG	RG	MOX	UK Pu	WG	RG	MOX	UK Pu
Maximum Pu limit (%wt)	30	30	30	30	30	20	20	20
Reactivity limited discharge burnup (GWd/tHM)	286.71	194.01	147.33	196.15	293.41	123.52	91.37	121.87
Pu consumed (kg/GW _y)	274.45	273.45	279.72	255.57	245.19	264.33	279.15	246.71
U233 accumulated by EOC (kg/tHM)	33.52	31.15	27.40	31.03	43.88	31.88	27.23	31.51
MAs accumulated by EOC (kg/tHM)	61.84	63.04	59.19	59.95	74.83	51.89	46.14	49.92

Table 9
Practical limitations where FVR is required to remain negative throughout the cycle.

Constraint	PWR				RMPWR			
	WG	RG	MOX	UK Pu	WG	RG	MOX	UK Pu
Maximum Pu limit (%wt)	20	20	20	20	20	20	20	20
Reactivity limited discharge burnup (GWd/tHM)	194.29	125.04	91.65	121.57	191.76	123.52	91.37	121.87
Pu consumed (kg/GW _y)	278.99	292.45	304.48	275.80	249.67	264.33	279.15	246.71
U233 accumulated by EOC (kg/tHM)	27.95	25.16	21.41	24.79	36.67	31.88	27.23	31.51
MAs accumulated by EOC (kg/tHM)	46.00	44.23	39.45	37.57	56.39	51.89	46.14	49.92

results in a significantly lower discharge burnup (although all burnup values still exceed current cladding limits). When the maximum Pu content is 20%wt for all Pu grades in both the PWR and the RMPWR, a higher destruction rate is possible in the PWR. It is worth noting that when the FVR limits the Pu loadings to 20% wt (as opposed to 30%wt in the MTC limited case) the Pu destruction in the PWR is higher for all Pu grades despite the lower Pu loading. This is due to the higher levels of U233 in the MTC limited case and subsequent competition between Pu239 and U233 as well as increased absorption due to higher levels of MAs. In the FVR limited case the U233 and total MA content in SNF is 5–10 kg/tHM higher in the RMPWR than the PWR for all Pu grades.

4. Conclusions

Results illustrate how sensitive operational and safety parameters are to changes in the isotopic composition of the fuel and the effects of spectral hardening. The k_{∞} curves were shown to be heavily influenced by the presence of Am241 in UK Pu despite UK Pu having a similar Pu composition to RG Pu.

DC was determined to be strongly dependent on the Pu240 content of the fuel due to the absorption cross-section of this isotope at 1 eV and its subsequent negative effect on reactivity. BW was shown to be reliant upon spectrum-related effects due to the monotonic decrease in the ratio of capture to absorption cross-sections of B10 with increasing neutron energy. MTC and FVR were both shown to be more complex and difficult to predict. Pu239 was found to have the most significant effect on both MTC and FVR throughout the cycle assuming sufficient quantities remain at EOC. Where Pu239 has depleted by EOC, U233 becomes the most dominant isotope at low Pu loadings and Pu241 the most dominant isotope at higher Pu loadings. Reductions in positive contribution peaks from the fissile isotopes in the energy range 0.1–1 eV were shown to offset the increased fissioning in the epithermal energy region at higher Pu loadings and may be used to make MTC marginally more negative in a limited number of cases. FVR was shown to display some similar trends to MTC. However, key differences were noted – particularly the absence of positive contributions between 0.1 and 1 eV. This was found to be caused by the fact that the spectrum in the FVR case is much harder than the spectrum in the MTC case, due to the more extreme perturbation to the moderator density.

Where a negative MTC is required to meet safety criteria, it was found that the PWR can tolerate a higher maximum fissile loading and can therefore achieve a higher discharge burnup and Pu destruction rate than the RMPWR in a once-through cycle. Where a negative FVR is required, the maximum fissile loading and discharge burnup are similar in both reactor types, while increased Pu consumption rates were possible in the PWR compared to the RMPWR due to lower levels of U233 and MAs in the SNF, thereby reducing both competition between Pu239 and U233 and neutron absorption in MAs.

5. Data availability statement

To the best of the authors' knowledge, this paper and references herein contain all the data needed to reproduce and validate the results presented.

Acknowledgements

The first author is grateful for the financial support of the Engineering and Physical Sciences Research Council (EPSRC), Thor Energy, the Whitworth Society and the Institution of Engineering and Technology. Thanks are also due to Dr Cheuk Wah Lau (Thor Energy) for his advice, guidance and support.

