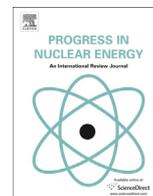




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The effectiveness of full actinide recycle as a nuclear waste management strategy when implemented over a limited timeframe – Part II: Thorium fuel cycle



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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 ²³³U 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 ⁹⁰Sr production, which subsequently decays into ⁹⁰Y. 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/GW_{yr} 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- U^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.

inventory when the old cores were discharged, which may result in insufficient material to refuel the reactors. Here, this is not modelled – the life of the preceding generation of reactors is instead extended. In practice, reactors would have slightly different start dates and lifetimes so this reduction in inventory would not occur on the same scale. 5 years cooling is assumed for all fuels before reprocessing (approximately the minimum required for aqueous reprocessing). Reprocessing and fuel fabrication take a single timestep in ORION – 3 months for the RBWR and 6 months for the SFR, which is in addition to the 5 years cooling time.²

For burner fuel cycle scenarios, the ratio of LEU-fuelled reactors to SFRs/RBWRs and the ratio of reactors in successive generations of SFRs are constrained by the core inventories required to start up and fuel the SFRs/RBWRs. This leads to the production of fuel inventories for recycling reactors from previous tiers which are not always used. The minimum material left at the end of the scenario is that of the final discharged core and fuel which has been cooled for a few years but not been reprocessed. This severely limits the proportion of heavy metal which can be recycled. In addition, there may be unused inventories at the end of the scenario which have not been burned. These inefficiencies will likely be unavoidable (discrete reactor effects, unplanned shutdowns, logistics etc.) and will likely be significant.

For break-even fuel cycle scenarios, the net Pu/TRU/U3 production is zero once the LEU-fuelled LWRs go offline. Here, the unused TRU from the LEU-fuelled LWRs is not counted in the spent fuel as it is assumed the fleet of recycling reactors can be more readily scaled to use all the TRU (e.g. by 'fine-tuning' of the conversion ratio of the recycling reactors over the first few cycles – indeed in reality a conversion ratio greater than 1 may be needed to properly scale the reactor fleet). The extra fission products from the LEU-fuelled LWRs which correlate to the production of unused TRU are not included in the long-term radiotoxicity. However, they are included in the repository decay heat over the scenario (Section 4.3). This is because these extra fission products correspond to roughly the last 20–30 years of operation for the LWRs. If the contribution of these fission products to the decay heat was removed, this could be accomplished by considering the LWRs to shut down early. However, this is considered unrepresentative of the repository decay heat in reality.

0.1% reprocessing losses are assumed in the ORION models, but the effect of 1% reprocessing losses is also discussed.

The scenarios considered are summarised in Table 1.

3. Scenario modelling

ORION uses cross-sections and spectra produced using a reactor physics code to calculate the discharged fuel composition as a function of the loaded fuel composition. The loaded fuel changes throughout the scenario due to decay processes, and changing inventories from other reactors in the scenario. Infinite dilution cross-sections from the TRAIL library (ANSWERS, 2013) are condensed to one group using flux spectra from the reactor physics code and used for isotopes not significant from a reactor physics perspective. The reactor parameters are given in Table 2.

4-loop Westinghouse LEU-fuelled PWRs are considered with a rating of 3411 MWth and 1150 MWe.

A 1000 MWth SFR is considered based on the Advanced Recycling Reactor (Dobson, 2008) with 3 batches and a 1 year cycle length. For the SFR operating with a burner fuel cycle, oxide fuel has been considered. The SFR U3+TRU loading is 44.2% and 38.1% with

Table 1

Scenarios considered. # denotes that 1, 2, 3, 4 and 5 generations of reactors are all considered.

Scenario	Reactor	Fuel	Fuel cycle
LEU-OT	PWR	LEU	Once-through
Th-SFR-Bu-MA#	SFR	Th-Pa-U3-TRU	Burner
Th-SFR-Bu-NoPa#	SFR	Th-U3-TRU	Burner
Th-SFR-Bu-Pu#	SFR	Th-U3-Pu	Burner
Th-SFR-BE-MA#	SFR	Th-Pa-U3-TRU	Break-even
Th-SFR-BE-NoPa#	SFR	Th-U3-TRU	Break-even
Th-SFR-BE-Pu#	SFR	Th-U3-Pu	Break-even
Th-RBWR-Bu-MA#	RBWR	Th-Pa-U3-TRU	Burner
Th-RBWR-Bu-NoPa#	RBWR	Th-U3-TRU	Burner
Th-RBWR-Bu-Pu#	RBWR	Th-U3-Pu	Burner
Th-RBWR-BE-MA#	RBWR	Th-Pa-U3-TRU	Break-even
Th-RBWR-BE-NoPa#	RBWR	Th-U3-TRU	Break-even
Th-RBWR-BE-Pu#	RBWR	Th-U3-Pu	Break-even

and without MAs respectively. This leads to a TRU incineration rate of ~16% and ~20% per pass respectively, corresponding to ~273 kg/GWthyr in both cases. For the SFR with a break-even fuel cycle, nitride fuel has been used, and over the first generation of SFRs, the seed has 25.9% and 21.5% TRU+U3 loading with and without MAs respectively. After this, the core contains predominantly Th-U3 with 20.5% TRU+U3 loading in both cases. The core configurations are shown in Fig. 1.

The RBWR utilises the same plant as an ABWR but with a tight pitch triangular lattice. The core contains 720 hexagonal assemblies (Fig. 2) and the core area is ~50% greater than an ABWR. The core rating is 3926 MWth. The average void fraction is ~53%. The RBWR operating with a burner fuel cycle utilises a heterogeneous assembly with Th-Pu-(MA) and Th-(Pa)-U3 pins in different areas of the fuel assembly as this greatly improves the neutronic performance (Fig. 3) (Lindley et al., 2013, 2014b).

With a burner fuel cycle, the RBWR U3+TRU loading is 23.9% and 20.5% with and without MAs respectively. This leads to a TRU incineration rate of ~13% and 17% per pass respectively, corresponding to ~130 kg/GWthyr with MAs and ~158 kg/GWthyr without MAs. The incineration rate is limited by the need to keep the void coefficient negative, and is therefore lower than in the SFR (Lindley et al., 2014b). Over the scenario, RBWRs with a break-even fuel cycle are loaded with 16.4% and 16% U3+TRU with and without MAs respectively. The start-up core of an RBWR operating a break-even fuel cycle may require a higher loading due to the lower fissibility of the initial TRU feed. Also, the RBWR with both the burner and break-even fuel cycles may not be able to start up while simultaneously satisfying void coefficient and acceptable cycle length constraints, as the higher TRU loading is likely to result in a positive void coefficient. The solution to this is to utilise an intermediate pass of Th-Pu MOX fuel, as considered by Lindley et al. (2014c, 2014d), or to utilise 'wetter' assemblies in the first pass through the RBWR core, which would result in a lower TRU loading being required to maintain criticality. This is not modelled here to simplify the analysis and maintain a consistent comparison between the SFR and the RBWR, as an intermediate pass with a different reactor configuration results in different mass flows and inventories. Only the first generation of recycling reactors is likely to be affected; and the difference in radiotoxicity from utilising this intermediate step is relatively small (Lindley et al., 2014c).

