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The effectiveness of full actinide recycle as a nuclear waste management strategy when implemented over a limited timeframe – Part II: Thorium fuel cycle

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ABSTRACT

Full recycling of transuranic (TRU) isotopes can in theory lead to a reduction in repository radiotoxicity to reference levels in as little as ~500 years provided reprocessing and fuel fabrication losses are limited. However, over a limited timeframe, the radiotoxicity of the 'final' core can dominate over reprocessing losses, leading to a much lower reduction in radiotoxicity compared to that achievable at equilibrium. In Part I of this paper, TRU recycle over up to 5 generations of light water reactors (LWRs) or sodium-cooled fast reactors (SFRs) is considered for uranium (U) fuel cycles. With full actinide recycling, at least 6 generations of SFRs are required in a gradual phase-out of nuclear power to achieve transmutation performance approaching the theoretical equilibrium performance. U-fuelled SFRs operating a break-even fuel cycle are not particularly effective at reducing repository radiotoxicity as the final core load dominates over a very long timeframe. In this paper, the analysis is extended to the thorium (Th) fuel cycle. Closed Th-based fuel cycles are well known to have lower equilibrium radiotoxicity than U-based fuel cycles but the time taken to reach equilibrium is generally very long. Th burner fuel cycles with SFRs are found to result in very similar radiotoxicity to U burner fuel cycles with SFRs for one less generation of reactors, provided that protactinium (Pa) is recycled. Th-fuelled reduced-moderation boiling water reactors (RBWRs) are also considered, but for burner fuel cycles their performance is substantially worse, with the waste taking ~3–5 times longer to decay to the reference level than for Th-fuelled SFRs with the same number of generations. Th break-even fuel cycles require ~3 generations of operation before their waste radiotoxicity benefits result in decay to the reference level in ~1000 years. While this is a very long timeframe, it is roughly half that required for waste from the Th or U burner fuel cycle to decay to the reference level, and less than a tenth that required for the U break-even fuel cycle. The improved performance over burner fuel cycles is due to a more substantial contribution of energy generated by ^{233}U leading to lower radiotoxicity per unit energy generation. To some extent this an argument based on how the radiotoxicity is normalised: operating a break-even fuel cycle rather than phasing out nuclear power using a burner fuel cycle results in higher repository radiotoxicity in absolute terms. The advantage of Th break-even fuel cycles is also contingent on recycling Pa, and reprocessing losses are significant also for a small number of generations due to the need to effectively burn down the TRU. The integrated decay heat over the scenario timeframe is almost twice as high for a break-even Th fuel cycle than a break-even U fuel cycle when using SFRs, as a result of much higher ^{90}Sr production, which subsequently decays into ^{90}Y. The peak decay heat is comparable. As decay heat at vitrification and repository decay heat affect repository sizing, this may weaken the argument for the Th cycle.

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1. Introduction

Full recycling of transuranic (TRU) isotopes can in theory lead to a reduction in 'repository radiotoxicity' (defined as the radiotoxicity in Sv/GWeyr of power generated of the waste to be sent to geological disposal at the end of the scenario, although in practice this may go to multiple repositories and much of it may be stored on the surface for an indefinite period of time) to reference levels in as little as ~500 years (Grouiller et al., 2002) provided reprocessing and fuel fabrication losses are limited. However, this requires a long-term commitment to recycling (OECD Nuclear Energy Agency, 2002). Over a limited timeframe, the radiotoxicity of the 'final' core can dominate over reprocessing losses, leading to a much lower reduction in radiotoxicity compared to that achievable at equilibrium (National Nuclear Laboratory, 2014; Gregg and Hesketh, 2013).

While the heavy metal content in the repository dominates the radiotoxicity, this is by no means the only measure of repository loading or radiological hazard. The decay heat at time of loading and over the first few hundred years affects the repository size. Fission product isotopes (e.g. of I, Cs and Tc) are often the most mobile and hence form a large part of the radiological hazard (Lalieux et al., 2012; Nuclear Decommissioning Authority, 2010).

Time-dependent modelling is necessary to consider the performance of non-equilibrium systems. Theoretical and computational modelling of accelerator-driven system-based transmutation of a fixed fissile inventory was considered in OECD (2006), showing that several generations of reactors are required to achieve performance resulting in a large reduction in repository radiotoxicity. Reprocessing losses become significant after a few generations.

In Part I of this paper (Lindley et al., 2014a), TRU recycle over up to 5 generations of light water reactors (LWRs) or sodium-cooled fast reactors (SFRs) is considered for uranium (U) fuel cycles. Break-even and burner fuel cycles were considered in SFRs, and mixed low enriched uranium (LEU)-TRU LWR cores with zero net TRU production were also considered. With full actinide recycling, at least 6 generations of SFRs are required in a gradual phase-out of nuclear power to achieve transmutation performance approaching the theoretical equilibrium performance. TRU recycle in PWRs with zero net actinide production provides similar performance to LEU-fuelled LWRs in equilibrium with a fleet of SFRs operating with a burner fuel cycle. However, it is not possible to reduce the TRU inventory over multiple generations of PWRs. TRU recycle in SFRs operating a break-even fuel cycle is much less effective from a point of view of reducing waste radiotoxicity.

