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

Benjamin A. Lindley [a, *], Carlo Fiorina [b], Robert Gregg [c], Fausto Franceschini [d], Geoffrey T. Parks [a]

[a] Department of Engineering, University of Cambridge, Cambridge, CB2 1PZ, UK
[b] Paul Scherrer Institut, Nuclear Energy and Safety, Laboratory for Reactor Physics and Systems Behaviour, Villigen PSI, Switzerland
[c] United Kingdom National Nuclear Laboratory, Springfield Works, Preston, PR4 0XJ, UK
[d] Westinghouse Electric Company LLC, Cranberry Township, PA, USA

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ABSTRACT

Disposal of spent nuclear fuel is a major political and public-perception problem for nuclear energy. From a radiological standpoint, the long-lived component of spent nuclear fuel primarily consists of transuranic (TRU) isotopes. Full recycling of TRU isotopes can, in theory, lead to a reduction in repository radiotoxicity to reference levels corresponds to the radiotoxicity of the unburned natural U required to fuel a conventional LWR in as little as ~500 years provided reprocessing and fuel fabrication losses are limited. This strategy forms part of many envisaged 'sustainable' nuclear fuel cycles. 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. The importance of low reprocessing losses and minor actinide (MA) recycling is also dependent on the timeframe during which actinides are recycled. In this paper, the fuel cycle code ORION is used to model the recycle of light water reactor (LWR)-produced TRUs in LWRs and sodium-cooled fast reactors (SFRs) over 1–5 generations of reactors, which is sufficient to infer general conclusions for higher numbers of generations. Here, a generation is defined as a fleet of reactors operating for 60 years, before being retired and potentially replaced. Over up to ~5 generations of full actinide recycle in SFR burners, the final core inventory tends to dominate over reprocessing losses, beyond which the radiotoxicity rapidly becomes sensitive to reprocessing losses. For a single generation of SFRs, there is little or no advantage to recycling MAs. However, for multiple generations, the reduction in repository radiotoxicity is severely limited without MA recycling, and repository radiotoxicity converges on equilibrium after around 3 generations of 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 – which appears challenging from an economic and energy security standpoint. TRU recycle in pressurized water reactors (PWRs) with zero net actinide production provides similar performance to low-enriched-uranium (LEU)-fueled LWRs in equilibrium with a fleet of burner SFRs. However, it is not possible to reduce the TRU inventory over multiple generations of PWRs. TRU recycle in break-even SFRs is much less effective from a point of view of reducing spent nuclear fuel radiotoxicity.

1. 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 (World Nuclear Association, 2014; IAEA, 2004) and hence represent the

List of abbreviations: CORAIL, LWR fuel assembly containing U-TRU fuel pins and LEU pins; EPR, European pressurized reactor; LEU, low enriched uranium; LWR, light water reactor; MA, minor actinide; MOX, mixed oxide fuel; PWR, pressurized water reactor; SFR, sodium-cooled fast reactor; TRU, transuranic.

* Corresponding author.
E-mail addresses: bal29@cam.ac.uk (B.A. Lindley), carlo.fiorina@psi.ch (C. Fiorina), robert.wh.gregg@nnl.co.uk (R. Gregg), francef@westinghouse.com (F. Franceschini), gtp10@cam.ac.uk (G.T. Parks).

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long-term storage liability in a nuclear waste repository and a major political and public-perception aversion towards nuclear power. Spent nuclear fuel decay time is often measured as the time taken for the spent nuclear fuel 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 (Grouiller et al., 2002) provided reprocessing and fuel fabrication losses are limited. This strategy is utilized in many envisaged future 'sustainable' nuclear fuel cycle schemes (OECD (Organisation for Economic Cooperation and Development) Nuclear Energy Agency, 2002; Generation and International Forum, 2002).

Most nuclear reactors currently operating are light water reactors (LWRs), which have a thermal neutron spectrum. However, 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 fueled with a mixture of conventional low-enriched-uranium (LEU) fuel and TRU-bearing fuel such as mixed-oxide fuel (MOX).

However, TRU recycling requires a long-term commitment to recycling (OECD (Organisation for Economic Cooperation and Development) 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, 1280; 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).

For direct disposal of spent nuclear fuel, the radiotoxicity of the Pu dominates. However, full Pu recycle without 'minor actinide' (MA – mostly Np, Am, Cm) recycling limits the reduction in spent nuclear fuel storage time (Grouiller et al., 2002). 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 (Delpech et al., 1998; Magill et al., 2003). The main considerations are (Lalieux et al., 2012):

- 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 (IAEA, 2008), 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 additional 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 (Varaine et al., 2010). This may be combined with homogeneous recycling of Np (Bonnerot et al., 2010).

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 (Pilate et al., 2000). This is not considered in this study.

Ref. OECD Nuclear Energy Agency (2006) 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 (reactors were assumed to have a lifetime of 60 years, before being shutdown and replaced with a new 'generation' of reactors), to burn the spent nuclear fuel 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, Pu + Am and Pu + MA recycling schemes are compared, allowing conclusions to be reached on the number of generations required for a scheme to deliver the claimed benefits. This paper primarily considers 'burner' scenarios where sodium-cooled fast reactors (SFRs) support a fleet of LWRs. The continued operation of LWRs is also considered, which allows comparison of scenarios with and without fleet reduction. Over hundreds of years, this seems questionable (in reality eventual deployment of 'fast breeders' is expected as U reserves are exhausted), but from this it can be inferred that some of the 'equilibrium' burner scenarios are themselves unlikely due to the timescales involved. 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 (IAEA, 2005).

Comparison is also made with 'break-even' SFR scenarios, where the SFRs operate in a self-sustaining mode where they produce as much fissile fuel as they consume, allowing a constant fleet size to be maintained with full recycle of TRUs during the scenario. 'CORAIL' scenarios are also considered, where LWRs operate with zero net Pu/TRU production by using a mix of LEU and MOX fuel (Kim et al., 2002).

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 time for the radiotoxicity to reduce to the reference level, when normalizing the radiotoxicity in per GWeyr terms and defining a reference level based on natural U ore, does not necessarily reflect the timeframe over which the repository represents a radiological hazard. If more electricity is generated per unit repository radiotoxicity, this leads to lower repository loading relative to nuclear generating capacity, but the radiological hazard of a given repository is related to the absolute radiotoxicity rather than the radiotoxicity normalized by electricity production. As discussed, radiotoxicity is not the only measure of radiological hazard, and the radiological hazard is likely to be non-negligible even after the repository radiotoxicity has reduced below the reference level.

Also, it is generally acknowledged (OECD Nuclear Energy Agency, 2006) that a deep geological repository is necessary in

any case.

Part I of this paper considers uranium fuel cycles. In Part II (Lindley et al., 2014), the thorium fuel cycle is investigated and compared to the results in this paper.

2. Scenarios considered

The fuel cycle code ORION (Gregg and Hesketh, 2013) has been used to model the transition from an open (relying on standard LWR technology) to a closed fuel cycle (involving SFRs or LWRs). For these scenarios, an 11.5 GWe (i.e. ten 1.15 GWe plants) fleet of LEU-fueled 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-fueled 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. Five 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 – six months in each case, which is in addition to the five years cooling time.