The ORION model consists of fuel fabrication facilities, reactors, buffers (which store material) and plants (which route and separate material). The inventories of 2500 isotopes were tracked, allowing the radiotoxicity to be accurately calculated. A typical ORION model for the burner fuel cycle utilising SFRs used in this study is shown in Fig. 4. For the break-even fuel cycle scenarios, the SFR core and

² The step size is determined by the maximum step size required to accurately model the reactors' batch strategies – hence the difference.

Table 2
Reactor parameters.

Reactor & fuel cycle	Fuel	Fuel residence time/number of batches	Discharge burn-up (GWd/t)	Specific power (MWth/t)	Isotope vector used for reactor physics calculations	Reactor physics method
PWR	LEU	4.5/3	52	38.1	4.4 wt% LEU	WIMS10 lattice calculation (Newton et al., 2008)
RBWR burner	Th-(Pa)-U3-Pu-(MA) oxide	9/4 ^a	86.1	26.2	Isotope vector from equilibrium study (Fiorina et al., 2013c; Lindley et al., 2014b)	SERPENT 3D pincell calculation (Leppänen, 2007)
Break-even RBWR		9/4 ^a	87.8 (seed) 3.9 (blanket)	26.7 (seed) 1.3 (blanket)		ERANOS core calculation (Rimpault et al., 2002)
SFR burner		3/3	97.1	104.2		
Break-even SFR	Th-(Pa)-U3-Pu-(MA) nitride	3/3 (seed) 6/3 (blanket) ^a	73.7 (seed) 5.0 (blanket)	79.2 (seed) 2.7 (blanket)		

^a The RBWRs in the equilibrium study operate a mixed 4/5-batch strategy with a cycle length of 2 years. This roughly corresponds to a 4-batch strategy with a cycle length of 9 years. Also, the axial blanket in the SFR in reality will reside in the core for the same length of time as the seed, i.e. 3 years. These approximations make very little difference to the ORION calculations and simplify the model, as having fuel elements operate with different batch strategies requires defining two reactors in the model.

blanket were modelled separately, with different ‘reactors’ and cross-sections. The blanket was fuelled exclusively with Th.

Th recovered from reactors is cooled for a further 20 years before fuel fabrication to allow ²²⁸Th and its daughters (notably high-energy gamma sources ²⁰⁸Tl and ²¹²Bi) to decay. ²²⁸Th is produced by ²³²U decay, and these have half-lives of 1.9 and 69 years respectively. The ²³²U in the U3 will decay into ²²⁸Th and its daughters, replenishing the high-energy gamma source in the short term. However, this takes a few years and hence the gamma source is greatly reduced compared to fabricating fuel containing recently irradiated Th. Similarly, after 20 years of cooling, the high-energy gamma emitters in the recovered Th have decayed away, meaning that the Th can be used in fuel fabrication. This has very little impact on the results presented in this paper.

For the burner fuel cycle scenarios, the ratio of LEU-fuelled PWRs, and SFRs/RBWRs in each generation is limited by TRU availability. The limiting point for the first generation of SFRs/RBWRs is reactor start-up (in Year 41) and the first few reloads until reprocessed TRU + U3 from the SFRs/RBWRs is available to supplement the TRU produced by the still-operating LWRs. For subsequent generations, the discharged cores from the previous generation are burned in a progressively smaller fleet of reactors. Each generation is smaller than the last, meaning that not all of the discharged core is loaded into the fresh core. The remainder of material from the discharged core is then used to provide fuel for

the subsequent generation over its lifetime. The SFR/RBWR capacity becomes lower than that of a single plant – but the ratio of reactors is the important parameter and it can be readily assumed that a large reactor fleet can be scaled accordingly. In any case, subsequent generations of LWRs and their associated SFRs/RBWRs will increase the SFR/RBWR capacity beyond that considered for the scenario. The number of reactors in each generation is shown in Table 3.

The resulting TRU+U3 inventory for Th-RBWR-Bu-MA5 is shown in Fig. 5. The TRU accumulated from the LEU-fuelled PWRs is used to start SFRs/RBWRs after 40 years. The TRU inventory increases after start-up due to continued operation of LEU-fuelled PWRs. From 60 years onwards, no further TRU is produced by the LEU-fuelled PWRs and the inventory decreases. After 100, 160, 220 and 280 years, unloading of one generation of SFRs/RBWRs provides inventory for the next generation. For the SFRs, each generation is roughly half the size of the preceding generation. For the RBWR, the incineration rate per GWthyr is slower than for the SFR and hence the RBWR fleet size decreases at a slower rate. The large core inventory required to start up the first generation of reactors limits the size of this generation, which combined with the low incineration rate means that there is no reduction in fleet size for the second generation.

For break-even fuel cycle scenarios, a constant fleet size of RBWRs/SFRs is maintained. As shown in Table 2, the RBWR requires

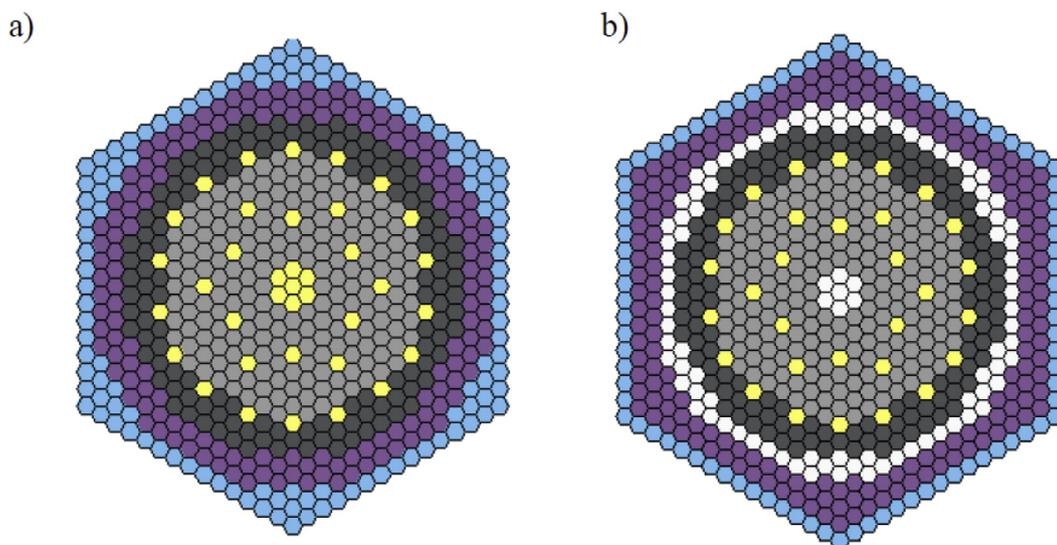


Fig. 1. SFR core designs for burner (a) and break-even (b) fuel cycles. Light grey = inner core, dark grey = outer core; yellow = control rods; violet = steel shield; blue = B4C shield; white = blanket. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

