Sustained recycle in light water and sodium-cooled reactors

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Abstract

From a physics standpoint, it is feasible to sustain recycle of used fuel in either thermal or fast reactors. This paper examines multi-recycle potential performance by considering three recycling approaches and calculating several fuel cycle parameters, including heat, gamma, and neutron emission of fresh fuel; radiotoxicity of waste; and uranium utilisation.

The first recycle approach is homogeneous mixed oxide (MOX) fuel assemblies in a light water reactor (LWR). The transuranic portion of the MOX was varied among Pu, NpPu, NpPuAm or all-TRU. (All-TRU means all isotopes through 252 Cf.) The Pu case was allowed to go to 10% Pu in fresh fuel, but when the minor actinides were included, the transuranic enrichment was kept below 8% to satisfy the expected void reactivity constraint. The uranium portion of the MOX was enriched uranium. That enrichment was increased (to as much as 6.5%) to keep the fuel critical for a typical LWR irradiation.

The second approach uses heterogeneous inert matrix fuel (IMF) assemblies in an LWR – a mix of IMF and traditional UOX pins. The uranium-free IMF fuel pins were Pu, NpPu, NpPuAm or all-TRU. The UOX pins were limited to 4.95%²³⁵U enrichment. The number of IMF pins was set so that the amount of TRU in discharged fuel from recycle N (from both IMF and UOX pins) was made into the new IMF pins for recycle N+1. Up to 60 of the 264 pins in a fuel assembly were IMF. The assembly-average TRU content was 1-6%.

The third approach uses fast reactor oxide fuel in a sodium-cooled fast reactor with transuranic conversion ratio of 0.50 and 1.00. The transuranic conversion ratio is the production of transuranics divided by destruction of transuranics. The FR at CR = 0.50 is similar to the CR for the MOX case. The fast reactor cases had a transuranic content of 33-38%, higher than IMF or MOX.

Introduction

This paper presents various fuel cycle parameters for three methods of sustainably recycling used nuclear fuel based on reactor physics calculations documented elsewhere [1-7]. We do not claim that these are the only ways or the best ways to sustainably recycle. The objective is to gain insights into the radioactivity of fresh fuel (heat, gamma, neutron emission), radiotoxicity of waste, and uranium utilisation from sustained recycle versus the first recycle. The baseline is always LWR with uranium oxide fuel at 51 MWth-day/kg-iHM burn-up.

The paper starts with a summary of the reactor physics calculations documented elsewhere. Next, we describe the heat, gamma, and neutron emission of fresh fuel. Finally, the paper addresses radiotoxicity and uranium utilisation.

Multi-recycle of MOX-UE in LWR

Two participants of this study analysed [1] the reactor physics of homogeneous recycling of different TRU mixtures (Pu, PuNp, PuNpAm, TRU) in pressurised water reactors (PWR) using MOX-UE fuel, i.e. MOX fuel with a ²³⁵U enriched uranium support instead of the depleted uranium typical of standard MOX fuel today. From a neutron physics point of view, the MOX-UE approach allows sustained multi-recycling of TRU as long as ²³⁵U is available. The key caveat to this sustainable recycle is keeping the TRU content per heavy metal at a value that ensures a negative moderator void coefficient. Once this value is determined, the ²³⁵U enrichment of the MOX-UE fuel is adjusted in order to reach the target burn-up (51 MWth-day/kg-iHM in this study). For the Pu recycle case, the Pu content is kept to 10% to ensure a negative void coefficient. When the minor actinides were included, the transuranic enrichment was kept below 8% to satisfy the expected void reactivity constraint. The uranium portion of the MOX was enriched uranium. That enrichment was increased (to 6.5%) to keep the fuel critical for a typical LWR irradiation.

