

Waste Classification based on Waste Form Heat Generation in Advanced Nuclear Fuel Cycles Using the Fuel-Cycle Integration and Tradeoffs (FIT) Model – 13413

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ABSTRACT

This study explores the impact of wastes generated from potential future fuel cycles and the issues presented by classifying these under current classification criteria, and discusses the possibility of a comprehensive and consistent characteristics-based classification framework based on new waste streams created from advanced fuel cycles. A static mass flow model, Fuel-Cycle Integration and Tradeoffs (FIT), was used to calculate the composition of waste streams resulting from different nuclear fuel cycle choices. This analysis focuses on the impact of waste form heat load on waste classification practices, although classifying by metrics of radiotoxicity, mass, and volume is also possible. The value of separation of heat-generating fission products and actinides in different fuel cycles is discussed. It was shown that the benefits of reducing the short-term fission-product heat load of waste destined for geologic disposal are neglected under the current source-based radioactive waste classification system, and that it is useful to classify waste streams based on how favorable the impact of interim storage is in increasing repository capacity.

INTRODUCTION

The radioactive waste classification system in the United States primarily relies on a source-based framework (e.g., U.S. wastes are categorized by where they originated, not the specific hazards posed by their disposal). This approach is known to have many deficiencies [1]. For one, the basis of U.S. classification within the framework is inconsistent; for example, high-level waste is defined by its source, whereas low-level waste is defined by exclusion [2]. Also, the classification rules leave some orphan wastes without any legal framework for disposal. This includes mixed low-level waste (MLLW) and depleted uranium (DU) from enrichment plants, among others, which the regulatory framework does not currently address. Furthermore, the possible option of closing the fuel cycle (and thus processing used nuclear fuel) would lead to the creation of different waste streams that, under the current source-based framework, could all be defined as high-level waste (HLW), even if their characteristics and hazard levels may not reflect the need for HLW disposal requirements or may simplify and improve the performance of disposal.

The primary issue in using a source-based classification system is the inability to minimize both the disposal cost and risk to the public by assuring that all types of waste are managed optimally. If

one purpose of developing advanced fuel cycles is to reduce the long-term burden of radioactive waste, a source-based waste classification system would overlook the benefit of a fuel cycle that directs materials to be appropriately managed where they would generate the lowest risk to the public.

Rethinking our radioactive waste classification system has been explored in the past [3]. Current revisions to 10CFR61 for low-level waste (LLW) definitions are being discussed, and on the international level, the IAEA recommends a characteristics-based classification system to its member states [4]. This study aims to apply previous investigations of how to redraw the classification boundaries for radioactive waste to the results of the advanced fuel cycle systems model considered in this study.

MODELS AND ASSUMPTIONS

The Fuel-cycle Integration and Tradeoffs (FIT) model, developed by Idaho National Laboratory (INL), is a static mass flow model that tracks isotopes through a fuel cycle of the user's choice; from discharged used fuel to recycle in a reactor to waste disposition [5]. FIT is a systems-level model that allows the user to compare nuclear fuel cycles with different parameters of choice, and to analyze how changes in these parameters in one part of the fuel cycle affect other parts of the fuel cycle.

Figure 1 depicts the basic mass flow and processes of the FIT model. The incoming used fuel, light water reactor uranium oxide (LWR UOX) at 51 MWth-day/kg-iHM burnup, goes through an aqueous separation process. The recovered actinides, the recovered uranium, and depleted uranium (DU) as a makeup feed, provide the material needed to fabricate metal or oxide fuel for irradiation in a second reactor, either a fast burner reactor or a mixed-oxide fuel light water reactor (MOX-LWR). After discharge, the used fuel goes through a second separation (electrochemical reprocessing if metal fuel and an aqueous process if oxide fuel). Some materials are recovered for another pass in the reactor and the rest are managed as waste. FIT is capable of modeling up to 18 recycle loops in appropriate cases. In the two fast burner cases in this study, 18 recycle iterations are sufficient for the fuel to approach equilibrium composition values.

