Effectiveness of Actinide Management in Fast Reactors over a Finite Recycling Period – 15123

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ABSTRACT

Long-lived nuclear waste primarily consists of transuranic (TRU) isotopes. Full recycling of 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 this paper, the fuel cycle code ORION is used to model the recycle of light water reactor (LWR)-produced TRUs in sodium-cooled fast reactors (SFRs). 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. Break-even SFRs are much less effective from a point of view of reducing waste radiotoxicity.

Thorium (Th)-burner fuel cycles are found to result in slightly lower radiotoxicity than U-burner fuel cycles for the same number of generations. Closed break-even Th fuel cycles require ~3 generations of operation before their waste radiotoxicity benefits result in decay to the reference level in ~1000 years, provided that Pa is recycled. While this is a very long timeframe, it is substantially better than the timeframe required by other strategies. The integrated decay heat over the scenario timeframe is substantially higher for break-even Th-SFRs than break-even U-SFRs as a result of much higher ^{90}Sr production. As decay heat affects repository sizing, this weakens the argument for the Th cycle.

INTRODUCTION

Spent nuclear fuel consists of uranium, fission products and transuranic (TRU) elements. While the remaining uranium is of low radiotoxicity, and fission products decay to safe levels within ~1000 years, many TRU isotopes take ~100,000 years to decay, and hence represent the long-term storage liability in a nuclear waste repository and a major political and public-perception impediment to nuclear power. Nuclear waste decay time is often measured as the time taken for the waste to decay to a 'reference level', which is typically taken as the radiotoxicity of the natural uranium (including 'daughter' isotopes produced by decay) used to fuel the reactor. Full recycling of transuranic isotopes can, in theory, lead to a reduction in repository radiotoxicity to reference levels in as little as ~500 years [1] provided reprocessing and fuel fabrication losses are limited. This strategy is utilized in many envisaged future 'sustainable' nuclear fuel cycle schemes [2,3].

Although most nuclear reactors currently operating are light water reactors (LWRs), which have a thermal neutron spectrum, fast reactors are usually considered for full recycle of TRU isotopes, as a fast neutron spectrum is beneficial for increasing the fission probability of many TRU isotopes. However, it is also possible to fully recycle TRU isotopes in LWRs, provided the LWRs are fuelled with a mixture of conventional low-enriched-uranium (LEU) fuel and TRU-bearing fuel such as mixed-oxide fuel (MOX).

TRU recycling, however implemented, requires a long-term commitment to recycling [2]. 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 [4,5].

While the heavy metal content in a 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 [6,7].

For direct disposal of waste, the radiotoxicity of the Pu dominates. However, full Pu recycle without 'minor actinide' (MA – mostly Np, Am, Cm) recycling limits the reduction in waste storage time [1]. Comparison of different partitioning and transmutation schemes, e.g. Pu-only, Pu+Am, Pu+Np, Pu+Np+Am, Pu+Np+Am+Cm, is the subject of numerous studies [8,9]. The main considerations are [8]:
- Pu-only recycle can only reduce the radiotoxicity by a factor of ~3 due to Am production.
- Np recycle, potentially performed by co-extraction with Pu [10], does not reduce the radiotoxicity until the ~1 million year mark (compared to recycle of Pu only), by which time the TRUs have decayed well below the reference level.
- Am recycle allows a reduction in radiotoxicity by a factor of ~10 over ~100–10,000 years, compared to recycle of Pu only, the effectiveness being limited by Cm production from the recycled Am.
- Am+Cm recycle allows a further reduction in radiotoxicity by 1–2 orders of magnitude over ~100–10,000 years, compared to recycle of Pu+Am, notionally allowing the radiotoxicity to decay to the reference level in <1000 years, depending on reprocessing losses.

While Np, Cm and Am all introduce fuel reprocessing, fabrication and handling challenges, this is particularly true of Cm. Hence Am-only transmutation, either homogeneously or in heterogeneous assemblies, is often considered as it is easier to implement [11]. This may be combined with homogeneous recycling of Np [12].

An attractive strategy is to burn Am in very-high-burn-up once-through moderated targets, such that the Cm is burned in situ without the need to fabricate Cm-bearing fuel. This is not considered in this study. Ref. [13] considered theoretical and computational modeling of time-dependent scenarios for accelerator-driven-system-based transmutation of a fixed initial inventory. The reactor fleet was assumed to reduce over successive generations, to burn the waste left over from the preceding generation. The findings included:
- A large number of reactor generations are necessary before the final core inventory does not. dominate the radiotoxicity, resulting in a timeframe of several hundred years for transmutation
- The radiotoxicity reduction factor became sensitive to the reprocessing losses after ~5 generations.
- Cm recycling became beneficial after ~4 generations of reactors.
- Delaying Cm recycling for ~1 generation, allowing it to decay (by α emission into isotopes of Pu), did not greatly reduce transmutation performance.

