

Radiotoxicity Characterization of HLW from Reprocessing of Uranium-Based and Thorium- Based Fuel- 11390

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ABSTRACT

The growing inventory of spent nuclear fuel generated in the current “open cycle” adopted in the US can hamper the long term growth of nuclear energy. A proper strategy for closing the fuel cycle and generating “acceptable waste” should hence be a priority of the nuclear industry. While satisfactory technological solutions exist addressing portions of the overall problem, a fully integrated effective solution satisfying all public concerns has yet to be developed. Westinghouse is proposing a new approach, which involves redefining the specifics of the main components involved (fuel form, reactor type and reprocessing technique) so that the nuclear system overall generates wastes which are not perceived as a hazard by the public and with reduced requirements on their disposal.

In a closed fuel cycle, the actinides in the spent fuel are typically recovered by reprocessing and recycled in a combination of reactors. Although the transmutation rate of actinides depends on the specific properties of the system adopted for transmutation (e.g. fast reactors, accelerator-driven systems etc), the final waste will be predominantly characterized by the reprocessing plants. The efficiency that can be achieved in the separation and recovery of some critical isotopes is a key to reduce the high-level waste radiotoxic content. This paper focuses on identifying these critical isotopes and their recovery ratio needed to produce an acceptable waste. Both the conventional uranium-based fuel cycle and a proposed alternative, the thorium-based fuel cycle are investigated.

INTRODUCTION

Commercial nuclear reactors in the US provide approximately 20.2% of the nation’s electric power generation, producing nearly 800 TWh in 2009 [1]. Although nuclear energy is regarded as a clean energy since no pollutants or greenhouse gases are emitted from a nuclear plant, it results in the generation of radioactivity. This radioactivity is effectively confined in wastes which are categorized according to their hazard (low-level waste, high-level waste and so on) and disposed accordingly. High-level waste (HLW) needs to be isolated from the environment for geological times through confinement in costly repositories. The capability of the barriers interposed in the repository to the release of the radiotoxic pollutants is often questioned by part of the public opinion, reluctant to trust any engineered or geological barrier supposed to resist over very long periods of time, e.g. hundreds of thousands of years. While the rationality of such a frame of mind can be debatable, the resistance from the public will inevitably lead to delaying, if not preventing, the construction of a permanent repository.

The current US policy allowing only “once through” fuel cycle for commercial nuclear plants exacerbates the problem of used nuclear fuel (UNF) disposal. As a result of this policy all the UNF discharged from a reactor (typically ~20 metric tons of UNF/GWe-yr) is effectively a HLW. Over the past four decades, the US nuclear industry has thus cumulated ~60,000 metric tons (MT) of UNF, increasing at a rate of ~2,000 MT UNF per year. This large and increasing volume of wastes is currently stored on site, in wait of permanent disposal in a yet-to-be built repository.

The projected growth of the nuclear industry may be hampered under these premises: the favorable framework for the nuclear energy favored by rising environmental concerns related to use of fossil fuels risks to be negated by what is perceived as the lack of an adequate solution for the backend of the fuel cycle.

Westinghouse’s primary goal is thus to devise a system (fuel, reactors, reprocessing) which generates HLW which are “acceptable” from the public perspective. The first challenge in this approach is conceptual. “Acceptability” relates to one’s perception and has therefore an intrinsic degree of variability. Still “acceptability” needs to be translated into a set of metrics and specifics in order to measure the compliance of the system with the objective proposed.

We will devote the next section to the definition of these metrics. We will then focus on the specifics for two representative fuel cycles, the uranium and thorium cycle, and show their potential to meet the objective. The conclusions from this comparison are the guide for the development of a system producing acceptable HLW.

METRICS

We propose to define “acceptable” HLW in relative terms, based on the selection of a figure of merit to characterize the HLW and its performance against an “equivalent amount of a reference material” at a “reference time”. The figure of merit that we select to characterize HLW is radiotoxicity. The reference material is freshly mined natural uranium and the reference time for the comparison is 300 years after the generation of the HLW. The amount of freshly mined natural uranium is calculated based on the fuel provision for typical PWR operation (open-cycle, UOX fuel, at typical discharge burnup and U-235 enrichment), which translates into ~210 metric tons of freshly mined natural uranium per GW-yr. The HLW radiotoxicity is compared against the reference open cycle on a per-energy generated basis, which ensures a fair assessment of systems promoting recovery and multiple recycle of spent fuel, thereby requiring very low amount of freshly mined natural uranium. For these systems, the HLW will consist predominantly of the losses from the fuel reprocessing.