For burner scenarios, the ratio of LEU-fueled reactors to SFRs and the ratio of reactors in successive generations of SFRs is constrained by the core inventories (i.e. TRU 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 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 severely limits the proportion of heavy metal which can be recycled.

For break-even and CORAIL scenarios, the net Pu/TRU production is zero once the LEU-fueled LWRs go offline. Here, the unused TRU from the LEU-fueled 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. In particular: LWRs can be only part-loaded with CORAIL fuel assemblies (with the remainder being LEU assemblies) if insufficient TRU is available at any step to fuel the reactors, hence it is relatively easy to ensure the TRU is efficiently used.

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 aqueous or pyrochemical separation of elements; and in fabrication. Therefore the effect of 1% reprocessing losses is also discussed.

The scenarios considered are summarized in Table 1.

Table 1
Scenarios considered. # 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-Bu-MA#	SFR	U–Pu–MA	Burner
SFR-Bu-Am#	SFR	U–Pu–Am	Burner
SFR-Bu-Pu#	SFR	U–Pu	Burner
SFR-BE-MA#	SFR	U–Pu–MA	Break-even
SFR-BE-Pu#	SFR	U–Pu	Break-even
CORAIL-MA#	PWR	LEU/U–Pu–MA	Zero net TRU
CORAIL-Pu#	PWR	LEU/U–Pu	Zero net TRU

3. 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 (ANSWERS, 2013) 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 2.

A 1000 MWth SFR is considered based on the Advanced Recycling Reactor (Dobson, 2008) with three batches and a one year cycle length. For the burner, the SFR TRU loading is 44.9% and 38.1% with and without MAs respectively. This leads to a TRU incineration rate of ~17.8% per pass in both cases, corresponding to ~249 kg/GWthyr with MAs, ~212 kg/GWthyr without MAs. The break-even SFR uses metallic fuel, with 18.7% and 16.9% TRU loading with and without MAs respectively. The core configurations are shown in Fig. 1.

Four-loop Westinghouse PWRs are considered in all cases. CORAIL-Pu and CORAIL-MA are based on designs considered in Ref. (Kim et al., 2002). These are heterogeneous assemblies containing a mixture of ~1/3 U-TRU and ~2/3 LEU pins. The CORAIL-Pu design uses the same pin diameter as a normal PWR, while the CORAIL-MA design utilizes a high moderation lattice to limit the equilibrium MA fraction in the pins. The fuel assembly designs are shown in Fig. 2. In this study, the CORAIL-Pu design utilized a Pu loading of 9.05% in the U–Pu pins, and the CORAIL-MA design utilized a TRU loading of 13% in the U-TRU pins, to give zero net TRU production in both cases. These are greater than the values of 8.45% and 10.56% found appropriate in Ref. (Kim et al., 2002).

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 used in this study 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 fueled exclusively with reprocessed U. Similarly, the U-TRU and LEU portions of the CORAIL LWR were modeled as separate 'reactors' in ORION, with separate cross-section libraries.

For the burner scenarios, the ratio of LEU-fueled 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

Table 2
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	Reactor physics method
PWR	LEU	4.5/3	52	38.1	4.4 wt% LEU	WIMS10 lattice calculation
PWR (CORAIL)	U–Pu–(MA) oxide, LEU	3/3	45	38.6 (U–Pu) 42.7 (U–Pu–MA)	4.62 wt% LEU (U–Pu)/5.11 wt% LEU (U–Pu–MA); Equilibrium TRU isotope vector from (Kim et al., 2002)	(Newton et al., 2008)
SFR burner	U–Pu–(MA) oxide	3/3	113.6	114.6	Isotope vector from equilibrium study (Fiorina et al., 2013)	ERANOS core calculation (Rimpault et al., 2002)
Break-even SFR	U–Pu–(MA)–Zr	3/3 (seed) 6/3 (blanket)[a]	65.5 (seed) 14.0 (blanket)	70.3 (core) 7.5 (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.

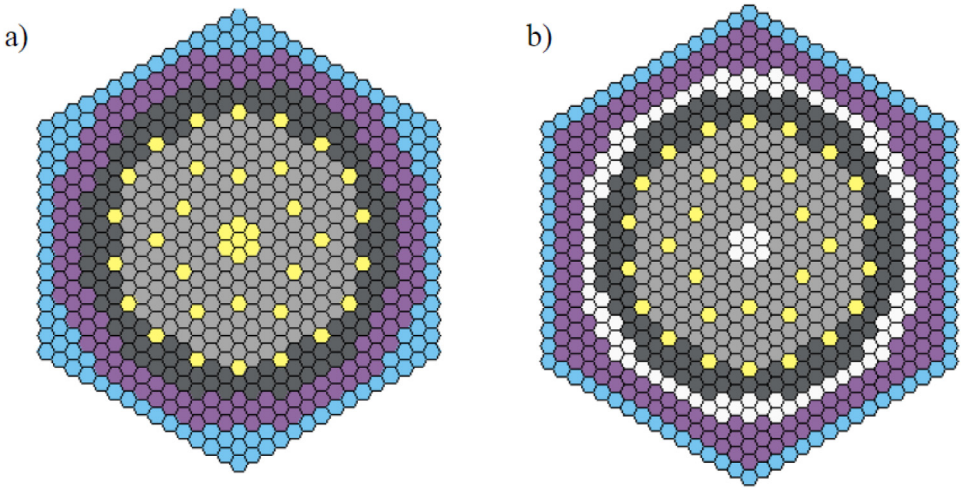

Fig. 1. SFR core layouts for burner (a) and break-even (b) designs. Light grey = inner core, dark grey = outer core; yellow = control rods; violet = steel shield; blue = B_4C shield; white = blanket. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

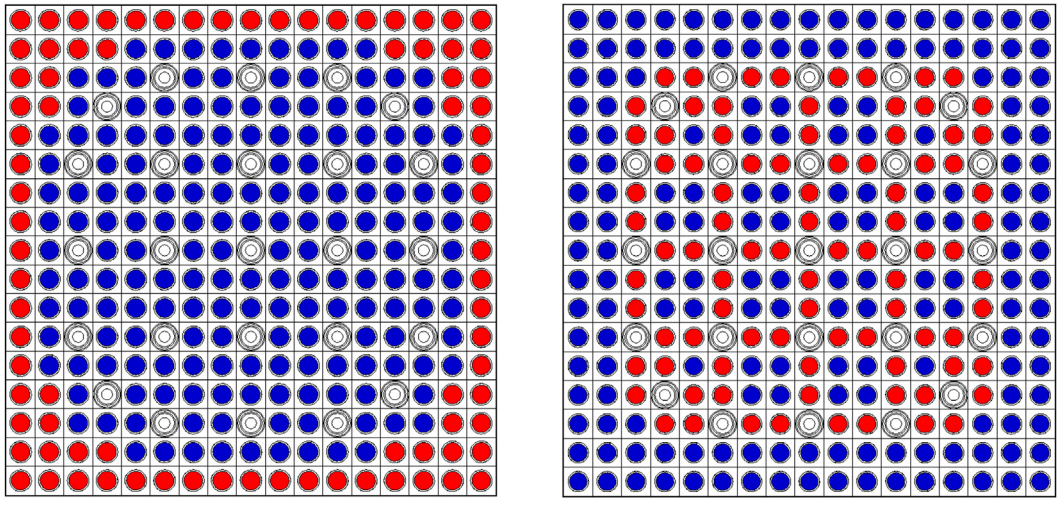

Fig. 2. CORAIL assemblies with LEU pins (blue) and U–Pu/U–Pu–MA pins (red). Left: CORAIL-Pu assembly; Right: CORAIL-TRU assembly. 1/8th of the fuel assembly is shown in each case. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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 capacity for reactors of each type in each generation is shown in Table 3.