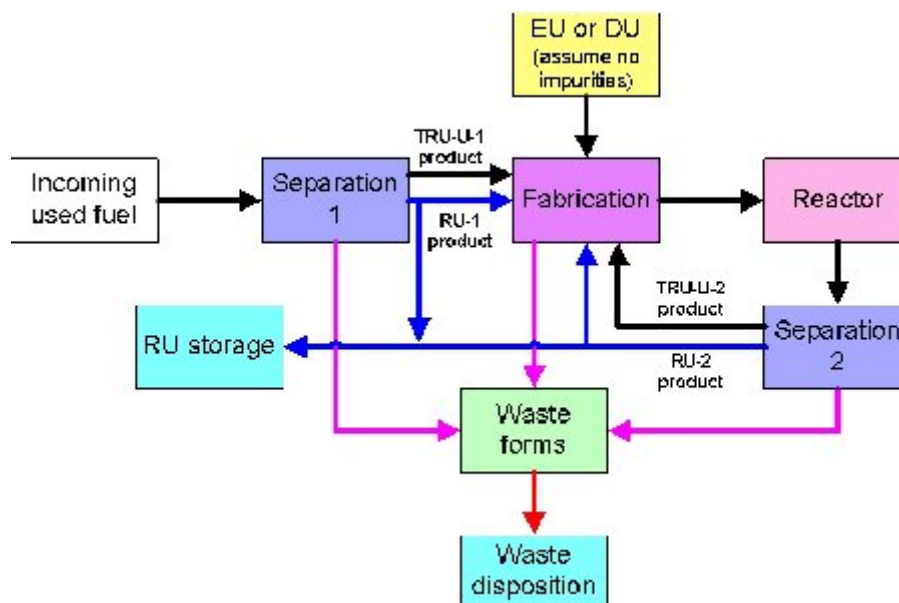


Figure 1: Basic mass flows; FIT does not yet address enriched uranium (EU) feed as an option.

For every case in this study (see Table 2), recovered uranium RU-1 was used for the first iteration (RU-2 is not generated unless the first cycle is completed, as shown in Figure 1), and RU-2 was used for every recycle after the first (RU-2 and recovered transuranics (TRUs) are kept together for fuel fabrication). No breeder cases were considered for this study. Therefore, every case considered has excess uranium in the system; so it is reasonable to assume that there will always be enough RU-2 as a makeup feed for fuel fabrication.

There are two separation modules: Separation 1, in which the partitioning of the incoming used fuel (LWR UOX-51) occurs, and Separation 2, where the used fuel of the second reactor is processed. For each of these separation modules, FIT allows for several separation technology options [5], but for this study, only UREX+1a, electrochemical processing, and PUREX were considered where appropriate.

For simplicity, the separation efficiencies for each module are assumed to be constant with every recycle. (In reality, as impurities accumulate with each recycle loop, separation efficiencies for different isotopes might increase or decrease.) In cases where no separation data were available, a default 99.9% isotopic recovery fraction was assumed. The user can define a loss fraction for undissolved solids (UDS) in aqueous separations processes. For this study, a 0.1% UDS loss fraction was assumed for UREX+1a separations. The user can also choose whether UDS gets vitrified along with the raffinate/residual waste in HLW glass for aqueous separations processes, or stays as a separate waste stream to be immobilized in a metal alloy ingot. In this study, UDS was

assumed to combine with the HLW glass waste form. For an electrochemical separations process, UDS, cladding, and baskets are immobilized in a metal alloy ingot.

Waste management is addressed in the model by tracking the masses of all materials separated from used fuel that are not recycled into new fuel. Waste streams generated during fuel fabrication are also tracked. All tracked waste streams are converted into a waste form suitable for disposal, based on current waste form feasibility studies. The waste stream characteristics can then be examined to make assessments about their management. At this time, FIT does not calculate waste volumes based on waste packaging design assumptions. This was done to avoid confusion with the actual volume of the waste forms.

No operational and maintenance (O&M) waste from front-end processes, the initial fuel fabrication, or the two reactors in the cycle (the initial LWR and the recycle reactor) have been considered in the model implementation to date. However, O&M wastes for separations and fuel fabrication are included; FIT currently only deals with “technology wastes” that specifically contain wastes separated from the used fuel.

Table 1 lists the various waste forms used to immobilize respective separated waste isotopes from their respective separations process. An attempt was made to classify the resulting differing waste streams according to current classification criteria based on their properties similar to those of existing waste classes. For example, the Cs/Sr waste stream, if separated from the used fuel, could go to decay storage as MLLW (mixed low-level waste) because it has a small long-term heat burden. However, under the current legal framework, most of the waste forms shown in Table 1 should be classified as HLW.