In this paper, the effectiveness Pu+MA recycling schemes are considered, allowing conclusions to be reached on the number of generations required for a scheme to deliver the claimed benefits. Scenarios consider reprocessing of TRUs produced by 'new build' LWRs, thus making them of reasonably general validity. Legacy stockpiles vary greatly between countries and in many cases may not be reprocessed [14].

Comparison is made between break-even and burner reactors, and the performance of U and Th as the fertile component of the fuel is compared. 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 [15,16]. This is only the case for a period up to ~35,000 years after which the radiotoxicity of [233]U and its daughters becomes most significant [17,18]. 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 [19,20,21]. 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 [22]. 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 [23].

The radiotoxicity beyond the shutdown of the 'final' reactors is considered. For scenarios of a few hundred years, the repository radiotoxicity (or the radiotoxicity of long-term surface storage) is also considerable. It must also be noted that the radiotoxicity, normalized in per GWeyr terms, is a somewhat abstract concept, as it is generally acknowledged [13] that a deep geological repository is necessary in any case.

SCENARIOS CONSIDERED

The fuel cycle code ORION [5] has been used to model the transition from an open (relying on standard LWR technology) to a closed fuel cycle (involving SFRs). For these scenarios, an 11.5 GWe (i.e. ten 1.15 GWe plants) 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 TRU/Pu inventory when the old cores were discharged, which may result in insufficient material to refuel the reactors. Here, this is not modeled – 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 time-step in ORION – 6 months in each case, which is in addition to the 5 years cooling time.

For burner scenarios, the ratio of LEU-fuelled reactors to SFRs and the ratio of reactors in successive generations of SFRs is constrained by the core inventories (i.e. TRU/U3[a] availability) required to start up and fuel the SFRs. In general, it is difficult or impractical to size the fleet of each successive generation of reactors such that it uses all the available TRU/U3 but does not run out of fuel. In any case, there will be out-of-core inventories at the end of scenario from recently discharged fuel which has not been reprocessed. In addition to the discharged core of the recycling reactors at the end of the scenario, this

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[a] U3 is used to refer to U bred from Th, i.e. [233-236]U.

severely limits the proportion of heavy metal which can be recycled.

For break-even 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, such that there is no unused TRU except for recently discharged fuel which has not yet been reprocessed.

0.1% reprocessing losses are assumed in the ORION models – this is a typical assumption for closed nuclear fuel cycles. In reality, reprocessing losses may be higher, with losses occurring: in the head end (where the fuel is chopped up); in the dissolver; in the aqueous or pyrochemical separation of elements; and in fabrication. Therefore the effect of 1% reprocessing losses is also discussed. For Th fuel cycles, the impact of reprocessing Pa is evaluated.

The scenarios considered are summarized in Table I.

TABLE I. Scenarios considered. 1-5 denotes that 1, 2, 3, 4 and 5 generations of reactors are considered respectively.

Scenario	Reactor	Fuel	Fuel Cycle
LEU-OT	PWR	LEU	Once-through
SFR-U-1-5	SFR	U-TRU	Burner
SFR-NoPa-1-5	SFR	Th-U3-TRU	Burner
SFR-Th-1-5	SFR	Th-Pa-U3-TRU	Burner
SFR-Iso-U-1-5	SFR	U-TRU	Break-even
SFR-Iso-NoPa-1-5	SFR	Th-U3-TRU	Break-even
SFR-Iso-Th-1-5	SFR	Th-Pa-U3-TRU	Break-even

METHOD

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 [24] library 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 II. LWR lattice calculations were performed using WIMS 10 [25]. SFR calculations were performed using ERANOS [26].

TABLE II. Reactor parameters.

Reactor	Fuel	Fuel residence time / number of batches	Discharge burn-up (GWd/t)	Power density (MWth/t)	Isotope vector used for reactor physics calculations
PWR	LEU	4.5 / 3	52	38.1	4.4 wt% LEU
SFR burner	U-TRU oxide	3 / 3	113.6	114.6	Isotope vector from equilibrium study [27]
Break-even SFR	U-TRU-Zr	3 / 3 (seed) 6 / 3 (blanket) [a]	65.5 (seed) 14.0 (blanket)	70.3 (seed) 7.5 (blanket)	
SFR burner	Th-(Pa)-U3-TRU oxide	3 / 3	97.1	104.2	
Break-even SFR	Th-(Pa)-U3-TRU nitride	3 / 3 (seed) 6 / 3 (blanket) [a]	73.7 (seed) 5.0 (blanket)	79.2 (seed) 2.7 (blanket)	

a In reality, the axial blanket will reside in the core for the same length of time as the seed, i.e. 3 years. This approximation makes very little difference to the ORION calculations and simplifies the model, as having fuel elements operate with different batch strategies requires defining two reactors in the model.

