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The factors affecting MTC of thorium–plutonium-fuelled PWRs

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ABSTRACT

Plutonium loading in a plutonium–thorium (Pu–Th) mixed oxide (MOX) fuelled pressurized water reactor (PWR) core is typically constrained by large maximum radial form factors (RFF) and positive moderator temperature coefficient (MTC). The large form factors in higher Pu content fuels stem from the large differences in burnup, and thus reactivity, between fresh and burnt fuel, while positive MTC can potentially be the result of the high soluble boron concentrations needed to maintain criticality for such reactive fuel. The conventional solution to these problems is the use of burnable poisons (BPs). While BPs are able to reduce RFF, the positive MTC is not entirely due to a large critical boron concentration (CBC) requirement. In fact, analysis shows a positive MTC in Th–Pu fuel is mainly caused by fissioning in the epithermal–fast energy range. A reduction in epithermal–fast fissioning through the use of certain BPs and the strategic employment of loading patterns that encourage leakage are more effective in attaining negative MTC, as a reduction in CBC has a negligible effect on MTC. This paper examines the contributions to positive MTC by isotope and energy and identifies characteristics of BPs that are able to mitigate positive MTC in a Pu–Th MOX PWR core.

1. Introduction

Plutonium (Pu) and minor actinides (MAs) present a significant nuclear proliferation concern as well as contributing to the long-term radiotoxicity of nuclear waste. There is a large amount of separated civil plutonium available today, with the largest stockpile in the world situated in the UK (Broomby, 2013).

With the delay in the deployment of fast reactors, the option to incinerate Pu in thermal reactors that are already widely in use is an attractive one. Typically, this is performed by mixing uranium (U) with Pu as mixed oxide (MOX) fuel (IAEA, 2003; Haas and Hamilton, 2007). An alternative approach is to use thorium (Th) as a fertile isotope instead of U. This increases the Pu incineration rate, as additional Pu is not bred from ^{238}U, although fissile ^{233}U is bred instead. Multiple lattice-physics-based investigations have confirmed this behaviour in pressurized water reactors (PWRs), e.g. studies by Galperin (1995) and Shwageraus et al. (2004), and such fuel has been shown to be feasible in a whole core (Forsberg and Kliem, 2011). Additionally, the Th–Pu matrix also shows potential for reducing radiotoxicity and decay heat when compared with standard U fuel waste (Wang et al., 2003; Hesketh, 2013). This offers an economic gain as it allows better management of spent fuel in repositories. Plutonium incineration can be pursued with either of the following competing objectives:

- *Fast burn:* Burn Pu quickly, i.e. increasing the *rate* of Pu incineration.
- *Deep burn:* Burn as large a fraction of the initially loaded Pu as possible, i.e. reducing the final amount of Pu in fuel.

A fast burn of Pu is achieved by decreasing the power share of all other nuclides (such as ^{233}U from the Th chain) in favour of primarily burning Pu, fast. Conversely, a deep burn means maximising the conversion of Th to ^{233}U, the presence of which will extend burnup of the fuel and allow more Pu to burn over time. However, a high Pu loading is required to sustain criticality as ^{233}U builds up slowly. The amount of Pu that can be loaded is limited by operational constraints on the maximum radial form factor (RFF) and prohibiting a positive moderator temperature coefficient (MTC).

Technically, there is no explicit requirement to keep the MTC negative at all operating conditions. The US NRC General Design Criterion No. 11 (NRC, 2016), for example, states: "The reactor core and associated coolant systems shall be designed so that in the power operating range, the net effect of the prompt inherent nuclear feedback characteristics tends to compensate for a rapid increase in reactivity. (This is negative feedback on a power transient)." However, a negative MTC requirement was imposed in this study to provide conservatism since detailed transient analysis

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was out of scope at this stage. The large form factors in higher Pu content fuels stem from the large differences in burnup between fresh and burnt fuel.

A positive MTC, on the other hand, can potentially be the result of the high soluble boron concentrations needed to maintain criticality for such reactive fuel due to a harder spectrum and reduced boron worth. The conventional solution to these problems is the use of burnable poisons (BPs).

This study investigates the contributions (by isotope and energy range) to positive MTC in high Pu content fuel. It then identifies characteristics of BPs that are able to mitigate positive MTC and presents potential BPs that can be used for this purpose.

1.1. Th–Pu fuel in PWRs

Fast reactors have not been widely commercially implemented yet. Hence the majority of spent nuclear fuel discharged from commercial reactors, which would have otherwise been efficiently incinerated in fast reactors, is now either due for interim storage, recycling, or direct final disposal. Civil Pu is recycled in some European countries (e.g. France, Belgium) while the United States and Russia have examined the option of incinerating their dismantled nuclear warheads (weapons-grade Pu). However, the presence of uranium (specifically ^{238}U) in conventional MOX fuel for the purpose of incinerating Pu means that Pu production can only be constrained, rather than being significantly reduced in content. Thus, the use of non-uranium fuels became of interest – inert-matrix (or fertile-free) fuel and thorium (Th). This work examines the use of thorium.