The resulting TRU inventory for SFR-Bu-MA5 is shown in Fig. 4. The TRU accumulated from the LEU-fueled PWRs is used to start SFRs after 40 years. The TRU inventory increases after start-up due

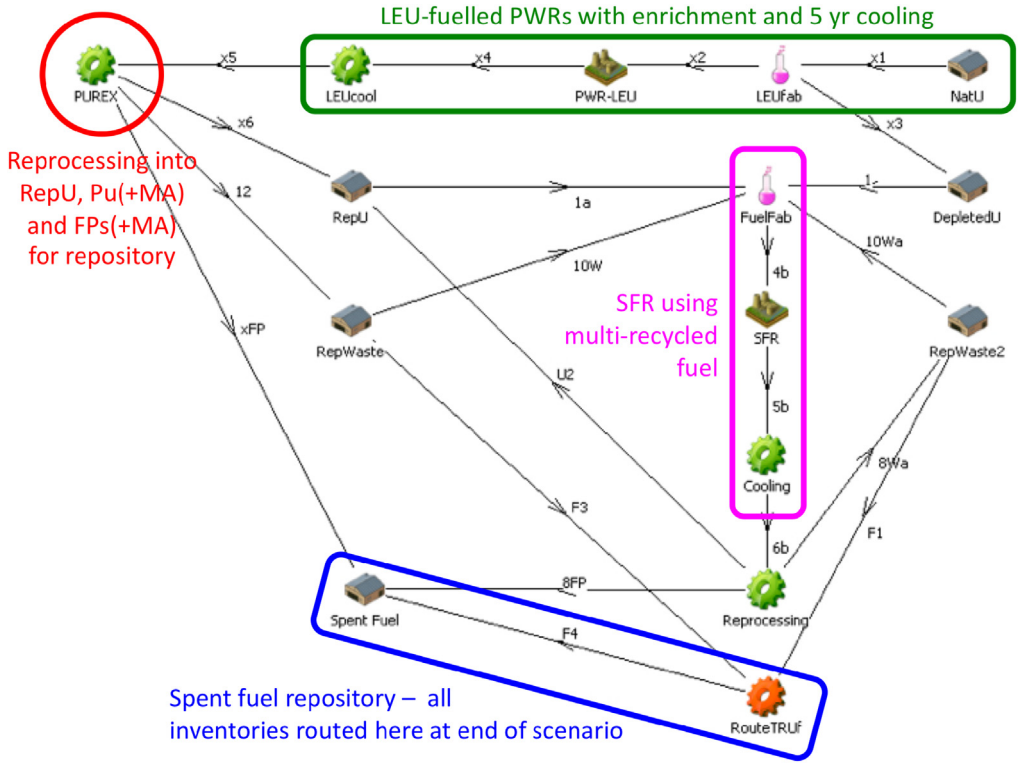

Fig. 3. ORION fuel cycle scenario model.

Table 3
Scenario reactor capacities.

Reactor generation	Starting year	Capacity (GWe)		
		SFR-Bu-MA	SFR-Bu-Am	SFR-Bu-Pu
LEU-PWR	1	11.50	11.50	11.50
SFR Generation 1	41	2.940	2.940	2.730
SFR Generation 2	101	1.470	1.470	1.155
SFR Generation 3	161	0.840	0.735	0.525
SFR Generation 4	221	0.420	0.315	0.268
SFR Generation 5	281	0.315	0.158	0.134

to continued operation of LEU-fueled PWRs. From 60 years onwards, no further TRU is produced by the LEU-fueled PWRs and the inventory decreases. After 100, 160, 220 and 280 years, unloading of one generation of SFRs provides inventory for the next generation. The capacity in GWe of each generation is roughly half the size of the preceding one.

The effect of having subsequent generations of LWRs on the TRU inventory is illustrated in Fig. 5. Here, SFR-Bu-MA5 is added to SFR-Bu-MA4 (delayed by 60 years), SFR-Bu-MA3 (delayed by 120 years), SFR-Bu-MA2 (delayed by 180 years) and SFR-Bu-MA1 (delayed by 240 years). Unless stated, the results presented here, e.g. for SFR-Bu-MA5, do not consider the subsequent generations of LWRs.

A reference level radiotoxicity is adopted (as considered, for example, in Ref. (OECD (Organisation for Economic Cooperation and Development) Nuclear Energy Agency, 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 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.

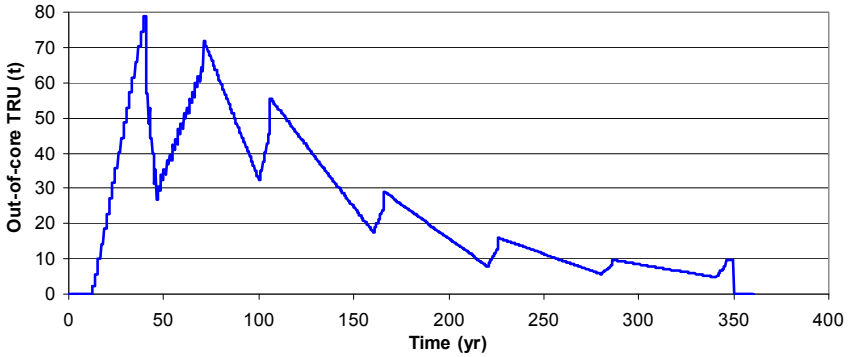

Fig. 4. TRU inventory for SFR-Bu-MA5.

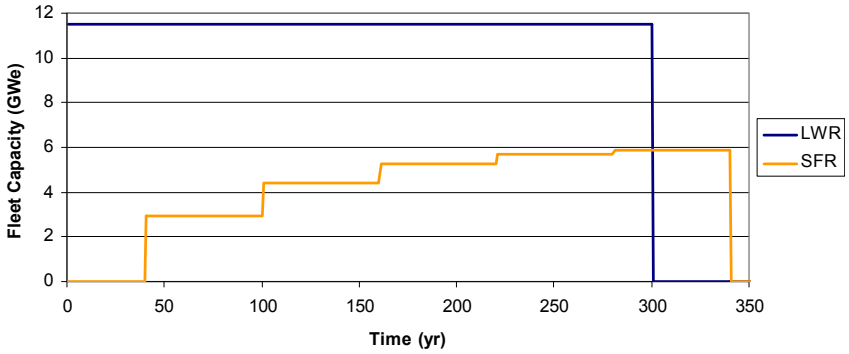

Fig. 5. Fleet capacity with 5 generations of LWRs (followed by TRU burning in SFRs), corresponding to sum of SFR-Bu-MA1–5 prior to reactor switch-off.