Implicit in the definition of the above metrics is the regard of HLW as a product that needs to fulfill certain expectations to be acceptable to the general public. These expectations go beyond what would be regarded as “safe” from a purely engineering perspective. People are more inclined to cope with and accept metrics expressed in relative terms and over times comparable with their generational span. This, more than their technical merit, is the logic underlying the choice of the “300 years” as time-frame, of radiotoxicity as a figure of merit, and of freshly mined natural uranium as term of comparison for the determination of an acceptable HLW.

Radiotoxicity definition

Radiotoxicity is a measure of the potential biological hazard of a nuclear material based on its radioactive quality rather than its chemical toxicity or radioactivity alone. Radiotoxicity is commonly expressed in “Sieverts”, ingested or inhaled. The ingested radiotoxicity in Sieverts (Sv) for a given isotope is determined by the activity (Bq) multiplied by the isotope effective dose coefficient, $e(T)$, for ingestion (similarly, but with a different coefficient, for inhalation). The effective dose coefficient (units Sv/Bq) accounts for radiation and tissue weighting factors, metabolic and biokinetic information of an adult based on ICRP 72. [2] Equation 1 shows the relationship between radiotoxicity, activity and the effective dose coefficient [3].

$$\text{Radiotoxicity [Sv]} = \text{Activity [Bq]} * e(T)[\text{Sv/Bq}] \quad (\text{Eq.1})$$

The radiotoxicity of a nuclide can also be expressed, as “m³ water” for ingestion or “m³ air” for inhalation. This radiotoxicity in volume corresponds to the amount of water or air needed to dilute the radioactive material potential intake by ingestion or inhalation below the threshold for the general public (50 mrem/year, based on average intakes of 2 litres/day of water or 22 m³/day of air).

Note that all the radiotoxicity values used in this paper are calculated as ingested radiotoxicity to maintain consistency and expressed in relative terms, i.e. divided by the ingested radiotoxicity of the equivalent freshly mined natural uranium as defined above.

The “300-year” goal

Figure 1 shows the relative radiotoxicity of UNF from standard PWR operation, open-cycle, at typical enrichment and discharge burnup (top-curve). For the first 100 years radiotoxicity is primarily dominated by fission products. From 100 to 700 years the radiotoxicity of the fission products decreases sharply due to their natural decay and the contribution from the actinides becomes predominant. Actinides and then their progeny going to secular equilibrium dominate the intermediate and long term radiological impact. Without separation UNF remains radiotoxic for several thousands of years (~300,000 years) before decaying to the same radiotoxicity level as the equivalent freshly mined uranium. The integrity of properly engineered and geological barriers will need to be ensured for an accordingly long period of time.

Figure 1 shows also that if all TRUs could be separated and recycled, leaving only the fission products to the HLW, the radiotoxicity could be reduced to that of the corresponding natural uranium ore in slightly below 300

years. Thus efficient separation and recovery of actinides from reprocessing, together with a viable strategy for in-core transmutation, is essential to achieving the “300-year” objective. The metrics we use to assess the elemental or isotopic recovery of a given separation process is the “recovery ratio” and is discussed next.