Over the years, interest in thorium has waxed and waned, with two recurring motivations for considering use of this fuel:

- *Reducing the risk of nuclear weapons proliferation:* Either in the form of using thorium with uranium, creating ^{233}U, which has self-protecting gamma-emitting daughter products in the decay chain of accompanying ^{232}U, or incinerating Pu from weapons or civilian stockpiles.
- *Cost cutting:* Increases in uranium spot prices and the fear of resource depletion prompted countries to investigate the use of thorium to reduce uranium ore and enrichment requirements in their nuclear fuel programmes.

While consideration of thorium for commercial use is still an academic exercise (IAEA, 2002, 2003, 2005), recent headway has been made by India and China (IThEO, 2015). However, these advances examine fuel mixes of either Th–U or Th–U–Pu. The natural progression is to consider pure Th–Pu mixes, which have better Pu incineration rates. Thus, studies on homogeneous Th–Pu fuel (without enriched or natural uranium) specifically in PWRs include:

- *Lattice calculations:* Galperin (1995), Puill (1999), Weaver and Herring (2004), Shwageraus et al. (2004), Dziadosz et al. (2004), Björk and Fhager (2009) and Herman (2009).
- *Full-core calculations:* Galperin and Raizes (1997), Lombardi et al. (1999), Joo et al. (2000), Forsberg and Kliem (2011), Trellue et al. (2011), Tsige-Tamirat (2011) and Björk et al. (2013).

These investigations compared the performance of different fuel mixes, e.g. Th–Pu/Th–Pu–U/weapons-grade/reactor-grade plutonium, quantified the materials attractiveness of the Pu product for use in weapons, analysed the fuel and potential SiC cladding at high burnup, and detailed the effects of hydrogen-to-heavy-metal (H/HM) ratio on Pu incineration.

The main observations concerning the fuel's behaviour are its reduced reactivity worth and delayed neutron yields which necessitate modification of reactivity control materials (e.g. use of boron enriched in ^{10}B), as well as more detailed transient analysis to confirm safe operation. As expected, the Th–Pu mix is found to yield substantially higher rates of Pu incineration compared to mixtures containing ^{235}U/^{238}U.

Note that in this study we consider a pure Th–Pu fuel mix, without any natural uranium denaturing. Denaturing is the process of adding a certain amount of natural uranium to fissile ^{233}U or ^{235}U in the fuel to inhibit weapons proliferation. As observed by Shwageraus et al. (2004) based on ^{233}U threshold limits proposed by Forsberg et al. (1999), there is a degradation in the efficiency of Pu destruction when the fuel is denatured. The repercussions of denaturing on our conclusions here will be analysed in future work.

1.2. Moderator temperature coefficient

The MTC is defined as the change in reactivity per degree change in moderator temperature. Changes in the moderator temperature subsequently insert positive or negative reactivity into the core. A negative MTC at hot full power is a mandatory requirement in all western countries operating PWRs. Thus, the MTC is tracked to ensure safe reactor operation. In a Th–Pu fuel mix, increasing the Pu content eventually causes the MTC to become positive. There are 5 main elements that have an effect on MTC (Martin, 2013):

- *Leakage:* For a fixed core pressure, an increase in temperature necessarily means a decrease in moderator density. This results in fewer collisions between fast neutrons and the hydrogen nuclei of the moderator, resulting in fewer neutrons slowing down or thermalising. Absorption cross-sections get smaller with the consequent harder neutron spectrum. This increases the mean free path of neutrons, and a larger proportion are therefore lost via leakage, causing a reduction in the effective multiplication factor (k_{eff}).
- *Thermal and resonance absorption:* A decrease in moderator density also reduces the number of neutrons absorbed in fissile material in the thermal range (especially in Pu fuel) creating a negative reactivity effect – a decrease in k_{eff}. However, depending on the nuclide mix, an increase in absorption in the epithermal–fast energy range might be either parasitic or contribute to fission.
- *Hydrogen absorption:* A moderator density decrease further reduces the number of neutrons lost to absorption by hydrogen in the coolant, creating a positive reactivity effect – an increase in k_{eff}.
- *Boron absorption:* If the reactivity of the core is controlled using soluble boron, then a decrease in coolant density also means that the amount of soluble boron in the reactor decreases, as some of it is forced out of the core, resulting in a decrease in neutron absorption in boron – an increase in k_{eff}.
- *Burnable poison absorption:* The shift of the neutron spectrum to higher energies generally causes a decrease in absorption in BPs, thus inserting positive reactivity into the core.

These effects all contribute to the overall MTC. Traditionally, the biggest effect on the MTC is due to the soluble boron. If critical boron concentration (CBC) is high, then a slight increase in temperature can cause a larger positive reactivity insertion. This could cause the net value of the MTC to be positive. Hence the use of BPs for overall reactivity control (rather than local power peaking reduction) is often in order to decrease the CBC requirement, rather