Table 1: Separated isotope(s), process of origin, respective waste forms, and their suggested disposition pathway.

Separated waste isotope(s)	From these separations processes	Waste form	Best estimate for waste class
C-14	All but electrochemical	Grouted carbonate (C-14 Grout)	HLW-Repository
I-129 (and co-collected halogens)	All	I-129 Glass Ceramic	HLW-Repository
Tc-99, UDS	UREX+1	Metal Alloy Ingot	HLW-Repository
Tritium	All	Grouted Tritiated water (H-3 Grout)	LLW
Kr-85 (and other Kr and Xe isotopes)	All	Compressed gas (Kr/Xe in cylinders)	LLW-Decay Storage

Cladding, UDS, baskets	Electrochemical	Metal Alloy Ingot (Echem)	HLW-Repository
Raffinate/residual (Could include Tc, UDS, Cs/Sr, and lanthanides)	All but electrochemical	Glass	HLW-Repository
Raffinate/residual (salt waste)	Electrochemical	Glass-bonded zeolite (Echem)	HLW-Repository
Cs/Sr	UREX+1	Mineralized monolith	MLLW-Decay Storage or HLW-Repository
Lanthanides	UREX+1	Glass	HLW-Repository
Cladding/coatings	Aqueous, Melt Refining, and AirOx	Compacted	HLW-Repository
Structure/hardware	All	Compacted (Remainder Metal)	HLW-Repository
Spent solvents-liquids (not yet in model)	UREX+1	Mineralized monolith	LLW
Fuel fabrication wastes	---	Compact	LLW GTCC
Operations and maintenance wastes (not yet in model)	All	Mineralized monolith (combustible wastes only, after combustion)	LLW or LLW GTCC, depending on TRU contamination

Table 2 illustrates the set of reactor and separations choices in FIT that were utilized in this study. Indicated are the type of reactor and corresponding fuel, the conversion ratio, and the age of the initial LWR UOX feedstock.

Table 2: The four representative fuel cycle cases considered.

Case	Separation 1	Reactor	Feedstock age (yr)	Separation 2
1	UREX+1a	metal fast reactor (CR=0.5)	5	Electrochemical
2	UREX+1a	oxide fast reactor (CR=0.5)	5	UREX+1a
3	PUREX	MOX	5	N/A (only 1 cycle)
4	PUREX	MOX	50	N/A (only 1 cycle)

These cases represent a standard sample set of possible fuel cycles alternative to the current open fuel cycle in the United States. Cases 1 and 2 represent two versions of a closed fuel cycle with 18 recycle loops, whereas cases 3 and 4 represent a modified open fuel cycle with one recycle into a MOX reactor, similar to the current French fuel cycle. For these MOX cases, two different feedstock cooling times, 5 years and 50 years, were compared. The 50-year-old feedstock case represents the option of processing legacy used fuel, and explores the impacts of decades long cooling of used fuel.

RESULTS AND DISCUSSION

Waste heat generation rate is the main metric of interest when attempting to make a first-order classification judgment. Since a geologic repository is ultimately necessary independent of fuel cycle choice, maximizing disposal capacity of the repository is a key issue. Disposal capacity directly correlates with heat load of emplaced waste packages; the peak near-field temperature determines the maximum heat loading of the waste package and the peak far-field peak temperature determines waste package spacing in the repository. The former is normally influenced by fission product heat load, and the latter is usually determined by heat-generating actinides. No thermal model was developed to model the heat profile of each package in its host geology, but it is still possible to make a deduction about repository impact based on the heat generation of each waste form meant to go into a package bound for the repository. Short-term fission product heat can be dealt with by decay storage, so it may be advisable to classify wastes according to which materials would benefit from interim storage.

For the cases presented in Table 2, the short- and long-term heat dose contribution from each resulting waste stream was evaluated. For relevant heat-contributing isotopes, a Bateman solution was used to calculate decay and account for ingrowth of the isotope and its decay chain. In the multi-recycle fast reactor cases 1 and 2, the heat generation rate from only the last recycle of the set of iterations is shown. This allows for a conservative estimate, as the heat contribution increases as the reactor approaches equilibrium after many recycles. All heat generation values are expressed in watts per kg of initial heavy metal (iHM), on the basis of the LWR-UOX fuel feed coming from a reactor with an assumed burnup of 51 MWth-day/kg-iHM, 1GW (electric) power output, a capacity factor of 0.9, and a thermal efficiency of 0.33.