A 1000 MWth SFR is considered based on the Advanced Recycling Reactor [28] with 3 batches and a 1 year cycle length. For the burner, the SFR TRU loading is 44.9% and 44.2% for U and Th fuel respectively. This leads to a TRU incineration rate of ~17.8% and ~16% per pass respectively, corresponding to ~249 kg/GWthyr and ~273 kg/GWthyr respectively. For the U and Th break-even SFRs, the seed has 18.7% TRU loading and 25.9% TRU+U3 loading respectively. The core configurations are shown in Fig. 1.
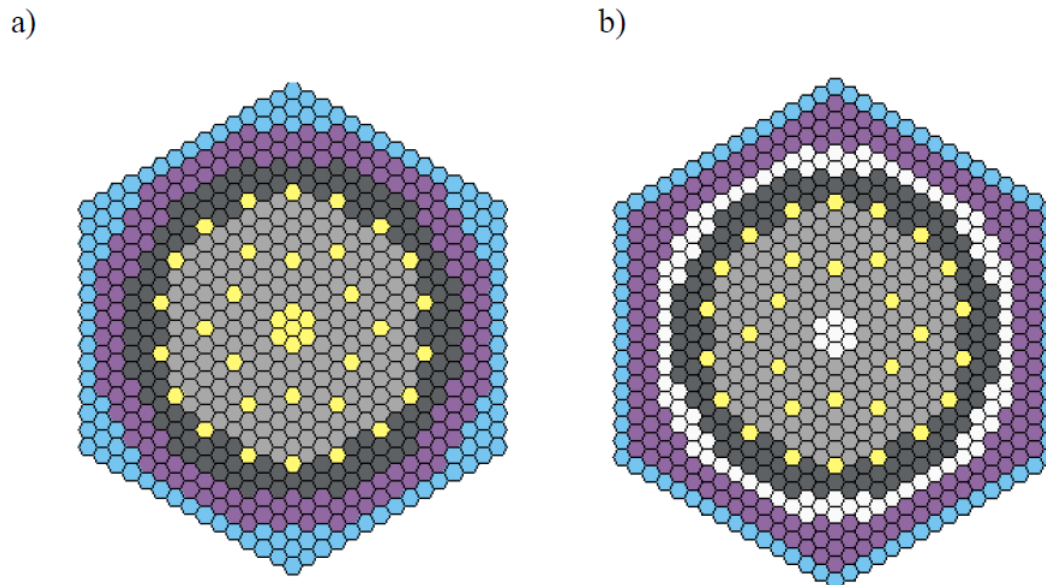

Fig. 1. SFR core layouts for burner (a) and break-even (b) designs. Light grey = inner core, dark grey = outer core; yellow = control rods; violet = steel shield; blue = B4C shield; white = blanket.

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 SFR burner for U cycles used in this study is shown in Fig. 2. For Th scenarios, Th recovered from reactors is cooled for a further 20 years before fuel fabrication to allow ^{228}Th and its daughters (notably high-energy gamma sources ^{208}Tl and ^{212}Bi) to decay. ^{228}Th is produced by ^{232}U decay, and these have half-lives of 1.9 and 69 years respectively. The ^{232}U in the U3 will decay into ^{228}Th 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. This is shown in Fig. 3.