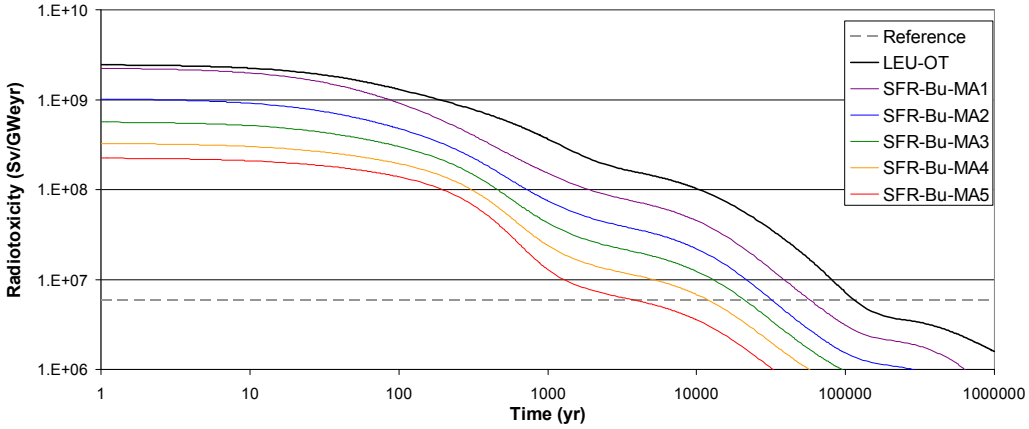

Fig. 6. Repository radiotoxicity for scenarios with MA recycling.

4. Results

4.1. Radiotoxicity with Pu + Am + Np + Cm recycle

The radiotoxicity over 5 generations of SFR burners 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 therefore 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.

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 radiotoxicity curve is non-linear, such that the time taken for the spent nuclear fuel to decay to the reference level is a non-linear function of TRU mass (Fig. 7). However, rough proportionality is still satisfied.

The ORION scenarios give a fleet size that roughly halves each generation. Assuming the radiotoxicity is a constant function of TRU mass, as in Fig. 8 (derived for 5 generations of SFRs in ORION), it is possible to derive the TRU mass and therefore radiotoxicity as a general function of: the number of SFR generations; reprocessing losses; cooling, reprocessing and fabrication times; and TRU utilization efficiency. To allow general conclusions to be drawn

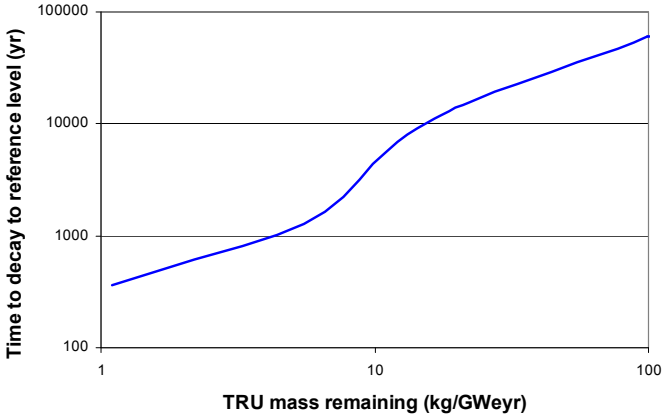

Fig. 7. Repository timeframe as a function of TRU mass.

from the calculations performed and limit computational overhead, it was assumed that the number of SFRs for generations 6–10 is half the number in the immediately preceding generation, which is a slight approximation – this is further discussed below. The time to decay to the reference level under these assumptions is shown in Fig. 8. Reprocessing losses and final core inventory are loaded into the repository at different times, but this is not distinguished here.

The fleet sizes in the ORION model are not optimized, i.e. the TRU is not utilized with 100% efficiency (~20–30% of the final TRU is

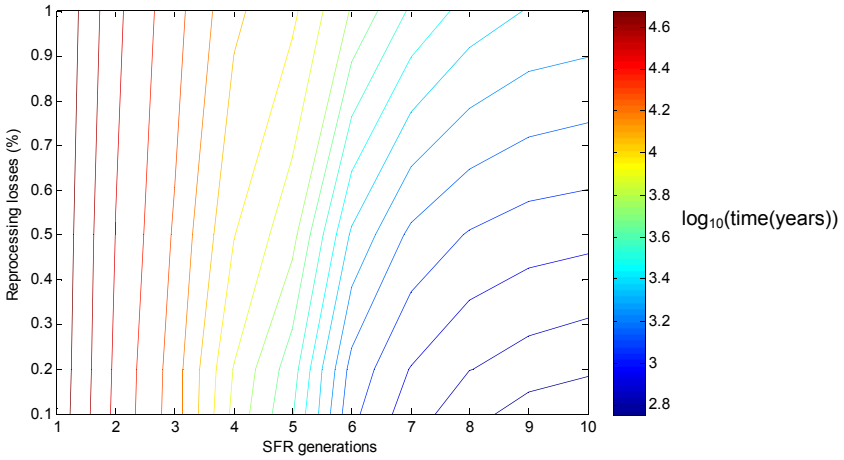

Fig. 8. $\log_{10}$ (time to decay to reference level) as a function of reprocessing losses and number of SFR generations, with 5.75 years out-of-core time.

not from the final core or the final out-of-core inventory). Note in particular that the TRU is utilized more efficiently in generation 5 than generation 4 which distorts Fig. 8. It is difficult to achieve 100% efficiency as the number of reactors of each generation much be exactly defined, such that all the TRU is either in the core or in cooling at end of life. In principle, if the TRU inventory is twice the minimum, then this corresponds to a loss of one reactor generation. The assumption that the fleet size successively halves for each generation after generation 5 is consistent with the observed trend from the ORION models reported in Table 3, but is an approximation as the exact size of each generation will depend on how efficiently the TRU can be utilized.

At least 7 generations of SFRs are required for the TRU to decay to the reference level within 1000 years. If out-of-core time is reduced to 1 year, then the out-of-core inventory is proportionally reduced. This allows the number of SFR generations to be reduced by ~1 (Fig. 9).

The above analysis assumes that only a single generation of LWRs is built. If the LWR fleet is held constant until the end of the fission program (as in Fig. 5, for 5 generations), then a much lower proportion of TRU can be incinerated before the end of the scenario. The scenario in Fig. 5 can be analyzed by summing the contributions to radiotoxicity levels and electricity from SFR-Bu-MA1–5. This results in spent nuclear fuel radiotoxicity somewhere between SFR-Bu-MA2 and SFR-Bu-MA3 (Fig. 10).