Figure 2 shows the total decay heat from all the waste forms resulting from each fuel cycle case. All waste streams generated in the FIT model “[result] from the reprocessing of spent nuclear fuel” [6]; therefore, under the current legal framework, all waste streams should be classified as HLW. However, there are waste streams produced, most notably Cs/Sr, that may have the potential to significantly alleviate the short-term heat burden if classified, managed, and disposed of differently.

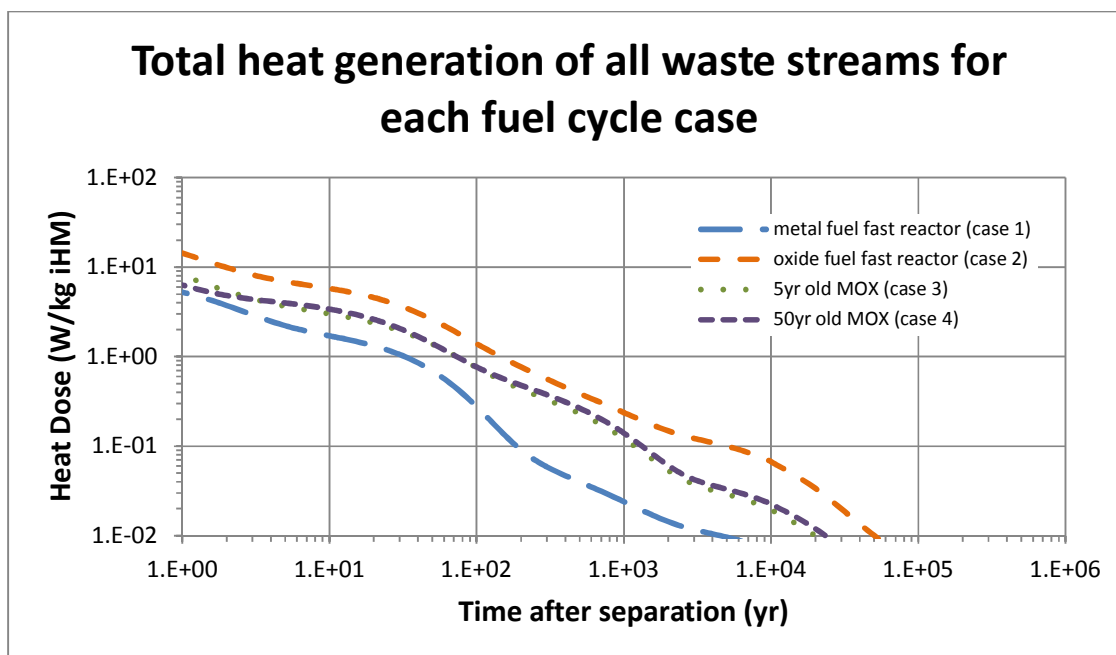


Figure 2: Comparison of heat contribution from all waste forms of cases 1-4. Total heat generation of all waste forms from fuel cycle if all waste streams were classified as HLW.

In Figure 2, it is important to note the curves show the total heat generation rate of the sum of each individual waste form for the last cycle of the set of iterations of each fuel cycle, normalized by the amount of initial heavy metal of the system. This includes the last discharged core of unprocessed used fuel, of which there is only one over the entire set of cycles; as well as Cs/Sr of the first aqueous reprocessing in the metal fuel case, of which there is also only one waste form per set of recycles. They do not yet show how many waste forms in total each fuel cycle produces; parametric assumptions about the fast and MOX reactors of the system are not made, and therefore the benefit of increased electricity generation with each recycle is not explicitly demonstrated in these graphs. The two fast reactor cases, which undergo multiple recycles, produce much larger cumulative electricity generation and thus have better uranium utilization over the lifetime of the fuel cycle. Expressing the metric in units of heat generation rate per MWh generated could show that the total heat generation from wastes in a fast reactor fuel cycle would be significantly lower than that of the MOX fuel cases.

Even so, it can be seen that older feedstock in a MOX recycle case contributes the greatest heat load in the very long term, which is consistent with a previous study [7]. This is due to the decay of fissile Pu-241 in the original spent fuel, with a half-life of 14.1 years, into Am-241, which is commonly the largest contributor to the long-term heat burden on a geologic repository.