For the break-even SFR scenarios, the SFR core and blanket were modeled separately, with different 'reactors' and cross-sections. The blanket was fuelled exclusively with reprocessed U or Th. For the burner scenarios, the ratio of LEU-fuelled PWRs and SFRs in each generation is limited by TRU availability. The limiting point for the first generation of SFRs is reactor start-up (in Year 41). For
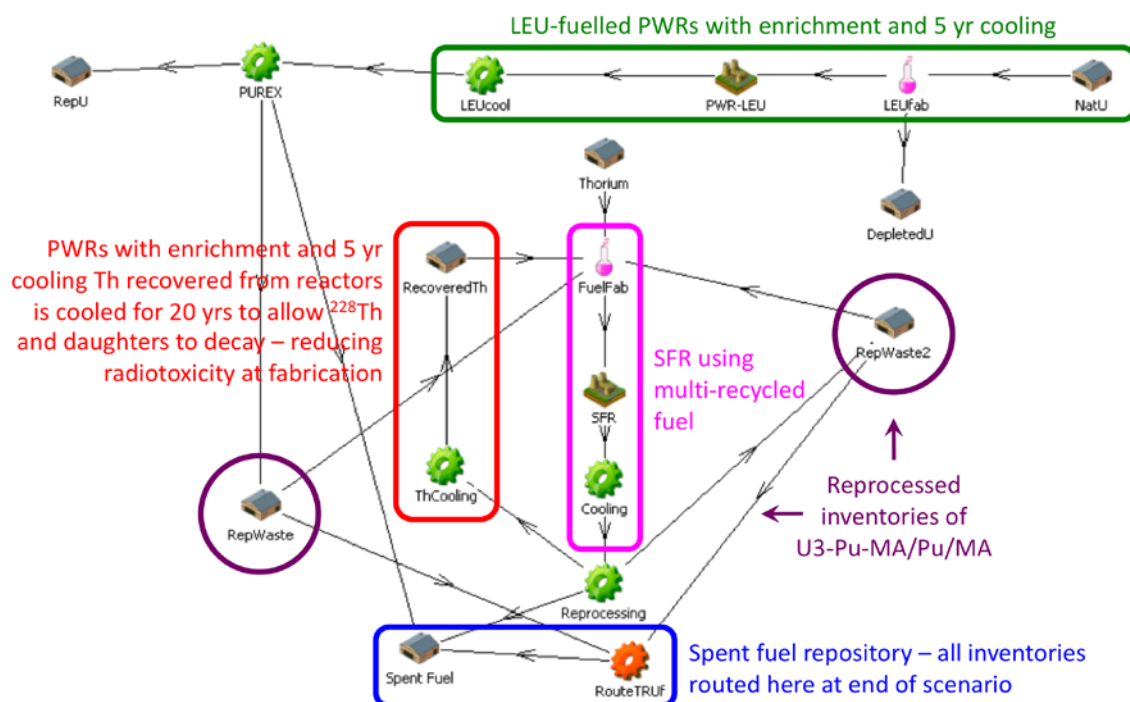
subsequent generations, the discharged cores from the previous generation are burned in a progressively smaller fleet of reactors. Each generation is smaller than the last, meaning that not all of the discharged core is loaded into the fresh core. The remainder of material from the discharged core is then used to provide fuel for the subsequent generation over its lifetime. The SFR 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 will increase the SFR capacity beyond that considered for the scenario. The number of reactors in each generation is shown in Table III.

Fig. 2. ORION fuel cycle scenario model (U scenario).

Fig. 3. ORION fuel cycle scenario model (Th scenario).

TABLE III. Scenario reactor capacities.

Reactor Generation	Starting Year	Capacity (GWe)	
		U	Th
LEU-PWR	1	11.50	11.50
SFR Generation 1	41	2.940	2.730
SFR Generation 2	101	1.470	1.470
SFR Generation 3	161	0.840	0.630
SFR Generation 4	221	0.420	0.315
SFR Generation 5	281	0.315	0.158

A reference level radiotoxicity is adopted (as considered, for example, in Ref. [2]), 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 Pressurized Reactor (EPR) as the reference 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. Fission products are included in radiotoxicity and decay heat calculations.

RESULTS

Burner fuel cycles
The radiotoxicity over 5 generations of U-SFR burners is plotted in Fig. 4. 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 therefore also relevant as the fission

products will be vitrified long before Year 1 in Fig. 4. 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.
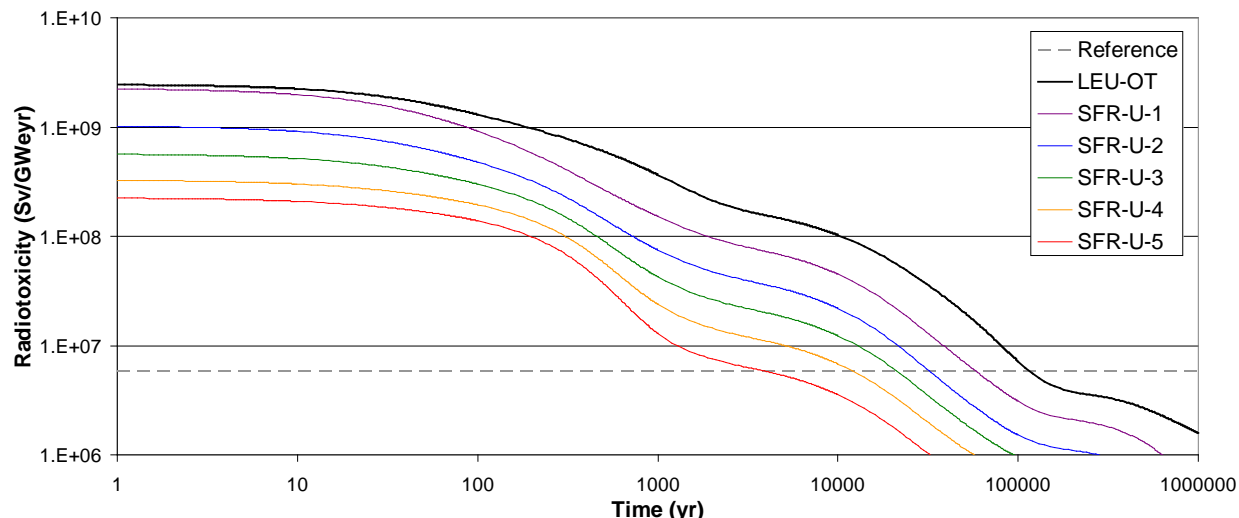