Over a larger number of generations (estimating the performance for SFR-Bu-MA6–10) then the reduction in performance becomes even worse – over 10 generations of SFRs, the time for decay to the reference level is of the order of 10,000 years (Fig. 11). The radiotoxicity of lower generations (corresponding to the latest constructed LWRs) dominates over higher generations. A relatively low proportion of the TRU from the last LWRs can be incinerated and this TRU dominates over the small amount of TRU left over from preceding generations.

This analysis is obviously limited by the consideration of a large number of generations of LWRs. U resources will ultimately become scarce (OECD Nuclear Energy Agency, 2011) such that if nuclear power continues for several hundred years fast breeder reactors are expected to be deployed.

Hence reduction of radiotoxicity to the reference level within ~1000 years would in practice require the reactor fleet to be steadily reduced over a period of a few hundred years. In the absence of a 300-year phase-out plan for nuclear energy, reduction of radiotoxicity to the reference level with SFRs within ~1000 years appears impractical: a longer decay time may need to be specified.

4.2. Radiotoxicity with Pu only recycle

The repository radiotoxicity for SFR-Bu-Pu1–5 is given in Fig. 12. The radiotoxicity reduction is limited by ^{241}Am and ^{243}Am

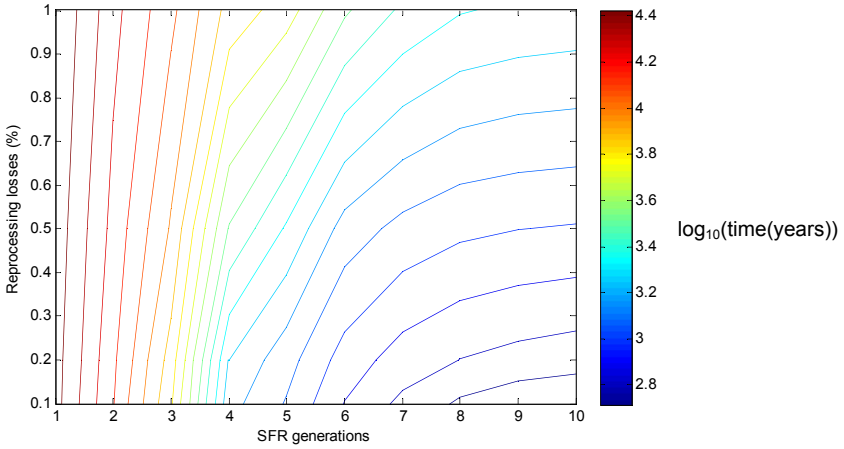

Fig. 9. $\log_{10}$ (time to decay to reference level) as a function of reprocessing losses and number of SFR generations, with 1 year out-of-core time.

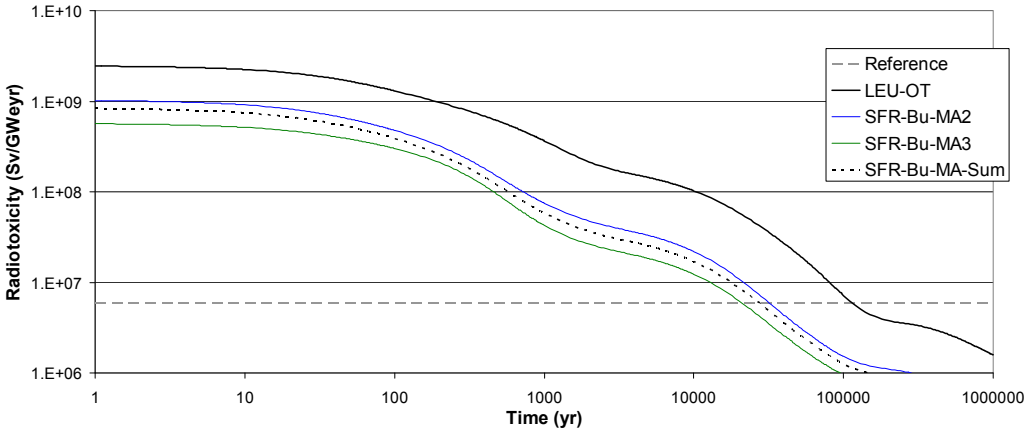

Fig. 10. Repository radiotoxicity for 5 generations of LWRs + SFRs.

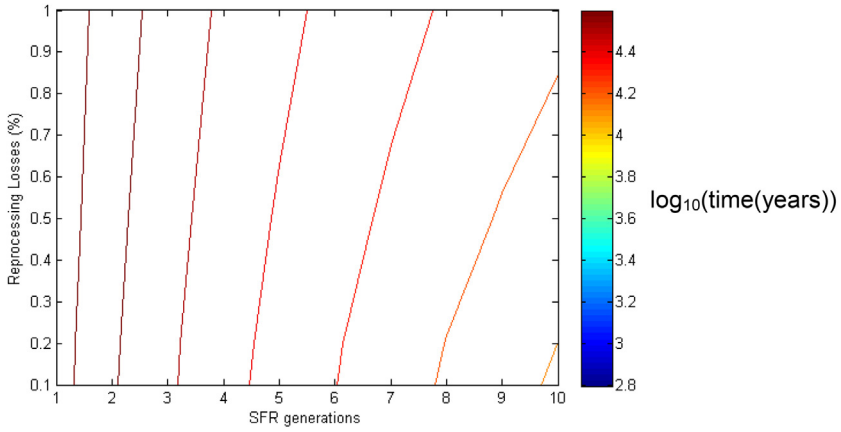

Fig. 11. $\log_{10}$ (time to decay to reference level) as a function of reprocessing losses and number of SFR generations, with 5.75 years out-of-core time and LWR operation over the scenario.

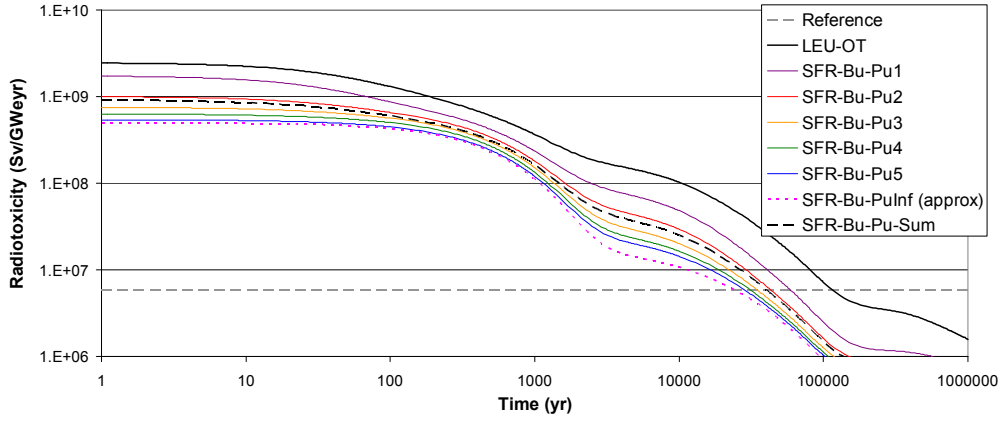

Fig. 12. Repository radiotoxicity for scenarios with Pu recycling.

accumulation in the repository, such that at least ~24,000 years are required for the spent nuclear fuel to decay to the reference level. The MA loading saturates within ~4 generations of SFRs (Fig. 13), allowing the radiotoxicity for an infinite number of recycles to be reliably estimated. ~3 generations of SFRs are sufficient to approach the minimum achievable time for the radiotoxicity to decay to the reference level.