Multi-recycle of heterogeneous IMF in LWR

Two participants of this study analysed [2] the reactor physics of multi-recycling using a heterogeneous inert matrix fuel (IMF) and UOX fuel assembly concept. The initial study was focused on direct thermal recycling of IMF fuels in a heterogeneous PWR 17×17 bundle design in a multi-recycle strategy. These analyses provided comparison of direct thermal recycling of the four actinide partitioning cases. The TRU loading in the IMF pins was adjusted to ensure a pin power peaking factor no greater than 1.2. The enrichment of the uranium in the UOX pins was adjusted to achieve a 1 500 day fuel life in a three-batch strategy, which is equivalent to ~51 MWth-day/kg-iHM in UOX or MOX. TRU elements of the given partitioning strategy were recycled from UOX and IMF pins in the previous cycle and loaded into IMF pins of the current cycle.

Multi-recycle in FR

One participant of this study analysed [5-7] the reactor physics of multi-recycling in fast reactors, first with transuranic (TRU) conversion ratio (CR) below 1.0 and more recently with CR above 1.0. The fast reactor concept is that of PRISM, a traditional fast reactor concept. These calculations estimated fuel composition for the first recycle and sustained recycle. A different team member then analysed the evolution of composition for TRU CR = 0.50 from first recycle to equilibrium sustained recycle [3].

A separate paper at this conference details parameters as a function of TRU CR. To study the impact of conversion ratio, only metal-fuelled fast reactors were the focus rather than oxide-fuelled because the former has higher fuel density and is believed to be capable of higher TRU conversion ratio. In this paper, to study the impact of minor actinide recycle strategies, oxide-fuelled fast reactors were the focus because of the presumption that aqueous separation (for which separation of one TRU element from the others) would be used whereas the common presumption is that electrochemical separation (for which separation among TRU elements is difficult or impossible) would be used with metal fuel.

Composition

Figure 1 compares the composition at equilibrium as a function of which minor actinides are recycled for multi-recycle MOX, multi-recycle IMF, and fast reactor oxide at CR = 0.50. The recycling of Cm-Bk-Cf is the dominant determinant to accumulation of higher actinide isotopes, not the reactor and fuel type. The following sections will show the impact of these composition trends on heat, gamma, and neutron emission.





One of the parameters that stem from the isotopic compositions is the support ratio, which is how much fuel for recycle N is made from used fuel at recycle N-1. For MOX-UE, the first recycle's support ratio is 6-8 (depending on minor actinide approach), i.e. the amount of first recycle MOX is 6-8 times lower than the amount of LWR-UOX-51. The support ratio thereafter is in the range of 1.1 to 1.3.

For heterogeneous IMF, the calculations were done with a support ratio near 1.

Similar to MOX-UE, the support ratio for the FR cases declines with successive recycles. At CR-0.50, the support ratio for the first recycle is about 26, declining to 6 at equilibrium recycle. For example, the first recycle fuel for FR at CR = 0.50 is 33% TRU. At discharge, LWR-UOX-51 is 1.3% TRU, therefore the support ratio is 33%/1.3% = 26. At CR = 1.00, the first recycle support ratio is 14; thereafter, the support ratio is infinite as this case is (barely) a breeder.

Fresh fuel decay heat

When one is comparing fuels from one reactor type to the next, there are different ways to compare or normalise the values. In this paper, we use three.

- Per fission energy (MWth-day) particularly appropriate when considering fuel cycle performance, indeed, MWth-day is the denominator in the burn-up parameter.
- Per kg-fuel particularly appropriate when considering the hazard posed during fabrication, transportation, etc.
- Per kg-TRU helpful to understand underlying physics trends because it shows the impact of TRU isotopic mix, whereas "kg-fuel" exhibits trends from both TRU isotopic mix and the TRU:U ratio that varies by reactor and TRU CR.