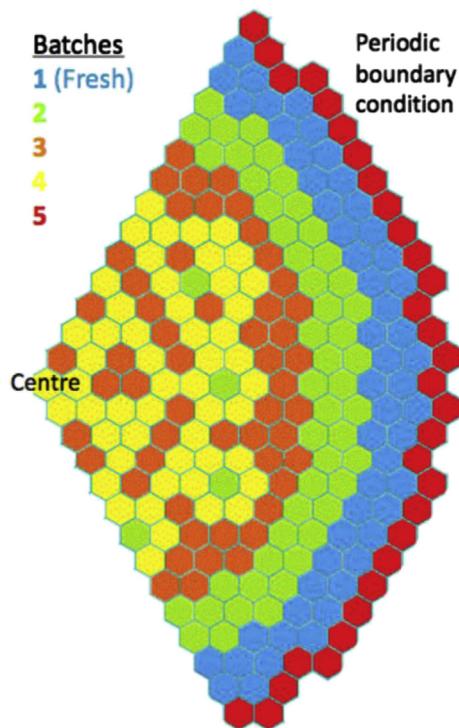


Fig. 2. RBWR core fuel loading pattern in one-third rotational symmetry.

a much larger fissile core inventory than the SFR which results in greater LWR capacity being required at start-up.

A reference level radiotoxicity is adopted (as considered, for example, in OECD (2002)), which corresponds to the radiotoxicity of the unburned natural U required to fuel a typical once-through LWR of the same electrical energy output. Daughter products from the decay of natural U are assumed to be at their equilibrium values. Using a European Pressurised Reactor (EPR) as the reference

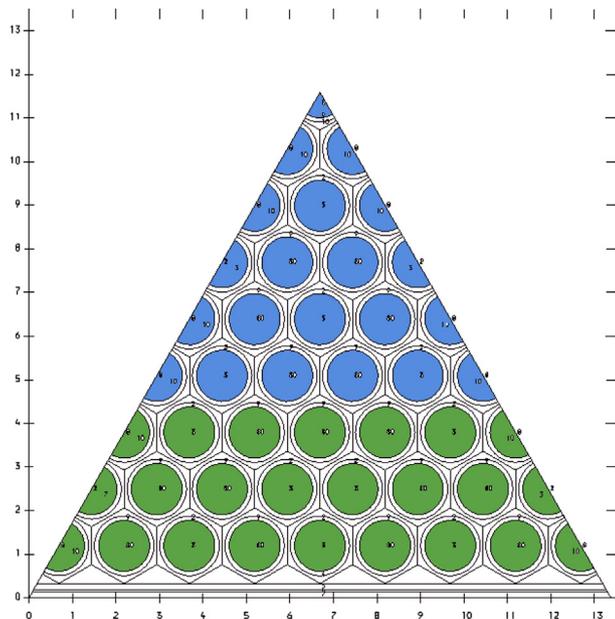


Fig. 3. RBWR fuel assembly design. Centre of assembly (blue) = Th-TRU. Periphery of assembly (green) = Th-U3. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

once-through LWR to determine natural U requirements, this results in a time-constant reference radiotoxicity level equal to 5.9×10^6 Sv/GWeyr.

4. Radiotoxicity and decay heat

4.1. Repository radiotoxicity for burner fuel cycles

The radiotoxicity over 5 generations of SFRs is plotted in Fig. 6. Time is measured relative to the scenario end, which for multiple generations of SFRs is up to 300 years after the LWRs (which produce the majority of the energy) are switched off – therefore the radiotoxicity in Year 1 decreases steadily with generation number. The radiotoxicity before Year 1 is also relevant as the fission products will be vitrified long before Year 1 in Fig. 6. However, on a timeframe of >1000 years, decay prior to the end of the scenario becomes irrelevant and the radiotoxicity of the different cases becomes comparable.

Scenarios with and without Pa recycling are presented. With a logarithmic representation of decay time, the effect of recycling Pa becomes perceptible after around 3 generations of SFRs. Beyond this point the radiotoxicity reduces such that ^{231}Pa and its daughter ^{227}Ac become significant contributors after ~1000 years.

In each generation, the mass of U3+TRU remaining roughly halves. For scenarios with Pa recycling, the U3+TRU specific radiotoxicity also slightly reduces as the proportion of U3 in the waste steadily rises over the scenario. This leads to a reduction of 30–40% in time taken for the waste to decay to the reference level for each additional generation.

The radiotoxicity over 5 generations of RBWRs is plotted in Fig. 7. In this case, the time taken for the waste to decay to the reference level initially increases relative to the once-through cycle, due to the effect of breeding a relatively large amount of ^{233}U and its daughters as the RBWR core is much larger than the SFR core. As the number of generations increases beyond 2, the end-of-scenario ^{233}U inventory decreases, leading to a reduction in time taken for waste to decay to the reference level. The reduction is much more modest than with SFRs due to the much lower specific power (which also results in a smaller fleet of reactors). ~3–4 generations of RBWRs are necessary before Pa recycling becomes worthwhile.

The burner fuel cycle scenarios are compared in Fig. 8, which shows the time taken to decay to the reference level for multiple generations of reactors under different recycling strategies. Also included are the results for multiple generations of SFRs operating using the U cycle, with results taken from Part I of this paper (Lindley et al., 2014a).

Without MA recycling, the reduction in repository radiotoxicity is much more limited and saturates after ~3–4 generations of SFRs. RBWRs again result in a lower reduction in time taken to decay to the reference level than SFRs, with 1–2 generations more of RBWRs required to achieve the same time reduction as SFRs.

Th-SFRs which recycle all actinides result in a lower time to decay to the reference level than U-SFRs for at least 3 generations of SFRs. This advantage is essentially contingent on recycling of Pa. Without Pa recycle, Th-SFRs and U-SFRs have very similar radiotoxicity for at least the first 5 generations of SFRs.