Classifying all waste “resulting from the reprocessing of spent nuclear fuel” [7] as high-level

waste potentially neglects the waste management benefits that these fuel cycle options have to offer. One of the main values of classifying waste streams according to their differing characteristics lies in providing appropriate credit to fuel cycle options for the reduction of the heat burden on the repository and the radiotoxicity burden on the environment (on a per electricity generation basis) by storing and/or disposing of different waste streams appropriately.

Figures 3-6 illustrate a more detailed breakdown of heat-contributing waste streams for all fuel cycle cases. In each figure, the heat contribution from fission products (FP) and actinides (U+TRU) in each waste form is shown. Fission products and actinides will still be present in the same waste form, despite partitioning efforts, due to impurities and material losses during the separations processes, but in certain waste streams they may be negligible.

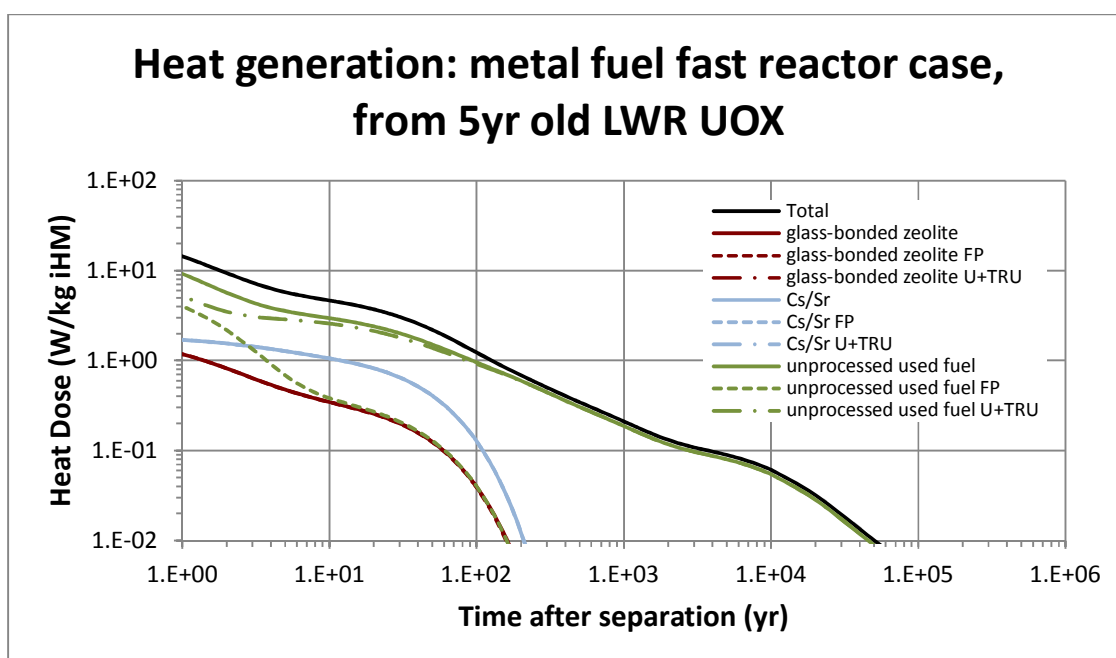


Figure 3: Comparison of heat generation from Cs/Sr, glass-bonded zeolite, unprocessed used fuel waste streams for metal fuel fast reactor case (case 1)

Again, it is important to reiterate that Figure 3 compares the heat generation rate of one individual waste form, and not the cumulative number of waste forms resulting from multiple recycles. Figure 3 shows that the highest heat generation rate is carried by the unprocessed used fuel bundle of the last recycle. For this waste form, decay storage would offer little benefit to reducing the near-field peak temperature of the repository, as its heat burden is mostly generated by actinides. In contrast, the heat curves for the glass-bonded zeolite (the main waste form for fission products from electrochemical separation) and Cs/Sr (from the first UREX+1a separation) waste forms indicate that surface decay storage may be beneficial in terms of repository heat management. The

main short-term heat load is carried by the isotopes Cs-137 and Sr-90, which decay below the heat dose level of the actinide impurities in the glass-bonded zeolite waste form after about 350 years and in the Cs/Sr waste form after about 450 years. After this timeframe, heat-generating actinides—mainly Am-241 from Pu-241 decay and Pu-238—drives the long-term far-field peak heat effects. Decay storage only makes sense for as long as the heat load of a waste form is dominated by short-term fission product heat, so the question thus becomes whether waiting for 350-450 years for fission products to decay is a reasonable timeframe for surface storage. The separated Cs/Sr was calculated to decay to LLW Class C levels after about 320 years, applying the concentration limit rules of 10 CFR 61 for Pu-241 (Table 1), Cs-137 and Sr-90 (Table 2) concentrations [2].