In this paper, the analysis is extended to the thorium (Th) fuel cycle. Closed Th-based fuel cycles are well known to have lower equilibrium radiotoxicity than U-based fuel cycles due to much lower TRU production from ^{232}Th than from ^{238}U (Franceschini et al., 2012; IAEA, 2005), although the period for which the radiotoxicity is lower is limited to ~35,000 years, after which the radiotoxicity of ^{233}U and its daughters becomes most significant (Coates, 2011; Fiorina et al., 2013a). However, it is also well known that it takes a long time for the advantages of 'equilibrium' Th fuel cycles to be realised due to the long transition time to equilibrium (Hesketh and Thomas, 2013; Fiorina et al., 2013b, 2013c).

Franceschini et al. (2013) compared Th- and U-based transmutation strategies from a point of view of fuel fabrication and reprocessing requirements. Th-based transmutation is a much less developed technology than U-based transmutation. While further developments are required in either case for full recycle of TRUs, notably for MA reprocessing and fuel fabrication, additional technology developments are required for the Th fuel cycle. Reprocessing of Th fuel is not currently an industrial-scale process, and the Th-TRU fuel cycle introduces a greater range of isotopes that need to be recovered compared to U-TRU and Th–U3[1] cycles in isolation. Remote fuel fabrication is required in any case due to spontaneous neutron (SN) emission from Cm isotopes (and Cf for thermal recycle schemes), but the presence of U3 further complicates this due to the high-energy gamma emitters present in its decay chain.

Here, the time-dependent performance of Th fuel cycles is modelled using the fuel cycle code ORION (Gregg and Hesketh, 2013) for up to 5 generations of recycling reactors. SFRs with break-even and burner fuel cycles and LWR-based recycling are also considered. For a burner fuel cycle, this requires a harder neutron spectrum to improve the fissibility of TRU isotopes (Lindley et al., 2013), which leads to consideration of reduced-moderation boiling water reactors (RBWRs). RBWRs have also been recently considered for a Th break-even fuel cycle (Ganda et al., 2011). The radiotoxicity of Th break-even and burner fuel cycles at equilibrium for SFRs and RBWRs is comparable (Lindley et al., 2014b). However, higher specific power reactors have a more rapid transition to equilibrium (Hesketh and Thomas, 2013), and RBWRs have a relatively low power density compared to SFRs, which is therefore expected to slow their transition to equilibrium. These scenarios are not exhaustive, but give representative cases for fast and epithermal reactors operating at typical power densities. In particular: other liquid metal or gas-cooled fast reactors can be expected to have similar performance to the SFR; molten salt reactors may have a fast or epithermal neutron spectrum with a power density somewhat similar to SFRs (Hesketh and Thomas, 2013); however, the cases considered may not be representative of highly-moderated reactors operating a Th break-even fuel cycle due to the substantially different neutron spectrum (e.g. Nuttin et al., 2012). Finally, hybrid scenarios which consider a mix of U and Th fuel are not considered, e.g. in RBWRs (Gorman et al., 2014) or using a combination of SFRs and heavy-water moderated reactors (World Nuclear Association, 2014).

The impact of minor actinide (MA: consisting of Pa, Np, Am, Cm, Cf) recycling is also considered. Reprocessing of Pa is a particular challenge of the Th cycle. Pa normally remains with the fission products for THOREX fuel reprocessing. Recycling of long-lived ^{231}Pa may be desirable to reduce long-term radiotoxicity (IAEA, 2003). However, ^{231}Pa capture is the principal route to ^{232}U production. ^{232}U production can be reduced by ~70% by not recycling Pa, reducing the gamma source at fuel fabrication (Lindley et al., 2014c).

2. Scenarios considered

The fuel cycle code ORION has been used to model the transition from an open (relying on standard LWR technology) to a closed fuel cycle (involving SFRs or RBWRs). For these scenarios, a fleet of LEU-fuelled LWRs is assumed to come online in Year 1. In Year 41, the closed cycle reactors are subsequently switched on. All reactors operate for 60 years, and the LWRs are not replaced at their end of life, as any future generations of LWRs may be supported by their own fleets of recycling reactors. The 40 year gap between LEU-fuelled LWRs and recycling reactors is similar to that typically assumed, e.g. scenarios with a 2015 start date with fast reactor switch-on in 2050. Reprocessing of fuel for a 40 year period before use of recycling reactors is longer than sometimes considered but here is utilised to simplify the scenario.

Successive generations of recycling reactors are then started when the preceding generation reaches end of life. The simultaneous replacement of all the reactors in the fleet would cause a sharp but temporary reduction in the separated Pu/TRU/U3

[1] U3 signifies U bred from Th.

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