References

- Alhaj, M.Y., Badawi, A., Abou-Gabel, H.H., Mohamed, N.M.A., 2016. Partial loading of thorium-plutonium fuel in a pressurized water reactor. *Nucl. Technol.* 194 (3), 314–323.
- Ashley, S.F. et al., 2014. Fuel cycle modelling of open cycle thorium-fuelled nuclear energy systems. *Ann. Nucl. Energy* 69, 314–330.
- Askew, J.R., Fayers, F.J., Kemshall, P.B., 1966. A general description of the lattice code WIMS. *J. Br. Nucl. Energy Society*.
- Chadwick, M.B. et al., 2006. ENDF/B-VII.0 next generation evaluated nuclear data library for nuclear science and technology. *Nucl. Data Sheets* 107, 2931–3060.
- Department of Energy & Climate Change, 2011. Management of the UK's plutonium stocks: A consultation response on the long-term management of UK-owned separated civil plutonium. S.l.: DECC.
- Driscoll, M.J., Downar, T.J., Pilat, E.E., 1991. *The Linear Reactivity Model for Nuclear Fuel Management*. American Nuclear Society, Illinois.
- Ernout, M. et al., 2015. Advanced plutonium management in PWR, complementarity of thorium and uranium. *Prog. Nucl. Energy* 78, 330–340.
- Forget, B., Xu, S., Smith, K., 2014. Direct doppler broadening in Monte Carlo simulations using the multipole representation. *Ann. Nucl. Energy* 64, 78–85.
- Fridman, E., Kliem, S., 2011. Pu recycling in a full Th-MOX PWR core. Part I: steady state analysis. *Nucl. Eng. Des.* 241 (1), 193–202.
- Galperin, A., 1995. Utilization of light water reactors for plutonium incineration. *Ann. Nucl. Energy* 22 (8), 507–511.
- Galperin, A., Raizes, G., 1997. A pressurized water reactor design for plutonium incineration: fuel cycle options. *Nucl. Technol.* 117 (2), 125–132.
- Ganda, F., Greenspan, E., 2010. Analysis of reactivity coefficients of hydride-fueled PWR cores. *Nucl. Sci. Eng.* 164, 1–32.
- Gill, M., 2016. *The Potential Impact of Fast Reactors and Fuel Recycling Schemes on the UK's Nuclear Waste Inventory*. University of Manchester, Manchester. PhD Thesis.
- Haas, D., Hamilton, D.J., 2007. Fuel cycle strategies and plutonium management in Europe. *Prog. Nucl. Energy* 49, 574–582.
- Hesketh, K., Thomas, M., 2013. *The Potential Role of the Thorium Fuel Cycle in Reducing the Radiotoxicity of Long-Lived Waste – 13477*. Waste Management Symposium, Phoenix.
- Institution of Mechanical Engineers, 2013. *UK Plutonium: The Way Forward*. Institution of Mechanical Engineers, Westminster.
- Insulander Bjork, K., 2013. A BWR fuel assembly design for efficient use of plutonium in thorium-plutonium fuel. *Prog. Nucl. Energy* 65, 56–63.
- Insulander Bjork, K., Kekkonen, L., 2015. Thermal-mechanical performance modeling of thorium-plutonium oxide fuel and comparison with online irradiation data. *J. Nucl. Mater.* 467, 876–885.
- International Atomic Energy Agency, 2002. *Thorium Fuel Utilization: Options and Trends*. International Atomic Energy Agency, Vienna.
- International Atomic Energy Agency, 2003a. *Potential of Thorium Based Fuel Cycles to Constrain Plutonium and reduce long lived waste toxicity (IAEA-TECDOC-1349)*. IAEA, Vienna.
- International Atomic Energy Agency, 2003b. *Status and Advances in MOX Fuel Technology*. International Atomic Energy Agency, Vienna.
- International Atomic Energy Agency, 2005. *Thorium Fuel Cycle – Potential Benefits and Challenges (TECDOC 1450)*. IAEA, Vienna.
- Kamei, T., Hakami, S., 2011. Evaluation of implementation of thorium fuel cycle with LWR and MSR. *Prog. Nucl. Energy*, 820–824.
- Lamarsh, J.R., Baratta, A.J., 2001. *Introduction to Nuclear Engineering*. Prentice Hall, s.l..