Figure 2 compares heat generated as a function of recycle pass and MA recycle strategy, per fission energy generated. The FR increase of heat from first to equilibrium recycle is the least, i.e. the lines are generally the most flat, especially for the TRU-recycle case. (We have not calculated the composition evolution for Pu or NpPuAm for the fast reactor.) Figure 3 shows the equilibrium heat for the cases in two sets of units – per kg-fuel and per kg-TRU. The "per kg-TRU" pattern in Figure 3(b) is particularly clear, heat increases in progressing from Pu to PuNp to PuNpAm to all-TRU. The pattern in Figure 3(a) (per kg-fuel) is more complex; but it demonstrates the differences among fuel and reactor types in terms of the TRU content in the fuel. The fast reactor has a higher enrichment by about a factor of three.





Note that nuclear material at 450 W/kg may be considered not directly usable for a nuclear weapon. The IAEA denotes Pu mixtures with 80% ²³⁸Pu as self-protecting because of heat generation. This is about 450 W/kg-Pu. None of the cases reach this value when the uranium is excluded, i.e. in Figure 3(b). The closest are the TRU cases: MOX is 215, IMF is 247, FR/CR = 0.50 is 89, FR/CR = 1.00 is 48. Of course, it is also possible to take the TRU mixtures and subsequently separate the high heat minor actinides from lower heat Pu, in which the Pu would not be generating such heat.



Figure 3: Fresh fuel decay heat at equilibrium recycle as a function of MA partitioning strategy (a) per mass of fuel, (b) per mass of TRU

Fresh fuel gamma emission

The gamma emission comparison is very similar to the heat comparison. Figure 4 shows the evolution of gamma emission upon multiple recycles. The NpPu cases are not shown; they are similar to the Pu cases. Figure 5 shows the gamma emission at equilibrium. Figure 5(b) (per kg TRU) shows the initial TRU feed from discharged UOX for comparison so that one can see how recycling has changed gamma emission from its starting point – a modest impact. Figure 5(a) (per kg HM) shows gamma emission normalised to mass of heavy metal, for which a comparison with used UOX is possible. Used UOX emits about 1.2 W/kg-HM at 5 years after discharge, dropping to 0.3 W/kg-HM at 30 years and 0.06 W/kg-HM at 100 years – such cases are considered "self-protecting" from the proliferation standpoint. Fresh recycled fuel is below such numbers.



FR-oxide @ CR = 0.50, MOX-UE, heterogeneous-IMF





Figure 5: Fresh fuel gamma emission at equilibrium recycle as a function of MA partitioning strategy (a) per mass of fuel, (b) per mass of TRU

Fresh fuel neutron emission

Figure 6 shows the evolution of neutron emission upon multiple recycles. The NpPu cases are not shown to keep the figure relatively readable; NpPu results are close to Pu. The only FR-NpPuAm data is a single point at equilibrium. Figure 7 shows the neutron emission at equilibrium. Use of fast reactors, rather than LWR mitigates some of the neutron penalty of recycling Cm-Bk-Cf but not by multiple orders of magnitude. Fast reactors have generally higher fission-to-absorption ratios than thermal reactors. Hence, fast reactors are more adept at transmuting by fission as opposed to neutron capture the higher mass actinides, Cm-Bk-Cf. Thus, the build-up of the primary spontaneous fission neutron emitters is less in fast reactors than it is in thermal reactors. Nevertheless, the effect of global reaction rate [i.e. gross transmutation(s)] is also relevant as it reflects the transient build-up of the ²⁴⁴Cm and ²⁵²Cf evolution as a function of calendar time over multiple recycles.

Radiotoxicity

An internationally used metric relevant to waste management is radiotoxicity, which is the inventory of waste normalised by each isotope's hazard. Here, the hazard metric is ICRP-72 ingestion dose conversion factors as the primary exposure pathway of concern is eventual dissolution of waste in water, followed by ingestion of water or food. Radiotoxicity only denotes the hazard of an inventory and not the performance of any waste form, disposal method or disposal site. In essence, it tells us how much performance must be attained by waste form, disposal and siting.