The effect of ~1% reprocessing losses was investigated in Part I of this paper (Lindley et al., 2014a). For up to 1% reprocessing losses, the effect becomes significant after ~5 generations of U-SFRs. This result will mostly generalise to the Th-fuelled SFRs and RBWRs considered here. However, unlike the U cycle, where radiotoxicity is essentially proportional to non-fertile inventory after a few generations, with the Th fuel cycle the proportion of U3 relative to TRU rises over time. With higher reprocessing losses, more of the initial TRU feed will be lost before it can be burned, leading to

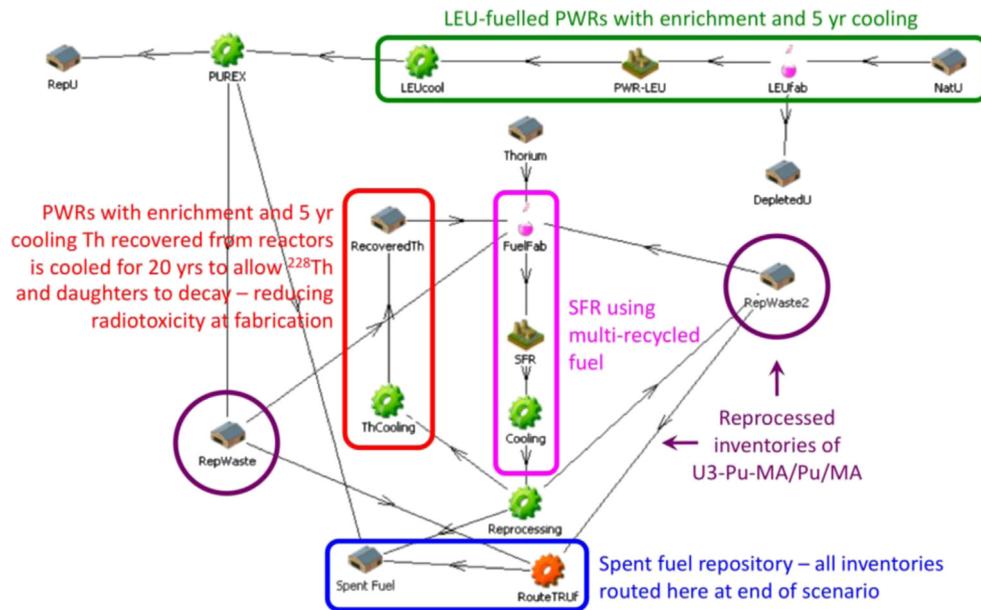


Fig. 4. ORION fuel cycle scenario model.

reprocessing losses having a more significant impact. Also, the higher end-of-scenario inventories for RBWR cases mean that a slightly higher number of generations may be required before reprocessing losses become significant.

Finally, it must be stressed that all results in this section assume a gradual phase-out of nuclear power over several generations of reactors. If further generations of LWRs are built, then the reduction in repository radiotoxicity is much smaller. This is discussed in detail in Part I of this paper (Lindley et al., 2014a).

Multiple generations of LEU-fuelled LWRs will ultimately be constrained by natural U reserves. At this point, reactors operating with a break-even or breeder fuel cycle are required if nuclear power is not phased out. From Part I of this paper (Lindley et al., 2014a), U-fuelled SFRs operating a break-even fuel cycle are not particularly effective at reducing repository radiotoxicity. Th-fuelled reactors operating a break-even fuel cycle are considered in the following section.

4.2. Repository radiotoxicity with a break-even fuel cycle

The repository radiotoxicity of a break-even fuel cycle with SFRs is shown in Fig. 9. Scenarios with and without Pa recycle are displayed with solid and dashed lines respectively. With Pa recycle, the time for the waste to decay to the reference level drops to ~1400 years within 3 generations.

Without Pa recycle, the radiotoxicity of ^{231}Pa and ^{227}Ac severely limit the achievable reduction in repository radiotoxicity. Little

further reduction is achieved beyond 2 generations of SFRs, leading to a long (~44,000 year) time to decay to the reference level. The radiotoxicity contributions for Th-SFR-BE-NoPa5 are shown in Fig. 10. The $^{231}\text{Pa} + ^{227}\text{Ac}$ radiotoxicity dominates over a timeframe of ~1000 to ~50,000 years, resulting in radiotoxicity around twice the reference level during this timeframe. Not recycling Pa reduces the ^{232}U in the fuel at fabrication by ~70%, and thus is advantageous from a fuel fabrication standpoint.

With RBWRs, the break-even fuel cycle radiotoxicity follows a similar trend to that with break-even SFRs (Fig. 11). Over ~1–3 generations with Pa recycle, RBWRs yield a lower reduction in radiotoxicity than the SFRs, but they slightly outperform SFRs over 4–5 generations (Fig. 12). Without Pa recycle, RBWRs yield a significantly higher reduction in time to decay to the reference level than SFRs. In this case, the radiotoxicity of $^{231}\text{Pa} + ^{227}\text{Ac}$ up until ~50,000 years is slightly below the reference level rather than slightly above it. This is partly a result of ~25% lower production of ^{231}Pa per GWe in the RBWR than the SFR (as a result of the different flux spectrum in the reactor), but mostly a result of normalisation per unit energy production: the RBWR requires a ~3× larger fleet of LWRs to generate sufficient Pu for start-up. This increases the radiotoxicity over the first few generations (due to a higher TRU inventory and longer transition time), but results in normalisation of the radiotoxicity over higher energy production (from the initial LWRs), which in turn results in reduced radiotoxicity in Sv/GWeyr over a higher number of generations. Hence the ‘improvement’ in radiotoxicity for the RBWR must be viewed with caution: the actual

Table 3
Scenario reactor capacities.

Reactor generation	Starting year	Capacity (GWe)			
		Th-SFR-Bu-MA/Th-SFR-Bu-NoPa	Th-SFR-Bu-Pu	Th-RBWR-Bu-MA/Th-RBWR-Bu-NoPa	Th-RBWR-Bu-Pu
LEU-PWR	1	11.50	11.50	11.50	11.50
Generation 1	41	2.730	2.520	2.034	2.034
Generation 2	101	1.470	1.155	2.034	1.582
Generation 3	161	0.630	0.420	1.243	0.904
Generation 4	221	0.315	0.210	0.791	0.452
Generation 5	281	0.158	0.105	0.452	0.226

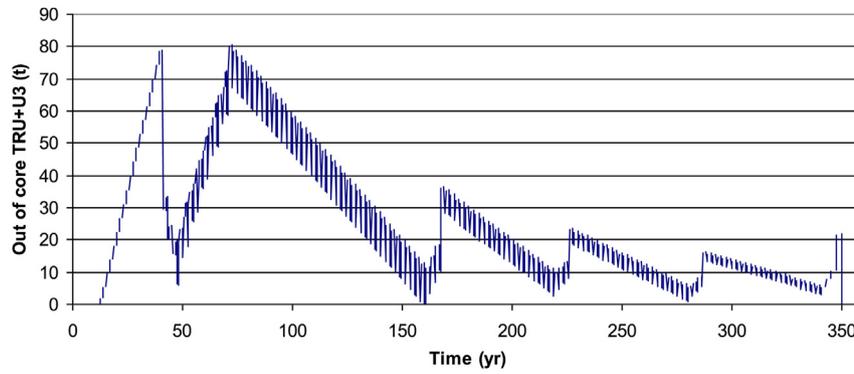


Fig. 5. TRU inventory for Th-RBWR-Bu-MA5.

repository loading will be similar to the SFR. In general, the radiotoxicity in Sv/GW_{yr} is essentially a measure of repository loading per unit energy production, and it must be stressed that in any case a long-term geological repository will be required.