Figure 4 tells a similar story for the oxide fuel fast reactor case (case 2). In this case, all materials are separated using UREX+1a and immobilized in appropriate waste forms shown in Table 1. The heat generation rate of the three waste forms with highest heat load (HLW glass, Cs/Sr, and the unprocessed used fuel bundle) is shown below.

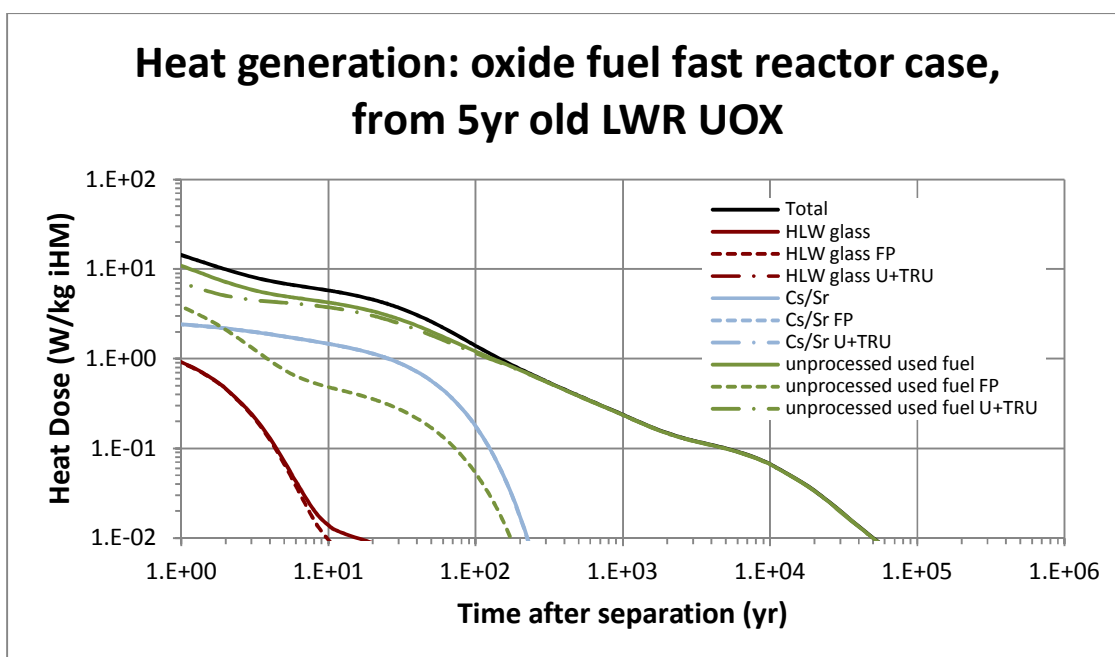


Figure 4: Comparison of heat generation from Cs/Sr, HLW glass, unprocessed used fuel waste streams for oxide fuel fast reactor case (case 2)

The heat decay profile of the oxide fuel fast reactor case in Figure 4 is similar to that of the metal fuel fast reactor case (Figure 3), showing that the main heat contributing waste form is the unprocessed spent fuel bundle discharged at the end of multiple recycles. In the Cs/Sr waste form, the fission product heat load falls below that of the actinides in the same waste form after about

350 years. In the case of HLW glass, this time point happens at about 50 years; thus, decay storage of several decades of HLW glass waste forms in this oxide fast reactor case may be an acceptable way of reducing the short-term heat burden and thus the near-field peak heat effects on the repository. Further purifying either of these waste streams by increasing the actinide recovery fraction could make long-term heat management of the repository even easier. As in the previous case, the separated Cs/Sr was found to decay to LLW Class C levels after about 320 years [2], after which near-surface burial would be acceptable.