Fig. 4. Repository radiotoxicity for U-SFR burner scenarios.

Results for Th-SFR burners are shown in Fig. 5. Scenarios with and without Pa recycling are presented with solid and dashed lines respectively. 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.
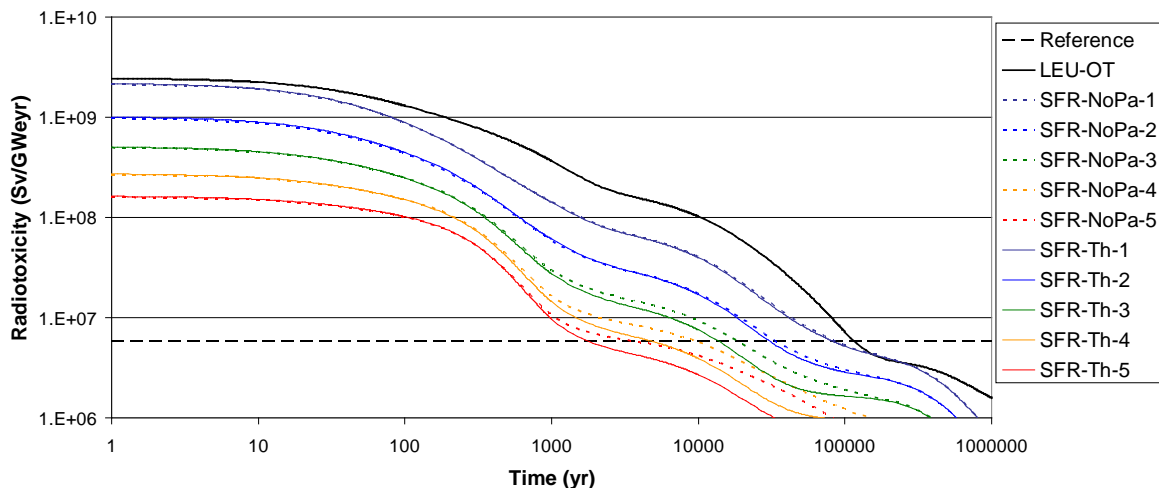
Fig. 5. Repository radiotoxicity for Th-SFR burner scenarios.

The time for waste to decay to the reference level is shown in Fig. 6. 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.

In each generation, the mass of TRU remaining roughly halves, and the time taken for the repository radiotoxicity to reduce to the reference level also roughly halves. After a few generations, the actinide

isotope vector converges such that the radiotoxicity is essentially proportional to the TRU mass. The ORION scenarios give an SFR fleet size that roughly halves each generation. Assuming the radiotoxicity is a constant function of TRU mass, it is possible to derive the TRU mass and therefore radiotoxicity as a function of the number of SFR generations. It is found that both U-SFRs and Th-SFRs require around ~7 generations of operation in order for the waste to decay to the reference level within 1000 years. 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 as radiotoxicity is dominated by the final cores of the LWRs.
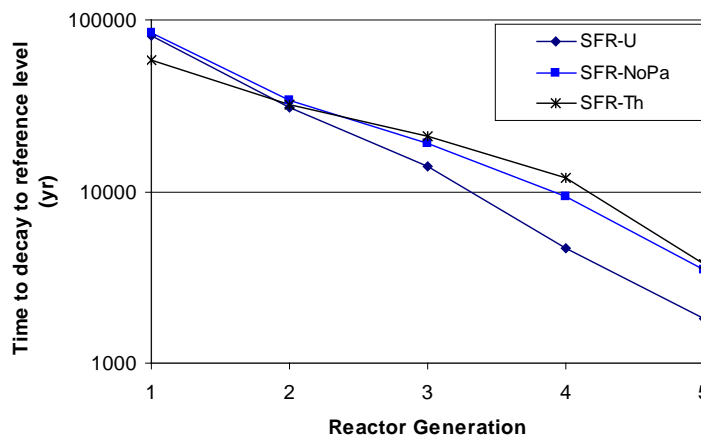

Fig. 6. Comparison of repository decay times for burner scenarios.