Fig. 12 also shows the effect of a break-even fuel cycle utilising U-fuelled SFRs (results from Part I of this paper (Lindley et al., 2014a)). The reduction in repository loading with the U fuel cycle is much lower, as it is dominated by the final core inventory, which contains a substantial amount of TRU, unlike the Th-fuelled cores. Indeed, the Th fuel cycle achieves comparable radiotoxicity

reduction to the U fuel cycle without MA recycle. However, it must be noted that the challenges of recycling Th and U3 are likely to be even greater than the challenges of recycling MAs (Franceschini et al., 2013).

Finally, the effect of 1% reprocessing losses is shown in Fig. 13. The ORION model is not otherwise altered to make the comparison fair, but this slightly reduces the available fissile inventory. As the same amount of energy is generated from each case, the main effects are to reduce the amount of TRU that is recycled but to slightly increase the amount of U3 that is burned (hence reducing the final

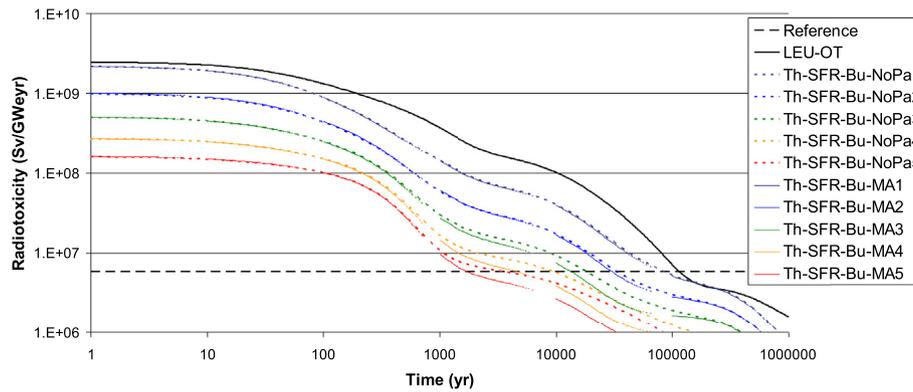


Fig. 6. Repository radiotoxicity for SFR burner fuel cycle scenarios.

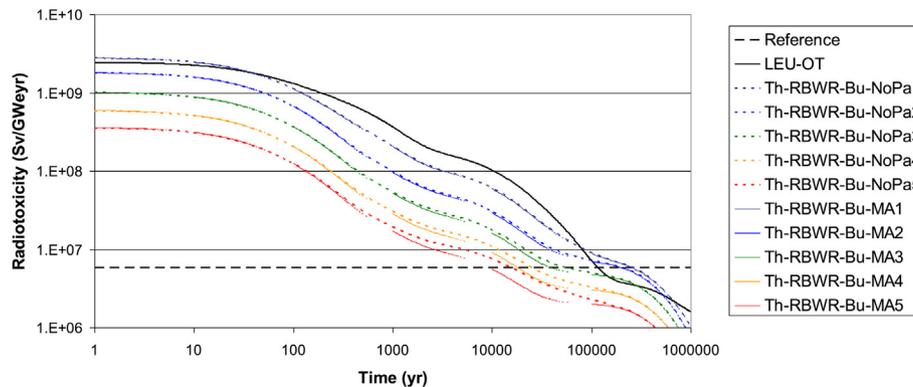


Fig. 7. Repository radiotoxicity for RBWR burner fuel cycle scenarios.

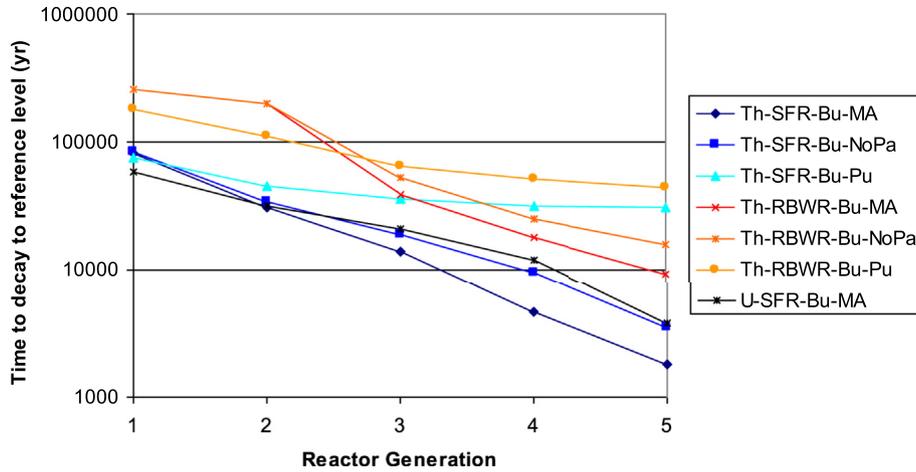


Fig. 8. Comparison of repository decay times for burner fuel cycle scenarios.

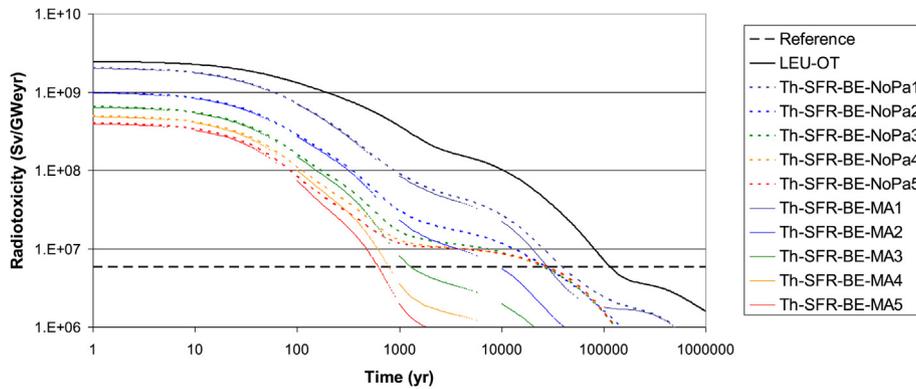


Fig. 9. Repository radiotoxicity for break-even fuel cycle scenarios with SFRs.

U3 inventory). Th-RBWR-BE-MA5 is not shown in Fig. 13 as the additional reprocessing losses mean that there is insufficient fissile feed for the final generation of RBWRs.

The higher reprocessing losses cause a noticeable impact within 1 generation, and over 5 generations the radiotoxicity of the repository becomes substantially higher than with 0.1% reprocessing losses, with the time taken for decay to the reference level increasing by a factor of ~3–6 for a given number of generations. This is in contrast to the U burner fuel cycle scenarios utilising SFRs considered in Part I of this paper (Lindley et al., 2014a), where ~5 generations of SFRs were required before the reprocessing losses

became significant. Here, the specific radiotoxicity of the final core inventory becomes much lower than the radiotoxicity of the reprocessing losses, as the reprocessing losses contain a relatively high proportion of TRU whereas the final core inventory contains very little TRU and is mostly Th+U3.