The black dashed line in Fig. 12 gives the effect of continuing to build LWRs over 5 generations (with 5 generations of SFRs, as in Fig. 5). As with SFR-Bu-MA, the radiotoxicity is between that of having 2 and 3 SFR generations with just 1 generation of LWRs (as in Table 3), corresponding to ~40,000 years for the spent nuclear fuel to decay to the reference level. This is already reasonably close to the performance for an infinite number of generations – therefore achieving close to the 'equilibrium' radiotoxicity reduction does not require a gradual phase-out of nuclear power.

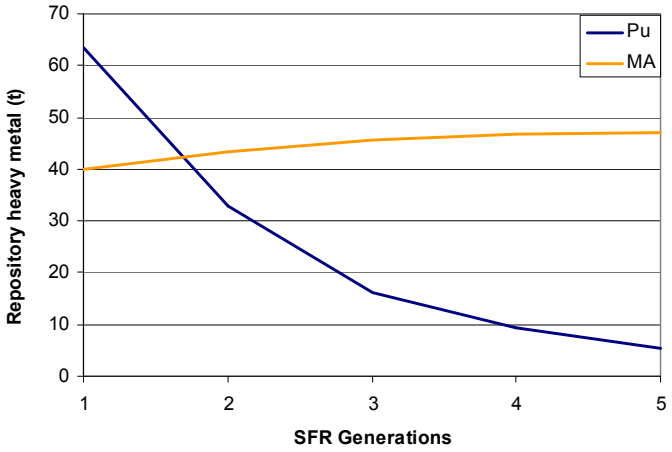

Fig. 13. Repository Pu and MA masses with Pu-only recycling.

4.3. Radiotoxicity with Pu + Am recycle

Am recycle reduces the radiotoxicity compared to Pu-only recycle, but is ultimately limited by a build-up of Cm (in particular ^{244}Cm and its daughter ^{240}Pu). Over 5 generations of SFRs, the reduction in radiotoxicity tends towards a maximum (Fig. 14). As before, the effect of continued LWR operation over this time results in radiotoxicity between that for 2 and 3 generations of SFRs without continued LWR operation.

4.4. Discussion and comparison with break-even SFRs and CORAIL LWRs

The time taken for the radiotoxicity to decay to the reference level is compared for all recycle strategies in Fig. 15.

For burner and break-even SFRs, recycling Am only results in a reduction in decay time after more than 1 generation of SFRs. Beyond this, there is a significant advantage to Am recycle. Recycling Cm is only advantageous after >3 SFR generations, i.e. >220 years after the start of the scenario and in this case >160 years after the LWRs are switched off. As discussed, numerous studies have confirmed that the benefits of recycling Np are minor from a radiotoxicity standpoint – the difference between SFR-Bu-Am and SFR-Bu-MA is due to Cm recycle.

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. The radiotoxicity for the SFR-BE-MA5 scenario 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 length of time can be shortened by

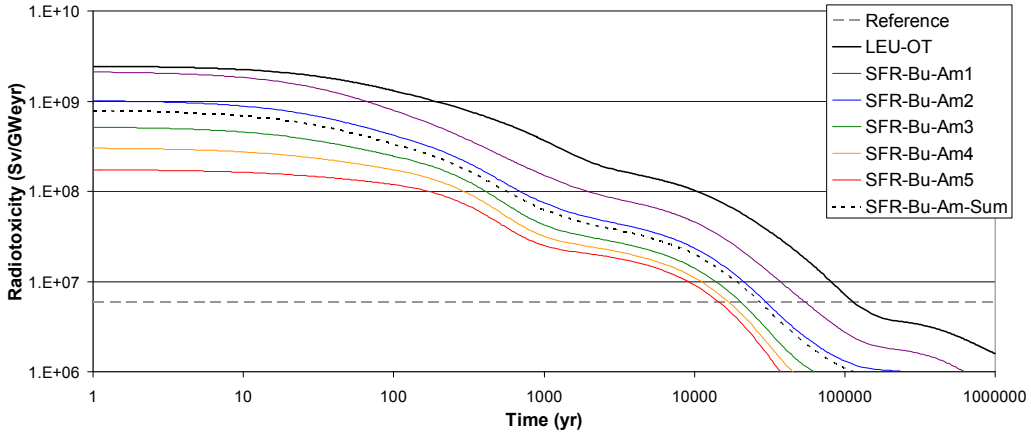

Fig. 14. Repository radiotoxicity with Pu + Am recycling.

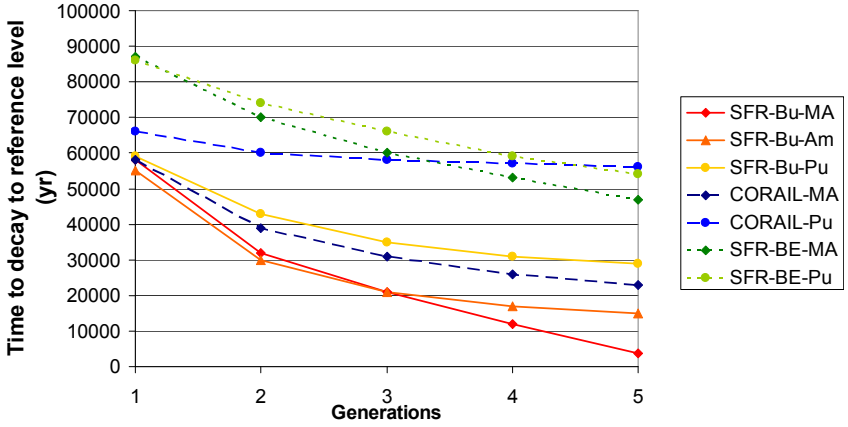

Fig. 15. Repository time to decay to reference level for different recycling strategies.

reducing the out-of-core inventory of the reactor (i.e. by reducing the cooling time).

After 5 generations, CORAIL with MA recycling performs worse than a 'tapering' fleet of SFR burners but slightly better than a fleet of SFR burners operating in conjunction with a fleet of LEU-fueled LWRs (as in Fig. 5). In both cases around 2/3 of the fleet is LEU-fueled LWRs. However, the total CORAIL in + out of core TRU inventory is slightly lower than the SFR burner case, due to the lower enrichment of TRU in the CORAIL core.

Contrastingly, the high MA generation rate in LWRs leads to the radiotoxicity reduction of CORAIL-Pu saturating within ~2 generations, with a much lower reduction in radiotoxicity than with SFR-Bu-Pu.