Radiotoxicity does not capture beneficial effects from dilution of radioactive isotopes in the environment, in the waste form and packaging, or in the waste contaminants themselves. For example, our analyses point to ^{166m}Ho as a key long-lived lanthanide isotope. Its dose factor and thus its radiotoxicity implicitly assume it is the only lanthanide ingested. Yet, ^{166m}Ho is only ~1 ppm of the total lanthanide mass in used fuel fission products, most of which are stable. It is not clear that ^{166m}Ho could be ingested and retained by the body to the degree it would be if it was the only lanthanide ingested. Similarly, long-lived ¹³⁵Cs is diluted by all the stable caesium isotopes in the waste itself, ¹²⁹I by stable iodine, etc.





FR-oxide @ CR = 0.50, MOX-UE, heterogeneous-IMF

Figure 7: Fresh fuel neutron emission at equilibrium recycle as a function of MA partitioning strategy (a) per mass of fuel, (b) per mass of TRU



(alpha,n) neutrons not included

Figure 8 shows the radiotoxicity for the three sustainable recycling approaches, with LWR UOX-51 as context.¹ The calculations are taken to a 1e9 years to show the underlying physics, even though waste management assessments tend to stop at a 1e6 years. The radiotoxicity of all waste is included except for depleted uranium. The radiotoxicity changes little with one recycle (if followed by direct disposal of the used recycled material), regardless of the approach used for that single recycle. The most effective single recycle found to date is homogeneous LWR-IMF or its HTGR analogue, called "deep burn" [8], but even that is a small change relative to LWR UOX-51.

The sustained recycle cases in Figure 8 are more interesting. From 100 years to ~1e7 years, sustained recycle of MOX-Pu has noticeably lower radiotoxicity than the once through and single recycle cases. All of these cases are flat after about 1e7 years as then the radiotoxicity is dominated by uranium isotopes and their progeny decay products. The TRU in waste and the recovered uranium from separation dominate radiotoxicity.

Sustained recycle of MOX-TRU and FR at CR = 0.50 (with 0.1%/recycle processing loss rate) has markedly lower radiotoxicity by a few hundred years after reactor discharge. On this graph, the natural uranium ore that gave rise to the fuel is several Sv/MWth-day. Thus, with zero or a single recycle, the time for waste to fall to uranium ore is 1e5 to 1e6 years; the time for MOX-Pu to fall to uranium ore is 1e4 to 1e5 years; but the time for MOX-TRU or FR-TRU to fall to uranium ore is at or below 1e3 years. Beyond 1e7 years, the radiotoxicity of MOX-TRU and FR at CR = 0.50 are dominated by the recovered uranium and the residual TRU in waste that has mostly decayed to uranium and its progeny. At such long times, the reduction relative to the original uranium ore mostly stems from removal of depleted uranium from the comparison.

The lowest radiotoxicity case is FR at CR = 1.00. For $CR \ge 1$, there is no excess recovered uranium from separation (and no excess depleted uranium). In such breeder cases, radiotoxicity continues to decline as all that remains is the 0.1% processing losses and fission products.



Figure 8: Radiotoxicity of FR-oxide @ CR = 0.50, MOX-UE, heterogeneous-IMF

^{1.} The unit in Figure 8, Sv/MWth-day, arises from dose factor (Sv/kg-isotope) × isotopic composition (kg-isotope/kg-HM) ÷ burn-up (MWth-day/kg-HM). For uranium ore, the ratio of kg-natural-uranium/kg-enriched uranium is included, which is about 8 for LWR-UOX.

Uranium utilisation

Figure 9 shows the uranium utilisation of MOX-UE as a function of recycle for the four minor actinide partitioning options. The highest uranium utilisation arises from recycling only Pu; it increases from 0.77% to 0.91%, higher than LWR UOX-51, which is 0.68%. Thus, sustainable MOX-Pu can increase utilisation relative to LWR UOX-51 by 34% (from 0.68% to 0.91%). Recycling NpPu provides improvement relative to LWR UOX-51 but less than recycling only Pu because Np is a net neutron absorber. Recycling NpPuAm further reduces uranium utilisation, but recycling all-TRU is slightly higher than NpPuAm because Cm is a net neutron provider.