4.3. Decay heat

Recycling of Pu and MAs can also reduce the peak and integrated heat load in the repository (Gregg and Hesketh, 2013). In this section, the decay heat for break-even fuel cycle scenarios is

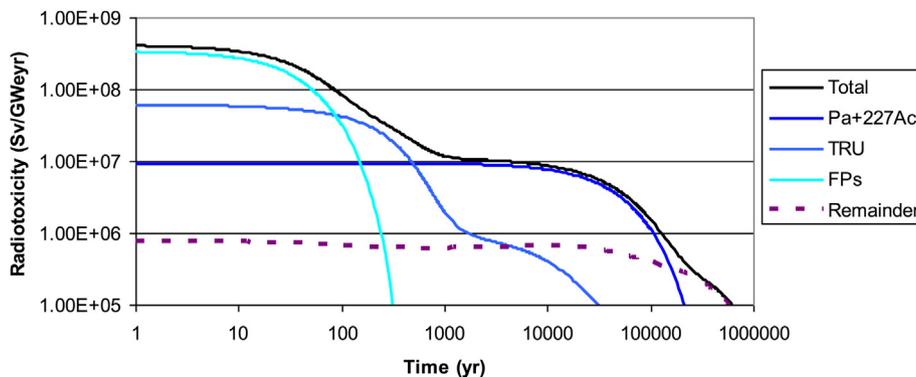


Fig. 10. Contributions to radiotoxicity of Th-SFR-BE-NoPa5.

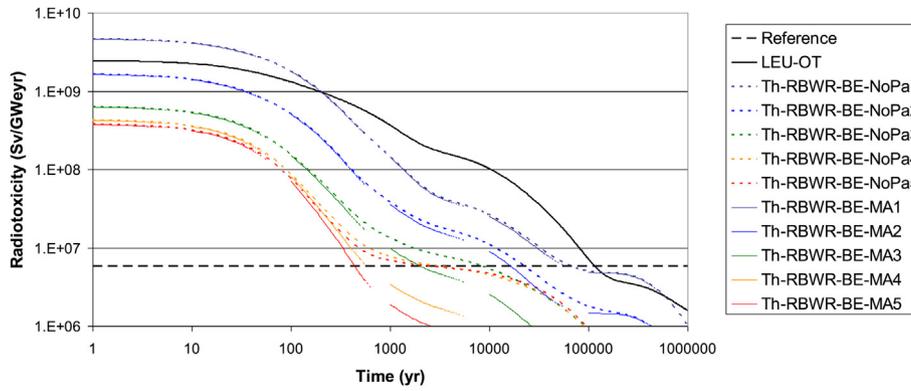


Fig. 11. Repository radiotoxicity for break-even fuel cycle scenarios with RBWRs.

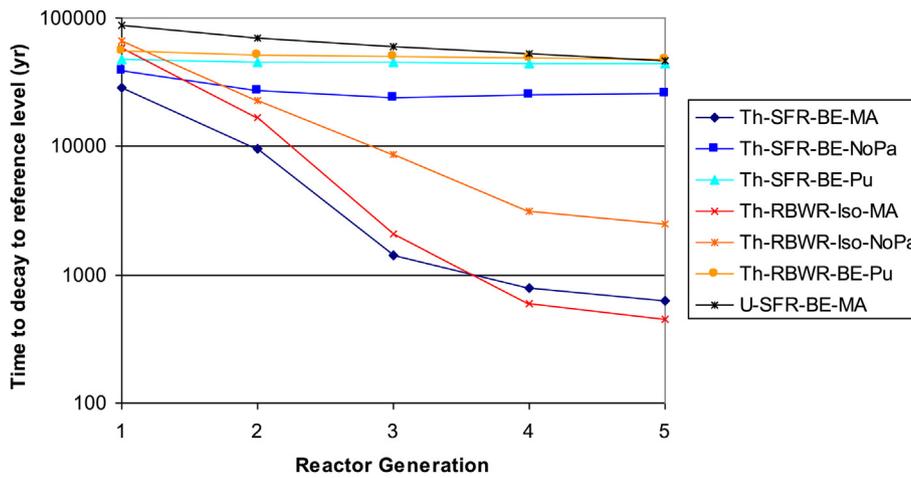


Fig. 12. Comparison of time to decay to the reference level for break-even fuel cycle scenarios.

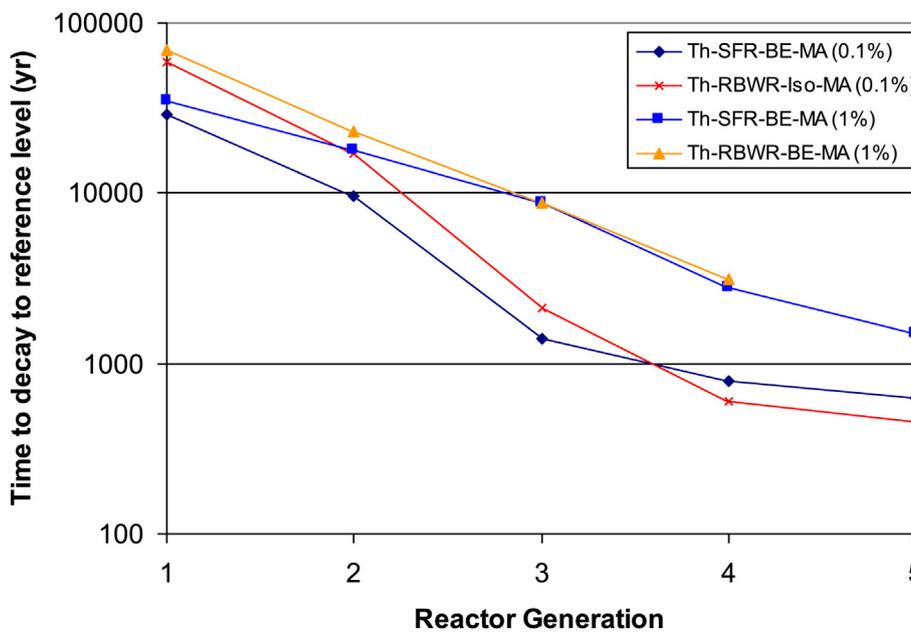


Fig. 13. Effect of 1% compared to 0.1% reprocessing losses on time to decay to the reference level for break-even fuel cycle scenarios.