Breeder fuel cycles

Break-even SFRs result in a much lower reduction in radiotoxicity as they do not reduce the TRU inventory, and this is not compensated for by the stabilization of the TRU inventory over a long electricity generation period (Fig. 7). Radiotoxicity of SFR-Iso-MA5 is ~26 times the reference level after 1000 years. Therefore, the scenario would have to be ~26 times longer for the energy generated by the reactors to be sufficient for the material to decay to the reference level within 1000 years (without accounting for reprocessing losses). This timeframe can be reduced by reducing the out-of-core inventory of the reactor (i.e. by reducing the cooling time).
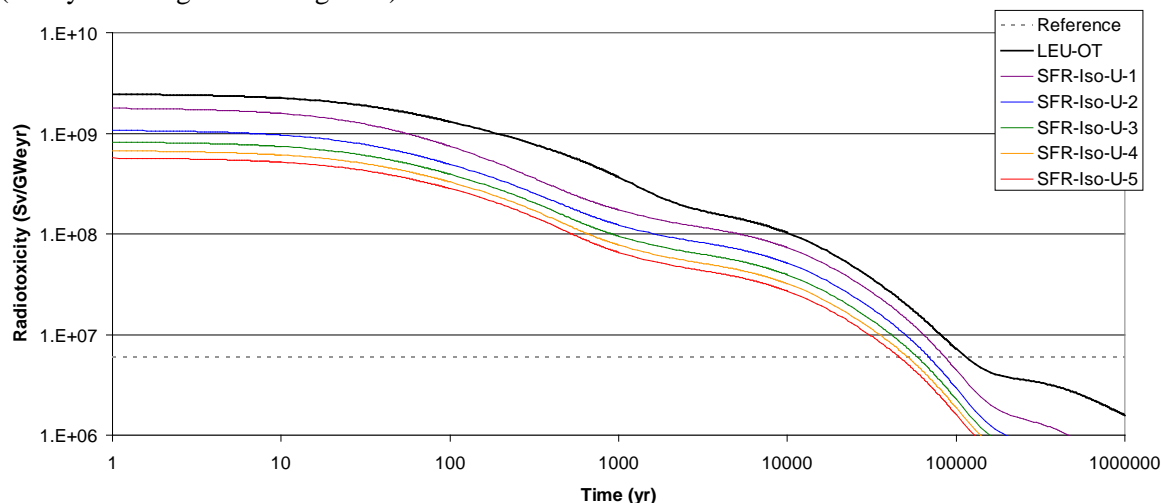

Fig. 7. Repository radiotoxicity for break-even U-SFR scenarios.

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However, for the Th fuel cycle, with Pa recycle, the time for the waste to decay to the reference level drops to ~1400 years within 3 generations (Fig. 8). 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 SFR-Iso-NoPa-5 are shown in Fig. 9. The [231]Pa + [227]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.
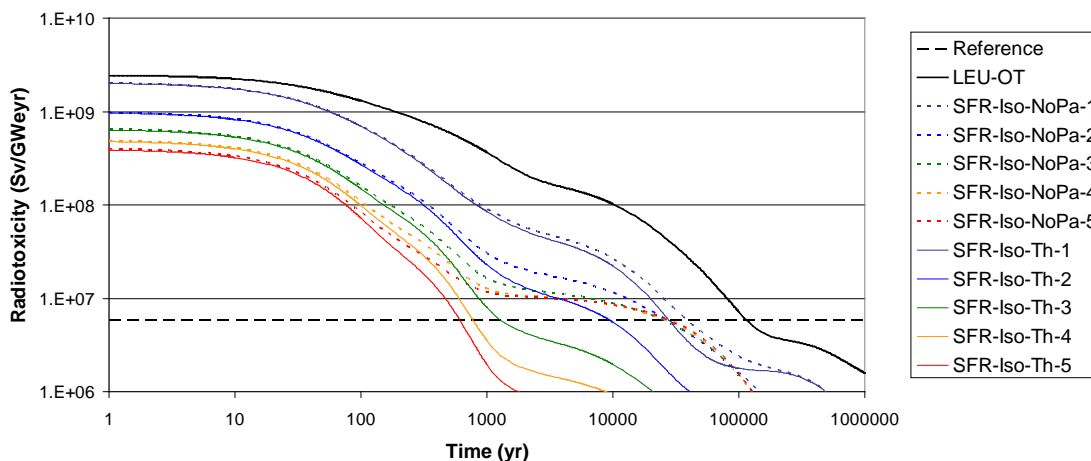