Figures 5 and 6 show the heat load carried by wastes produced from the two MOX recycle cases.

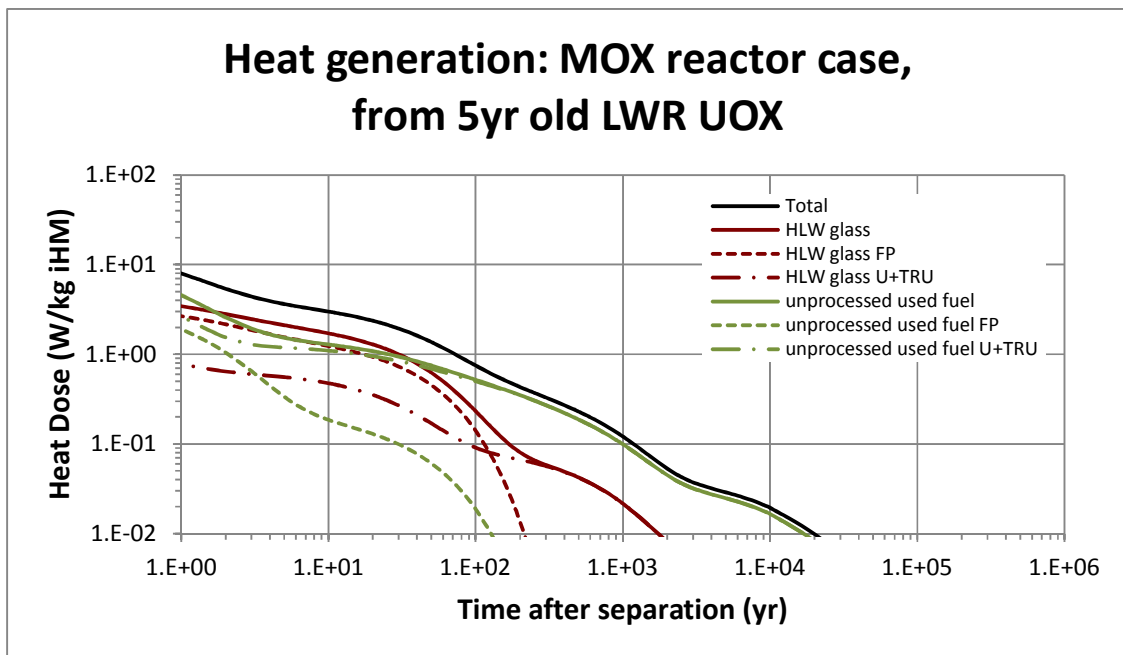


Figure 5: Comparison of heat generation from HLW glass and unprocessed used fuel for 5-year-old feedstock MOX recycle case (case 3)