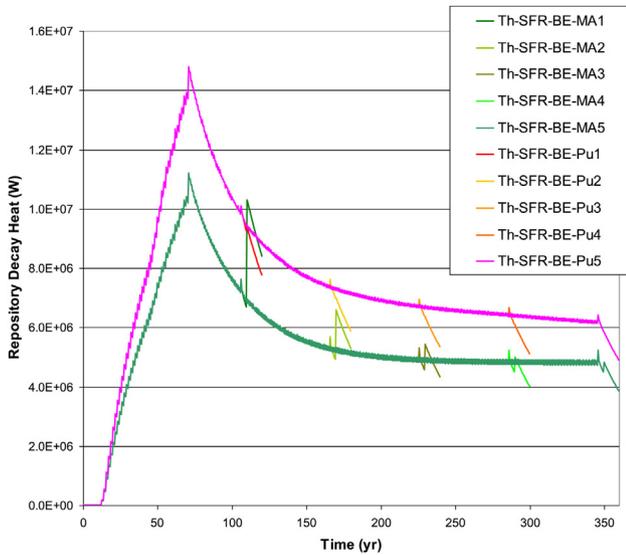


Fig. 14. Repository decay heat for break-even fuel cycle scenarios with SFRs.

investigated, to derive general conclusions on the relative behaviour of Th and U fuel cycles and SFRs compared to RBWRs.

The decay heat for break-even fuel cycle scenarios with SFRs is shown in Fig. 14. The decay heat is not sensitive to whether Pa is recycled as the ^{232}U contribution is relatively small. A fleet of 10 LEU-fuelled LWRs followed by 10 Th-fuelled SFRs is considered, for direct comparison with the U-SFR results in Part I of this paper (Lindley et al., 2014a). The initial peak in Fig. 14 occurs when the LWRs are unloaded, after which there is a sizeable reduction in fleet size and hence this peak is essentially an artefact of the simplified scenario considered here. However, the relative magnitude of the peaks with and without MA recycle is indicative of the effect of MA recycle. In some cases, there is a subsequent peak when the final cores are unloaded. Before the final core is unloaded, the repository decay heat is higher for Th-SFRs as a result of higher fission product decay heat, predominantly due to $\sim 3\times$ higher ^{90}Y production (a decay product from ^{90}Sr , which is produced in greater quantities in the Th-SFR) (Fig. 15).

Following reprocessing, high-level waste is vitrified and stored on the surface prior to loading in the repository (Gregg and Hesketh, 2013). The heat output from the glass is limited by material constraints: the maximum proportion of waste that can be incorporated into the glass depends on the decay heat. It is possible

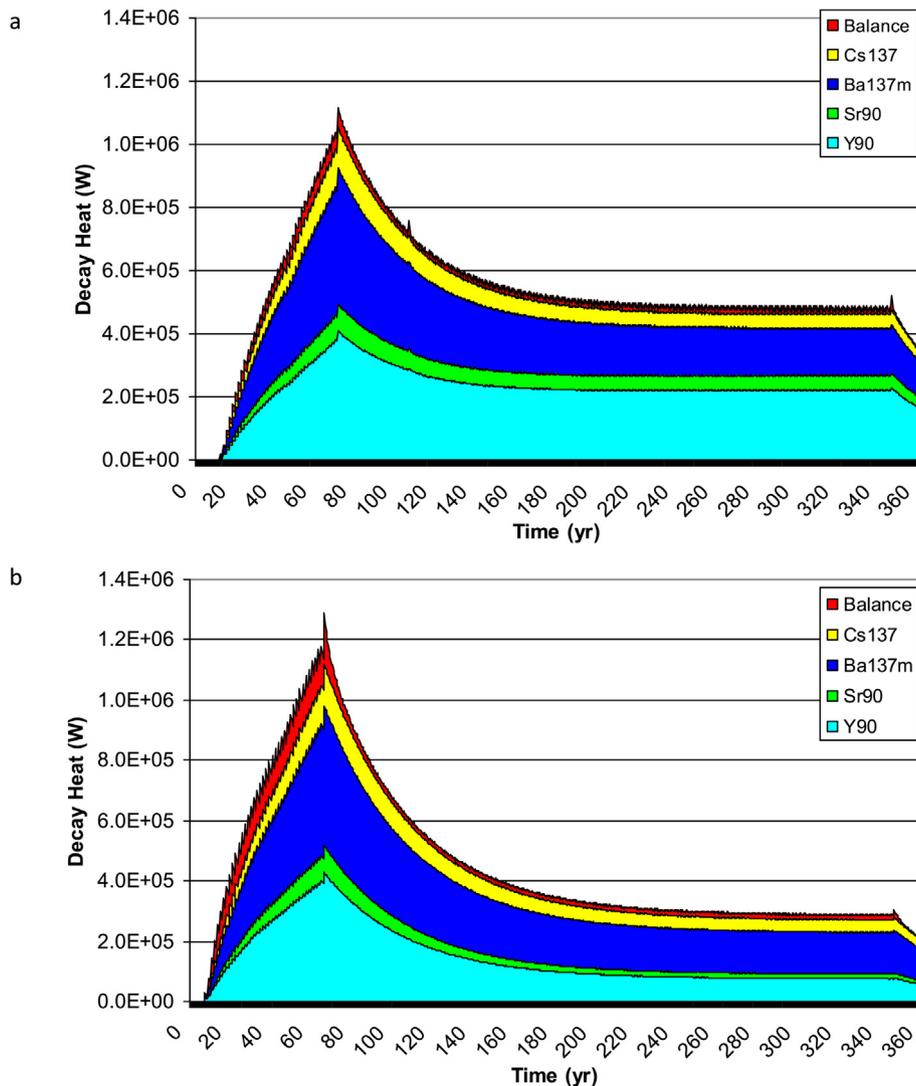


Fig. 15. Fission product decay heat for (a) Th-SFR-BE-MA5 and (b) U-SFR-BE-MA5. Th-SFR decay heat is generally substantially higher as a result of higher ^{90}Y production.

Table 4

Decay heat (MW) before and after final core discharge for Th and U SFRs with break-even fuel cycles.

SFR generations	Before		After	
	Th-SFR-BE-MA#	U-SFR-BE-MA#	Th-SFR-BE-MA#	U-SFR-BE-MA#
1	6.7	5.7	10.3	8.2
2	4.9	3.3	6.6	5.4
3	4.6	2.7	5.4	4.9
4	4.5	2.6	5.0	4.8
5	4.5	2.6	4.8	4.8

to store high-level waste before it is vitrified, giving time for the ^{90}Sr and ^{137}Cs to decay, but this is undesirable as it involves temporary storage of high-level waste as a liquid, which is more hazardous than storing the vitrified waste. In theory, it is possible to envisage mitigating this problem by separating ^{90}Sr and ^{137}Cs from the remaining waste, to be vitrified separately and stored with forced cooling for a period of ~ 100 years in a near-ground repository. This would reduce the amount of high-level waste, although the overall size and cost of the repository would be roughly the same.