4.5. Brief discussion of alternative scenarios

Scenarios utilizing SFRs with a breeding ratio greater than unity are now briefly considered. In this case, the SFR fleet size increases over the scenario. The final cores will continue to dominate repository radiotoxicity. The final core inventory can be assumed to be similar that of a break-even SFR and hence the final radiotoxicity will be similar to that of a scenario with break-even SFRs for a given fleet size. However, as the average fleet size over the course of the scenario is less than the final fleet size in this case, the repository radiotoxicity will be normalized over a lower amount of electricity production. Therefore, scenarios utilizing SFRs with a breeding ratio greater than unity will result in higher repository raditoxicity in per GWeyr terms than scenarios utilizing break-even SFRs only.

If SFRs with a breeding ratio greater than unity are first employed for a few generation(s) (implying an initial expansion of SFR capacity and Pu inventory), followed by stabilization of generating capacity with break-even SFRs, the repository radiotoxicity is again higher in per GWeyr terms than for the case with only break-even SFRs. However, the effect of the initial fleet expansion will become less significant over a greater number of generations, as the time-averaged fleet size tends towards the final fleet size.

For scenarios utilizing break-even SFRs, the repository radiotoxicity can be reduced by utilizing SFR burners towards the end of the scenario to reduce the final core inventory. As each generation of SFR burners roughly halves the TRU inventory, utilizing a single generation of SFR burners in this manner can roughly halve the number of generations of SFRs required to achieve a given reduction in repository radiotoxicity.

4.6. Decay heat

Recycling of Pu and MAs can also reduce the peak and integrated heat load in the repository (Generation and International Forum, 2002). This is plotted for all 15 SFR burner scenarios in Fig. 16 and for 5 generations of SFRs in Fig. 17. All recycle strategies are effective at reducing the peak repository decay heat load, although the advantage is quite low for Pu and Pu + Am recycle strategies. For MA and Pu + Am recycle scenarios is a substantial increase in decay heat when the final core is discharged, which is particularly pronounced with MA recycle (for which it is plotted on Fig. 17). The decay heat at discharge of the final core for SFR-Bu-MA1 and SFR-Bu-Am1 are comparable, while for subsequent generations the decay heat at discharge is lower with MA recycling. The increase in decay heat at core discharge for Pu-only recycle is relatively small. The effect of breeding ^{238}Pu from ^{237}Np is significant.

With MA recycle, the integrated decay heat is up to ~50% lower than with an open cycle, and is roughly constant after ~200 years for the scenario considered. Pu and Pu + Am recycle perform less well by this measure, with only a small advantage over the open

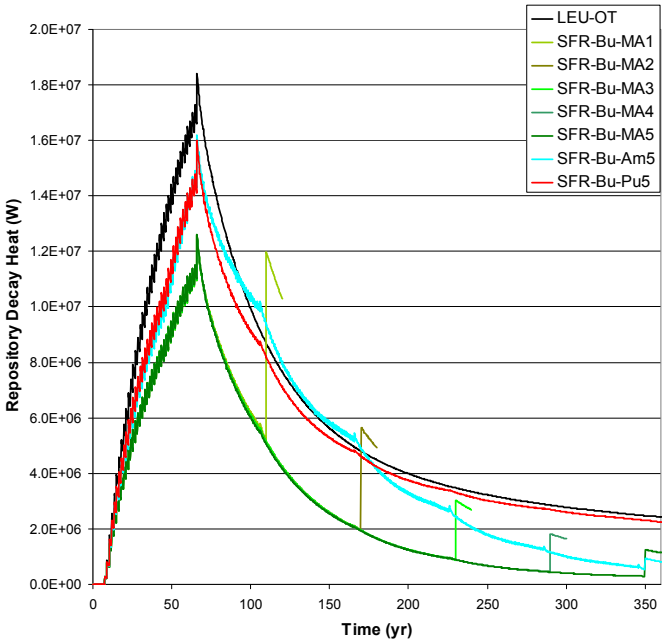

Fig. 16. Repository decay heat for SFR burner scenarios.

cycle. Pu-only recycle results in the integrated decay heat being very similar to the open cycle by the end of the scenario. In general, these strategies result in continuous production and discharge of Am/Cm from Pu/Am capture which leads to substantial decay heat over the longer term.

A few generations are required before Am recycle becomes advantageous relative to Pu-only recycle, which could limit its merits. The beneficial effect of recycling Am is countered by increased ^{238}Pu production through neutron capture and subsequent decay of ^{241}Am: some of this ^{238}Pu is ultimately loaded in the repository at the end of the scenario. This is roughly consistent with (Generation and International Forum, 2002) which shows advantages to Am + Np recycle after ~200 years of break-even fast reactor operation.

With continued LWR construction (as in Fig. 5) and full MA recycle, the repository decay heat tends to a constant value ~200 years into the scenario (Fig. 18). When the nuclear program is terminated, the decay heat initially reduces while the remaining SFRs operate (as the SFRs lag slightly behind the LWRs for the scenarios considered here). There is then a jump in repository decay heat when the remaining TRUs (either as unreprocessed spent fuel or separated TRU) are disposed of. The peak repository decay heat would be slightly higher if reprocessing stopped early –

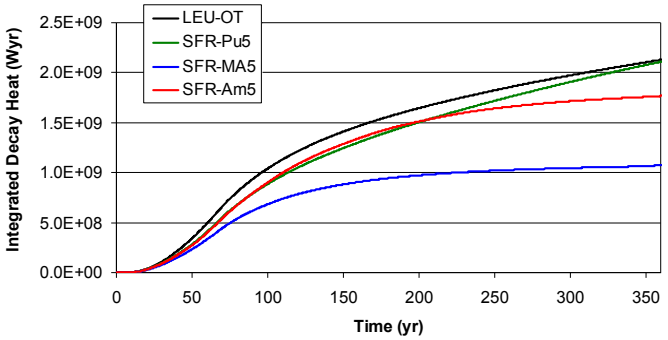

Fig. 17. Integrated repository decay heat.

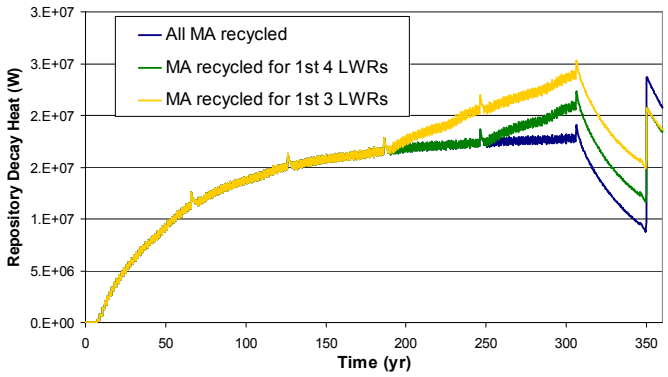

Fig. 18. Repository decay heat for reactor fleet shown in Fig. 5.

as the decay heat from separated TRU inventories between 300 and 350 years is not included in the repository decay heat. If MA reprocessing is stopped early, then it appears possible to slightly reduce the maximum decay heat in the repository.

Repository decay heat for the SFR break-even and CORAIL scenarios is shown in Figs. 19 and 20 respectively. As before, the decay heat over ~40–100 years is determined by the transition from one type of reactor to another and hence is unlikely to be representative of a realistic, gradual transition. For these scenarios, it is also higher as a result of including fission products from the entire fleet of LEU-fueled LWRs, not just the LWRs required to generate the TRU required to start the recycling reactors. The decay heat is not directly comparable between cases as the fleet sizes are also slightly different.