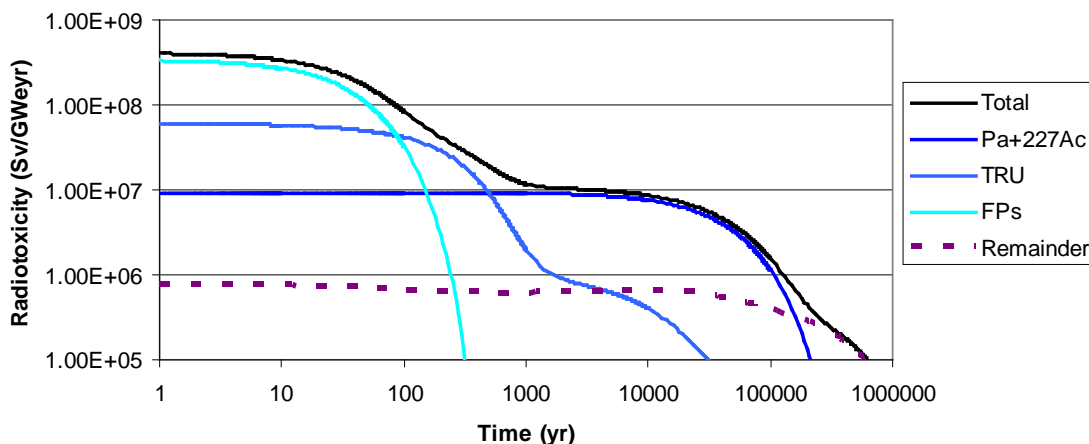Fig. 8. Repository radiotoxicity for break-even Th-SFR scenarios.

Fig. 9. Contributions to radiotoxicity of SFR-Iso-NoPa-5.

Decay heat
Recycling of Pu and MAs can also reduce the peak and integrated heat load in the repository [5]. In this section, the decay heat for break-even scenarios is investigated, to derive general conclusions on the relative behaviour of Th and U fuel cycles. For Th cycles, the decay heat is not sensitive to whether Pa is recycled as the [232]U contribution is relatively small. 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 much higher ^{90}Y production (a decay product from ^{90}Sr, which is produced in greater quantities in the Th-

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SFR) (Fig. 10). The initial peak in Fig. 10 occurs towards the end of the lifetime of the LWRs, after which the recycling reactors result in repository decay heat stabilizing at a lower level.
After a few generations the final core decay heat for Th-SFRs 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 higher for the Th-SFR scenarios and the decay heat at core discharge is comparable (Table IV).

TABLE IV. Decay heat (MW) before and after final core discharge for Th and U break-even cores.