After a few generations the final core decay heat becomes almost negligible, in contrast to U-SFRs where the significant TRU loading increases repository decay heat. Even so, the integrated repository decay heat is almost twice as high for the Th-SFR scenarios and the decay heat at core discharge is comparable (Table 4). In Fig. 14, the peak repository decay heat at ~ 60 years is dominated by the LEU-fuelled LWRs and is virtually identical for the Th-SFRs and U-SFRs. However, the size of the second peak (at SFR unloading) is higher for Th-SFRs for 1 or 2 generations of SFRs, after which it is lower. In reality, the relative magnitudes of the two peaks depend on relative fleet sizes of the LWRs and SFRs. In the present analysis, the LWR fleet has a larger electricity capacity than the SFR fleet, hence the first peak is larger. Recall that the SFR fleet size in this analysis is limited by TRU availability to start up the SFRs.

MA recycle is only effective at reducing the peak decay heat load compared to Pu recycle for >1 generation of SFRs. Similarly, MA recycle reduces the peak repository heat load over Pu+MA recycle for >1 generation of SFRs. Initially, the recycled MAs can increase the peak heat load. The effect of breeding ^{238}Pu from ^{237}Np is significant. Without MA recycle, the radiotoxicity of the final core is essentially insignificant for any number of generations. If reprocessing losses are increased to 1%, the repository decay heat is not significantly increased.

For the break-even fuel cycle, decay heat for RBWRs is not directly comparable to decay heat for SFRs due to the different relative sizes of the LEU-fuelled LWR and SFR/RBWR fleets. In each case, the LWR fleet size is 11.5 GWe, but this can only start up a 1.356 GWe fleet of RBWRs compared to a 4.2 GWe fleet of SFRs. The RBWR repository decay heat plotted in Fig. 16 is therefore substantially lower than for the SFRs after the initial peak as the fleet size is lower. Conversely, the decay heat per GWe capacity for RBWRs is larger than the decay heat per GWe capacity for SFRs as it contains a larger contribution from the preceding LWR fleet. However, it can be noted that the relatively low TRU burning rate in RBWRs leads to a much higher peak decay heat for 1 or 2 generations of RBWRs compared to SFRs. As before, the effect of Pa recycle is negligible.

For burner fuel cycle scenarios, the decay heat reduces with the fleet size (Lindley et al., 2014a). When the final cores are discharged, there is a peak in decay heat which again rapidly reduces as with the fleet size. This effect will be slightly more pronounced for Th fuel cycles as the TRU loading in the final core decreases.

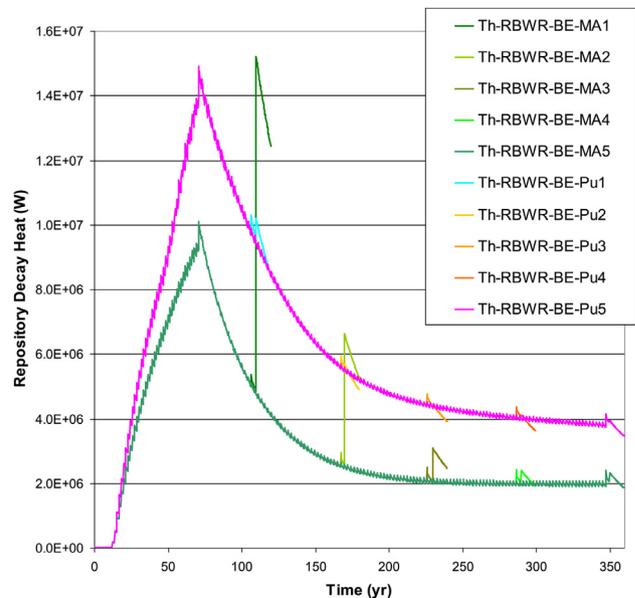


Fig. 16. Repository decay heat for break-even fuel cycle scenarios with RBWRs.

5. Conclusions

Closed Th break-even fuel cycles in SFRs or RBWRs require ~ 3 generations of operation before their waste radiotoxicity and decay heat benefits begin to approach their equilibrium performance. While this is a very long timeframe, it is substantially better than the timeframe required by other strategies. U and Th burner fuel cycles take ~ 6 generations to achieve the same result, and require a gradual phase-out of nuclear power to do so, while the U break-even fuel cycle requires over 100 generations. The 'long transition time' argument against Th fuel cycles, e.g. (Hesketh and Thomas, 2013), must be considered in conjunction with the 'long transition time' arguments against fuel cycles aimed at reducing radiotoxicity in general, e.g. (OECD, 2006).

Th break-even fuel cycles perform better from a radiotoxicity standpoint than Th burner fuel cycles, as while in both cases the waste from the TRU remaining at scenario end dominates the long-term radiotoxicity, in the former case this is normalised against the more substantial contribution of energy generated by U3. Therefore this is to some extent an argument based on how the radiotoxicity is normalised: operating a break-even fuel cycle rather than phasing out nuclear power using 'burner' reactors results in higher repository radiotoxicity in absolute terms (although this is balanced by less radiotoxicity where the Th was mined).³ The advantage of Th break-even fuel cycles is also contingent on recycling Pa, and reprocessing losses are also significant for a small number of generations due to the need to effectively burn down the TRU.

The radiotoxicity in Sv/GWe_{yr} is essentially a measure of repository loading per unit energy production, and it must be stressed that in any case a long-term geological repository will be required.

Th burner fuel cycles utilising SFRs result in lower radiotoxicity than U burner fuel cycles utilising SFRs after ~ 3 generations of SFRs. This is also contingent on Pa reprocessing, without which Th and U burner fuel cycles implemented using SFRs result in a very similar

³ Hence it could also be argued that the reference level for Th cycles should be based on the natural radiotoxicity of Th.

time to decay to the reference level. If RBWRs are utilised for the Th burner fuel cycle, at least 3 generations are required to reduce the time for the waste to decay to the reference level relative to LEU-fuelled LWRs. Beyond this, they require ~2 generations more than Th-SFRs to achieve the same radiotoxicity reduction (and provided Pa is recycled ~1 generation more than U-SFRs).

The repository integrated decay heat over the scenario time-frame is almost twice as high for SFRs operating a Th break-even fuel cycle compared to a U break-even fuel cycle as a result of much higher ^{90}Sr production, which subsequently decays into ^{90}Y . At end-of-scenario, the final core decay heat from U-SFRs increases the decay heat somewhat, while in Th-SFRs the final core is much less significant – this results in comparable peak decay heat. As decay heat at vitrification and repository decay heat affect repository sizing, this may weaken the argument for the Th cycle. This can potentially be mitigated by separation of ^{137}Cs and ^{90}Sr from the remaining waste. However, the cost of the repository is probably not sensitive to its exact size.

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List of abbreviations

EPR	European Pressurised Reactor
LEU	Low enriched uranium
LWR	Light water reactor
MA	Minor actinide
MOX	Mixed oxide fuel
PWR	Pressurised water reactor
RBWR	Reduced-moderation boiling water reactor
SFR	Sodium-cooled fast reactor
TRU	Transuranic
U3	Uranium bred from thorium, predominantly ^{233}U – ^{236}U

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