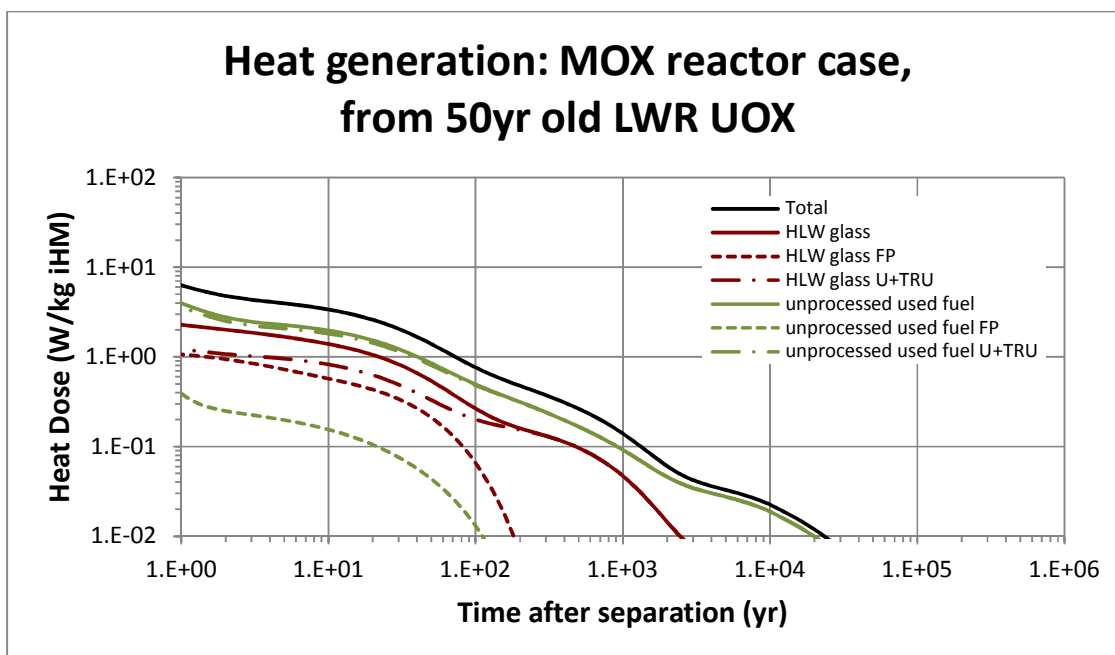


Figure 6: Comparison of heat generation from HLW glass and unprocessed used fuel for 50-year-old feedstock MOX recycle case (case 4)

Figures 5 and 6 show that the total heat load of both HLW glass and unprocessed used fuel for MOX-recycle cases are comparable. For the 5-year-old feedstock case, the heat generation from fission products dominates for the first 125 years until the actinide heat contribution takes over; for the 50-year-old feedstock case, the heat load is dominated by actinides (in both waste forms) from the beginning. In both cases, the total heat generation rate drops in half after about 450 years, which is when all the Cs and Sr have decayed to negligible levels and approximately half of the original Pu-241 and Am-241 have decayed. Whether there is a benefit to Cs/Sr separation for a MOX case is questionable; the idea of a 450-year-long surface storage institution, since it would need to be institutionally monitored for a very long period of time, may not be acceptable. Separation of Cs/Sr only makes sense if a separations process was used where actinides need to be recovered for utilization in a reactor, as in cases 1 and 2. At least, it would be beneficial to store the high-level wastes resulting from a MOX recycle case above ground for some reasonable temporary period to reduce the heat load before emplacement in the repository.

CONCLUSION

Under the current radioactive waste classification framework, all separated materials from advanced fuel cycles are classified as high-level waste, therefore neglecting benefits of separation from the waste management perspective. However, some of these separated wastes could benefit from interim storage to increase repository capacity (and thus reduce repository cost), and

changing the class of these wastes appropriately would better inform the heat management strategy of a waste disposition regime. The results of this study indicate the possibility to use repository capacity as a function of decay heat management as a metric for waste classification.

Interim decay storage for Cs/Sr (separated in the UREX+1a process) would result in either decaying to LLW levels after about three centuries (and go to a near-surface disposal if reclassified as such), or, in shorter decay storage timeframes, reducing the near-field peak temperature of the repository. This decision would depend on whether it is considered reasonable, socially stable, and cost-effective to operate a surface storage institution for 300 years. In the case of HLW glass form from an oxide fuel fast reactor case, it seems sensible to decay the waste form for several decades (a more reasonable timeframe), when the fission product-driven heat load falls below the heat dose contribution of the actinides.

If Cs/Sr separation is not necessarily an option, as in the MOX recycle cases, the results of this study indicate that some decay storage to let fission products decay to reduce the heat load is nevertheless beneficial for increased repository capacity. However, the high actinide heat load of MOX fuel reduces the benefits provided by surface storage, and storage for sufficient time for actinide heat load to drop significantly would be difficult, costly and impractical. Also, it was shown that the long-term heat burden is slightly smaller if fresher UOX feedstock is used to produce MOX, so from this perspective it is prudent to recycle UOX feedstock into MOX fuel as soon as possible.

It is important to point out that there will always be a need for a geologic repository even in the case of recycling and transmuting transuranics. A final used fuel bundle from the last reactor core as well as structural material losses and undissolved solids will always exist, no matter how many fast reactor recycle loops are performed.

Suggestions for future work include a more robust quantification of repository impact (high/medium/low), evaluating the heat generation impact of structural material contamination, and the comparison of heat generation of waste forms normalized to the amount of electricity generated. Also, a parametric study should be performed to explore the sensitivity of waste metrics such as heat dose rate due to varying fuel cycle parameters. For example, it would be good to assess the impact on heat load due to varying actinide contamination of different HLW waste streams. It is also relevant to compare the impact of heat load if Cs/Sr is left in the HLW glass waste stream (potentially eliminating some costs associated with separation and long-term above-surface monitoring).

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