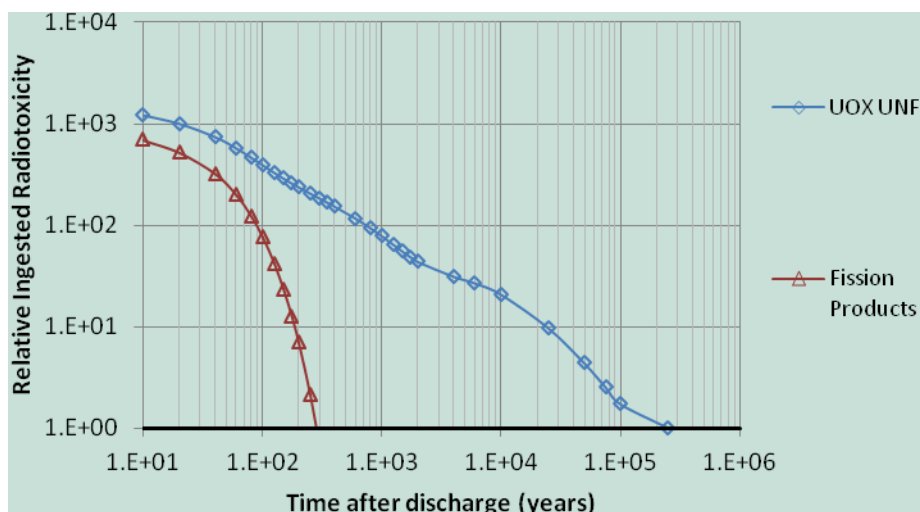


Figure 1: Relative radiotoxicity of UOX UNF and contribution from the fission products

Recovery ratios from reprocessing

Due to variation in chemical properties, specific activity and heat generation the separation efficiencies achievable for various elements will be different and, in many cases, not conclusively defined. The reprocessing technique that is selected here as indicative of the current state-of-the-art separation efficiency is the UREX+ suite, and in particular the UREX+3 [4], with the recovery ratios given in Table II.

Table I: Recovery ratios for an UREX+3 reprocessing plant [4]

UREX +3	
Element separated	Recovery ratios
U	0.9995
Pu	0.995
Am	0.98
Cm	0.79
Np	0.71
Cs	0.96
Sr	0.99
Tc	0.95
Lanthanides	-
Rare Earths	0.272

UREX+3 is a comprehensive reprocessing technology developed by ANL (Argonne National Laboratory) that efficiently separates a wide range of actinides. UREX+ has only been conducted at the laboratory scale. The process however appears adaptable to an industrial process. The reason is that UREX+ is essentially an extension of PUREX, which has gained substantial industrial maturity for reprocessing and separation of plutonium and uranium. A lower efficiency for U and Pu should be expected, at least until sufficient technological maturity is established, for the recovery of Minor Actinides (MA), e.g. Am, Cm and Np (see recovery ratios in Table 1). The short term higher radiotoxicity and decay heat contributors, Cs and Sr, both having half lives of ~30 years can also be efficiently

recovered. Sequestration of these very active isotopes will ease fuel handling and disposal. A possible strategy for Cs, Sr is interim on-site storage and future final disposal as HLW after their radiotoxicity has reduced to the acceptable level. However, we will not deal here with the treatment of fission products as only actinides recovery and recycle is essential to meet the proposed 300-year objective.

We calculated the HLW radiotoxicity from the reprocessing of UOX UNF using the UREX+3 recovery ratios in Table 1 through SCALE depletion calculations of LWR UOX and ORIGEN decay calculations on the waste stream [6,8]. The results are shown in Figure 2, together with a comparison to open-cycle (non-reprocessed) UOX UNF and waste stream from the fluoride volatility process [5].