- Lau, C.W., Nylan, H., Bjork, K.I., Sandberg, U., 2014. Feasibility Study of 1/3 thorium-plutonium mixed oxide core. *Sci. Technol. Nucl. Installations*.
- Lindley, B.A., 2014. *The Use of Reduced-Moderation in Light Water Reactors for Transuranic Isotope Burning in Thorium Fuel*. University of Cambridge, Cambridge.
- Lindley, B.A. et al., 2014. Steady state and Transient Core Feasibility Analysis for a Thorium-Fuelled Reduced Moderation PWR Performing Full Transuranic Recycle. *Ann. Nucl. Energy* 72, 320–337.
- Lindley, B.A., Fiorina, C., Gregg, R., Franceschini, F., 2015a. The effectiveness of full actinide recycle as a nuclear waste management strategy when implemented over a limited timeframe – Part I: Uranium fuel cycle. *Prog. Nucl. Energy* 85, 498–510.
- Lindley, B.A. et al., 2015b. Release of WIMS10: A Versatile Reactor Physics Code for Thermal and Fast Systems. Nice, Proceedings of ICAPP 2015.
- Lindley, B.A., Fiorina, C., Gregg, R., Franceschini, F., 2016. The effectiveness of full actinide recycle as a nuclear waste management strategy when implemented over a limited timeframe. Part II: thorium fuel cycle. *Prog. Nucl. Energy* 87, 144–155.
- Lindley, B.A. et al., 2017. Current status of the reactor physics code WIMS and recent developments. *Ann. Nucl. Energy* 102, 148–157.
- Lindley, B.A., Parks, G.T., 2012a. Near-complete transuranic waste incineration in a thorium fuelled pressurised water reactor. *Ann. Nucl. Energy* 40 (1), 106–115.
- Lindley, B.A., Parks, G.T., 2012b. The performance of closed reactor grade plutonium-thorium fuel cycles in reduced moderation pressurised water reactors. *Ann. Nucl. Energy* 47, 192–203.
- Lombardi, C., Mazzola, A., Padovani, E., Ricotti, M.E., 1999. Neutronic analysis of u-free inert matrix and thoria fuels for plutonium disposition in pressurised water reactors. *J. Nucl. Mater.* 274 (1–2), 181–188.
- Mark, J.C., 1993. Explosive properties of reactor-grade plutonium. *Sci. Glob. Secur.* 4 (1), 111–128.
- Nuclear Decommissioning Authority, 2010. *Plutonium: Credible Options Analysis (Gate A)*. Nuclear Decommissioning Authority. s.l..
- Nuclear Decommissioning Authority, 2014. *Progress on Approaches to the Management of Separated Plutonium*. Nuclear Decommissioning Authority. s.l..
- Nuclear Energy Agency, 2006. *Very High Burn-ups in Light Water Reactors*. OECD. s.l.
- Sasahara, A., Matsumura, T., Nicolaou, G., Papaioannou, D., 2004. Neutron and gamma ray source evaluation of LWR High Burn-up UO2 and MOX Spent Fuels. *Nucl. Sci. Technol.* 41 (4), 448–456.
- Schram, R.P.C., Klaassen, F.C., 2007. Plutonium management with thorium-based fuels and inert matrix fuels in thermal reactor systems. *Prog. Nucl. Energy* 49 (8), 617–622.
- Shapiro, N.L., Rec, J.R., Matzie, R.A., 1977. *Assessment of Thorium Fuel Cycles in Pressurized Water Reactors*. Electric Power Research Institute. s.l..
- Shwageraus, E., Hejzlar, P., Kazimi, M.S., 2004. Use of thorium for transmutation of plutonium and minor actinides in PWRs. *Nucl. Technol.* 147 (1), 53–68.
- The Royal Society, 1998. *Management of Separated Plutonium*. The Royal Society, London.
- Weaver, K.D., Herring, J.S., 2003. Performance of thorium-based mixed-oxide fuels for the consumption of plutonium in current and advanced reactors. *Nucl. Technol.* 143 (1), 22–36.
- Weber, W.J. et al., 1997. Radiation effects in glasses used for immobilization of high-level waste and plutonium disposition. *J. Mater. Res.*, 1947–1975
- Zainuddin, N.Z., Parks, G.T., Shwageraus, E., 2016. The factors affecting MTC of thorium-plutonium-fuelled PWRs. *Ann. Nucl. Energy* 98, 132–143.