However, it can be observed that the decay heat follows a similar trend for the SFR burner and break-even scenarios, although without MA recycling the decay heat begins to rise slightly for the break-even SFRs after ~200 years as the fleet size does not reduce. After the initial transient, peak repository loading with and without MA recycling remains similar for break-even SFRs. The additional decay heat at discharge of the final core is very small for SFR-BE-Pu# scenarios, while it is substantial and varies little with the number of generations for SFR-BE-MA#.

For CORAIL, the decay heat continues to rise steadily without MA recycling due to the accumulation of MAs in the repository. Similarly, the high MA population in the CORAIL assembly leads to a large increase in repository decay heat when the CORAIL-TRU assemblies are discharged at the end of the scenario (which as with SFR-BE-MA# varies little with number of generations). At the end of the CORAIL-MA5 scenario, the MA inventory is ~14 kg/GWeyr, compared to ~3.5 kg/GWeyr for the SFR-BE-MA5 scenario. As with SFR-BE-PU#, the additional decay heat at discharge of the final cores for CORAIL-Pu# is very small. Therefore, from a decay heat perspective, several generations of PWRs may be necessary before MA recycle in CORAIL assemblies becomes worthwhile relative to recycle of Pu only.

4.7. Effect of varying reprocessing and fuel fabrication losses over scenario

It is possible that reprocessing and fuel fabrication losses would reduce over time due to improvements in technology. For scenarios utilizing break-even SFRs or CORAIL assemblies, lower reprocessing and fuel fabrication losses later in the scenario would have a limited impact on repository radiotoxicity, as this is dominated by the final cores. However, the decay heat over the scenario would somewhat reduce due to lower discharge of actinides to the repository from reprocessing and fuel fabrication. For burner scenarios, reprocessing losses become significant over a large number of generations. However, the reprocessing and fuel fabrication losses of the earlier generations dominate as the fleet size and hence the mass flows for these generations is larger, hence the impact of reduced reprocessing and fuel fabrication losses later in the scenario is again limited, and losses early on in the scenario will tend to dominate.

5. Conclusions

To achieve a repository radiotoxicity reduction approaching that achievable at equilibrium, ~6 generations of SFRs are

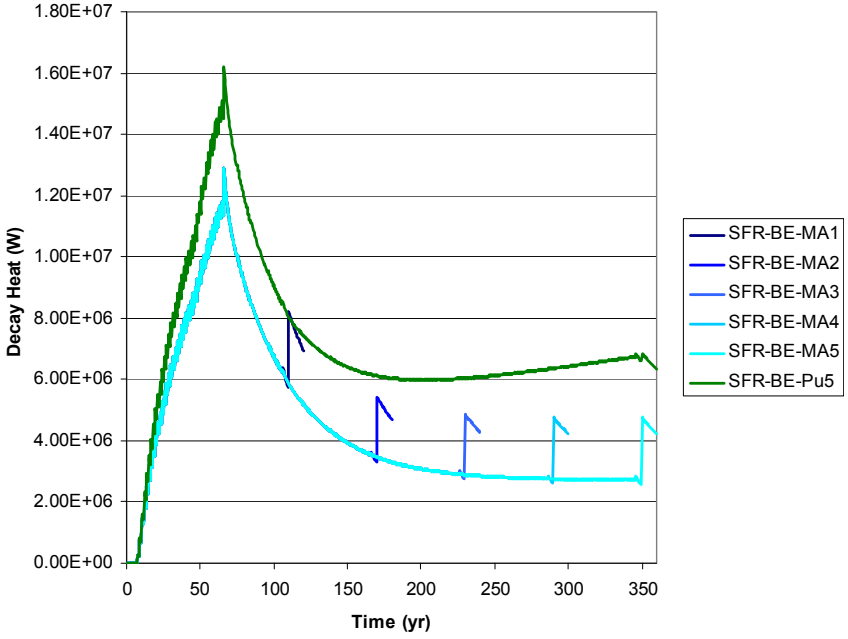

Fig. 19. Repository decay heat for break-even SFR scenarios.

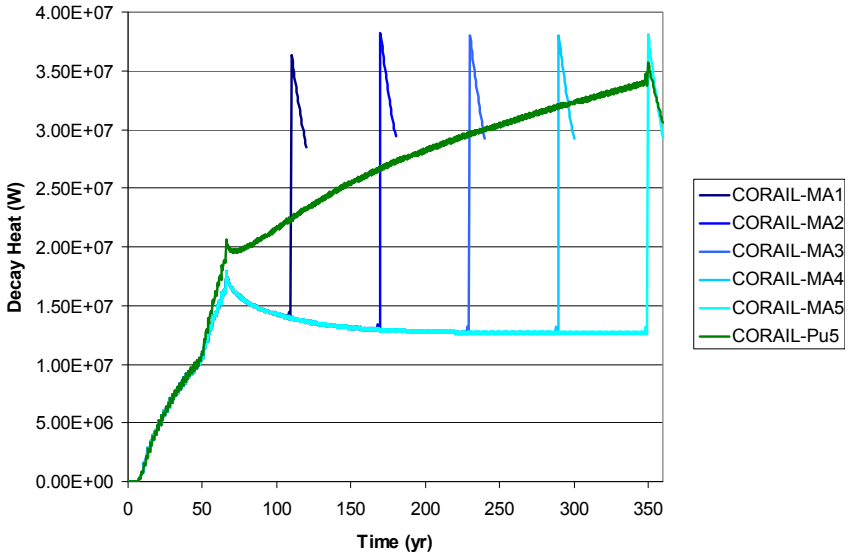

Fig. 20. Repository decay heat for CORAIL scenarios.

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 an economic and energy security standpoint.

To realize the more limited radiotoxicity reduction from recycling Pu + Am or Pu only, fewer SFR generations are required. ~3 generations are sufficient for Pu recycle to achieve radiotoxicity approaching the minimum achievable, and it is not generally required to reduce the fleet size prior to the end of the nuclear program.

More than 3 generations of SFRs are required (>220 years for the scenarios considered) before Cm recycle becomes worthwhile from a radiotoxicity standpoint, although over a large number of generations it may be practical to wait for the Cm to decay before recycling it.

Pu, Pu + Am and MA recycle are progressively more effective at reducing peak and integrated repository decay heat, although >1 generation of SFRs is required to realize this. From a decay heat standpoint, >3 generations of SFRs are required before recycling Pu + Am becomes worthwhile relative to recycling just Pu.

TRU recycle in PWRs with zero net actinide production provides similar performance to LEU-fueled LWRs in equilibrium with a fleet of burner SFRs. However, it is not possible to reduce the TRU inventory over multiple generations of PWRs. Also, the high rate of MA production leads to a much larger repository decay heat than for the open cycle or SFR scenarios.

TRU recycle in break-even SFRs is much less effective from a point of view of reducing spent nuclear fuel radiotoxicity, although still effective from the point of view of reducing repository decay heat.

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