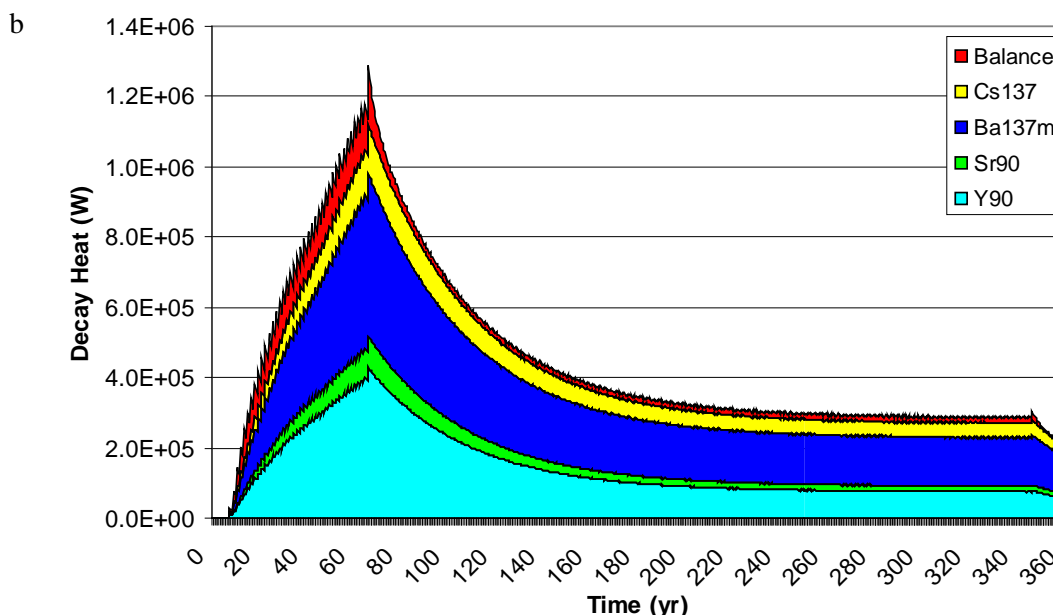| SFR Generations	Before		After	
	Th-SFR	U-SFR	Th-SFR	U-SFR
1	6.7	5.7	10.3	8.2
2	4.9	3.3	6.6	5.4
3	4.6	2.7	5.4	4.9
4	4.5	2.6	5.0	4.8
5	4.5	2.6	4.8	4.8
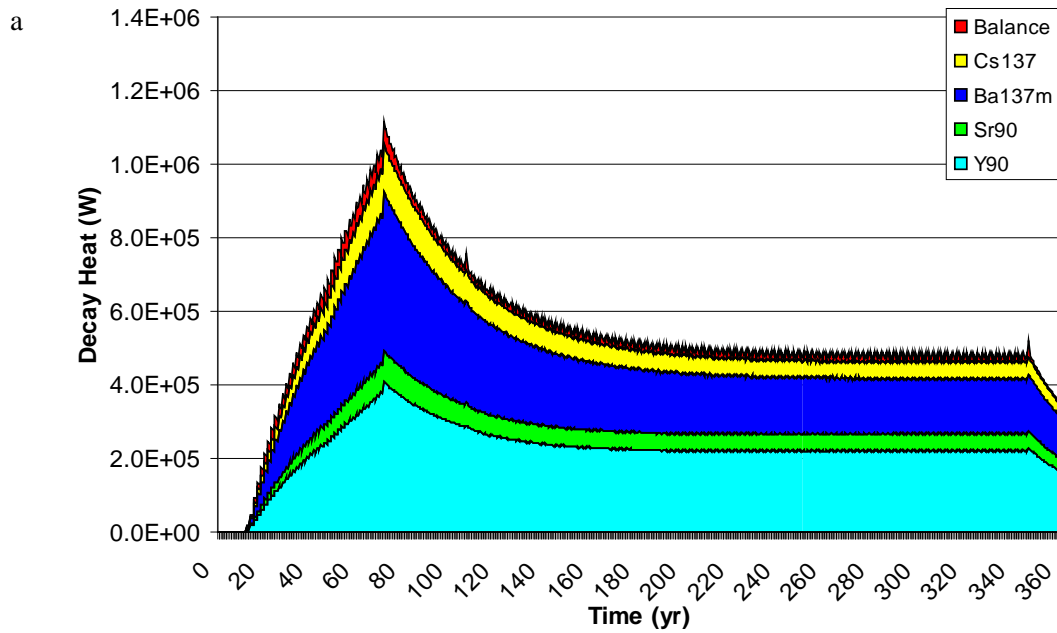
Fig. 10. Fission product decay heat for (a) SFR-Iso-Th-5 and (b) SFR-Iso-U-5. Th-SFR decay heat is generally substantially higher as a result of higher ^{90}Y production.

CONCLUSIONS

To achieve a repository radiotoxicity reduction approaching that achievable at equilibrium with the U fuel cycle, ~6 generations of SFRs are required to recycle the TRUs produced by LWRs. The fleet size must exponentially decay over a timeframe of several hundred years in a gradual phase-out of nuclear power. Otherwise, repository radiotoxicity is dominated by the final core inventory. This appears challenging from economic and energy security standpoints. TRU recycle in break-even SFRs is much less effective from a point of view of reducing waste radiotoxicity, although still effective from the point of view of reducing repository decay heat. Th-SFR burners result in slightly reduced radiotoxicity compared to U-SFR burners, provided that Pa is recycled.

In contrast, closed break-even Th fuel cycles in SFRs require only ~3 generations of operation before their waste radiotoxicity and decay heat benefits begin to approach their equilibrium performance, and a phase-out of nuclear power is not required. While this is a very long timeframe, it is substantially better than the timeframe required by other strategies. The 'long transition time' argument against Th fuel cycles (e.g. Ref. [19]) must be considered in conjunction with the 'long transition time' arguments against fuel cycles aimed at reducing radiotoxicity in general (e.g. Ref. [13]). Break-even Th fuel cycles perform better from a radiotoxicity standpoint than burner Th 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 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). The advantage of break-even Th 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 repository integrated decay heat over the scenario timeframe is substantially higher for break-even

Th-SFRs than break-even U-SFRs 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.

It must be emphasized that 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.

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