The utilisation for all MOX cases is below 1%. The heterogeneous IMF results are similar. Figure 10 shows uranium utilisation when FR cases are added. Figure 10 only shows the first and equilibrium recycle for MOX-Pu and MOX-TRU. The first recycle (all TRU) with traditional fast reactors is about the same as MOX-Pu. (This paper does not address exotic once-through "breed and burn" fast breeder reactors in which an initial enriched ²³⁵U zone breeds ²³⁹Pu in surrounding ²³⁸U, which is allowed to fission *in situ* assuming that the fuel and its cladding survive very high neutron fluence. These cases can reach uranium utilisation ~10% [8,9].)

Subsequent FR recycles further increase uranium utilisation, with the improvement increasing as the FR TRU CR increases For CR \leq 0.50, the uranium utilisation stays below 1% even at equilibrium sustainable recycle. At CR = 0.75, it is 1.5%. At CR \geq 1.0 (breeders), the uranium utilisation approaches 100%, limited by processing loss rate each recycle. At equilibrium, processing loss rates of 0.1% and 1.0%/recycle lead to utilisation of 99% and 87% respectively, if all TRU are recycled. If only Pu is recycled, the minor actinides increase the effective processing losses. Then, the raw process loss rates of 0.1% and 1.0% become 0.55% and 1.4%, with corresponding uranium utilisation of 93% and 81%. Achieving utilisation significantly over 1% requires a breeder reactor.



Heavy metal consumption rate (tonne of natural uranium HM/GWth-day)

Conclusions

From a physics standpoint, it is feasible to sustain multi-recycle in either thermal or fast reactors with enriched uranium support.

When recycling Pu, NpPu, or NpPuAm, there is about an order of magnitude variation of heat, gamma, and neutron emission per kg of fuel among MOX, IMF, and fast reactors with CR = 0.50.

If all the transuranics are recycled, there is an additional half-order of magnitude increase in heat and gamma. Isotopes that dominate heat and gamma emission are scattered throughout the actinide chain, so the modest impact of recycling Cm-Bk-Cf with Np-Pu-Am is unsurprising. Neutron emitters are preferentially among the higher actinides, so the neutron emission increases more when Cm-Bk-Cf are recycled. The fast reactor with CR = 0.50 with all-TRU recycling has two orders of magnitude *lower* neutron emission (per kg-TRU) than the LWR options with all-TRU recycling, but more than two orders of magnitude *higher* neutron emission than LWR options with NpPuAm recycling.

Even though the isotopic mix continues to evolve, heat and gamma tends to increase little after ~5 recycles. Neutron emission tends to increase for more recycles as the highest actinides are the part of the chain that takes longest to reach equilibrium (if all TRU are recycled).

The radiotoxicity of waste is only slightly reduced by a single recycle in the cases studied. Sustained recycle via MOX-Pu reduces radiotoxicity, but by less than an order of magnitude. Still, this can lead to a significant (more than an order of magnitude) reduction in the time required before waste drops to the level of natural uranium ore. Sustained recycle of all-TRU leads to orders of magnitude reduction in the time periods that have tended to dominate in waste management assessments (1e3 to 1e6 years). Sustained recycle of all-TRU in a breeder reactor additionally reduces radiotoxicity at yet longer times because there is no excess recovered or depleted uranium.

Even with recycling taken to equilibrium, the uranium utilisation of both MOX and IMF remains below 1%. The uranium utilisation of traditional fast reactors starts below 1% and can increase slightly over 1% for TRU CR below 1. Achieving uranium utilisation significantly over 1% requires a breeder reactor (TR CR \geq 1).

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