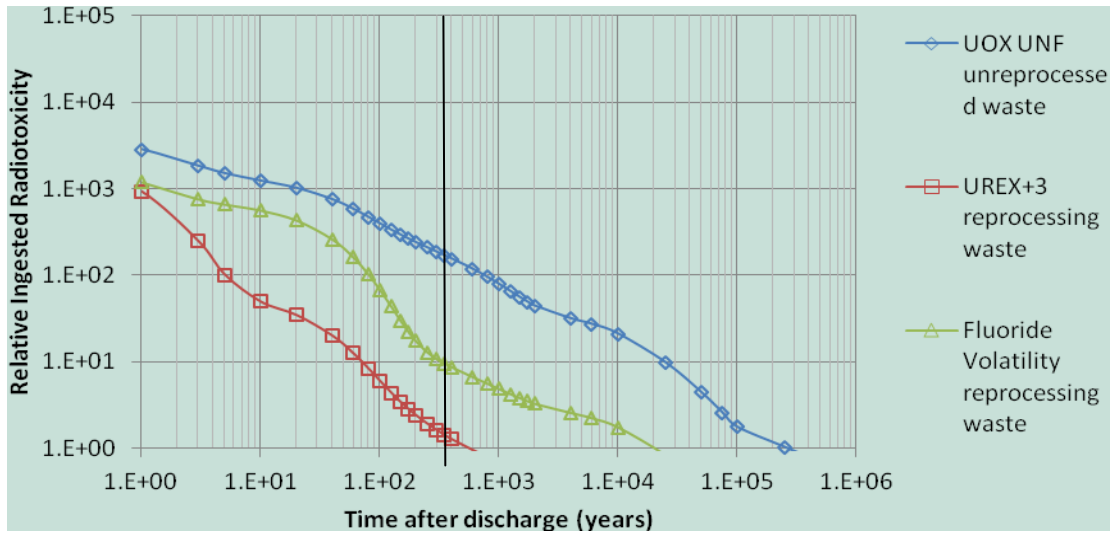


Figure 2: Relative Radiotoxicity from reprocessing of UOX UNF with UREX+3 and fluoride volatility.

Figure 2 shows that the radiotoxicity of the waste stream from the UREX+3 process decreases to that of the equivalent amount of uranium ore in ~300 years. As a counter example, the radiotoxicity from the fluoride volatility separation technique does not meet the 300-year waste criteria. The main reason is that fluoride volatility does not separate MAs from the wastes as the process targets mostly fission products (beside U and Pu). Table II shows the fluoride volatility recovery ratios assumed. Fluoride volatility however has other advantages over aqueous processes such as low risk of criticality due to the dry nature of the process and does not contribute to high volume of liquid waste unlike aqueous processes. Even though the waste from fluoride volatility does not meet the 300 year objective, it can be paired up with other simple reprocessing techniques to translate its radiotoxicity plot to the left in order to comply with our '300 year' waste criteria.

Table II: Recovery Ratios for a Fluoride Volatility reprocessing plant [5].

Fluoride Volatility	
Element	Recovery Ratio
U	0.99
Pu	0.99
H	1
C	1
Kr	1
I	1
Cs	0.98
Tc	1
Ru	1
Rh	0.8

Te	0.9
Mo	0.8

As a next step we focus our analysis on the waste characterization of current, uranium-based fuel vs. a possible alternative: thorium-based fuel. The specifics for UNF reprocessing and actinide recovery to meet the “300-year objective” for both options will be discussed. The in-core transmutation behavior of the recycled fuel is investigated in ongoing work and will be reported in the future.

WASTE ANALYSIS FOR THORIUM- VS. URANIUM-BASED FUELS

Thorium-based vs. uranium-based fuels have been considered and analyzed focusing on their potential impact on HLW radiotoxicity. The implications on the front-end, economics, in-core physics, fuel management, etc. of switching from a predominant uranium-based fuel cycle to a possible thorium alternative are not discussed.

In the perspective of HLW radiotoxicity reduction, the major benefit of the thorium fuel cycle is that it leads to the generation of substantially fewer transuranics (TRU) with respect to the uranium cycle, especially when fertile thorium is admixed only with fissile U-233. The reason is the “lower” position of Th-232 and U-233 in the transmutation isotopic chain leading to TRU generation vs. typical uranium fuel (U-238 fertile with U-235 fissile). Note however that other non-TRU isotopes are generated in irradiated thorium-based fuel. These isotopes contribute significantly to the long-term radiotoxicity from thorium UNF and associated reprocessing wastes. The major radiotoxic contributors and their behavior over different timelines will be discussed in the following sections.

Characterization of UNF radiotoxicity for thorium and uranium in the LWR open cycle

Irradiated fuel isotopic composition for a typical 17x17 Westinghouse 4-loop PWR has been calculated using the SCALE 6 [6] package for both a UOX and ThOX fuel assembly. We refer ThOX as an oxide fuel which has U-233 as fissile material and Th-232 as fertile material. For this analysis UOX fuel has 4.3 w/o U-235 enrichment. ThOX has Th-232 with 4.3 w/o U-233. The detailed mass flow at charge and discharge is shown in

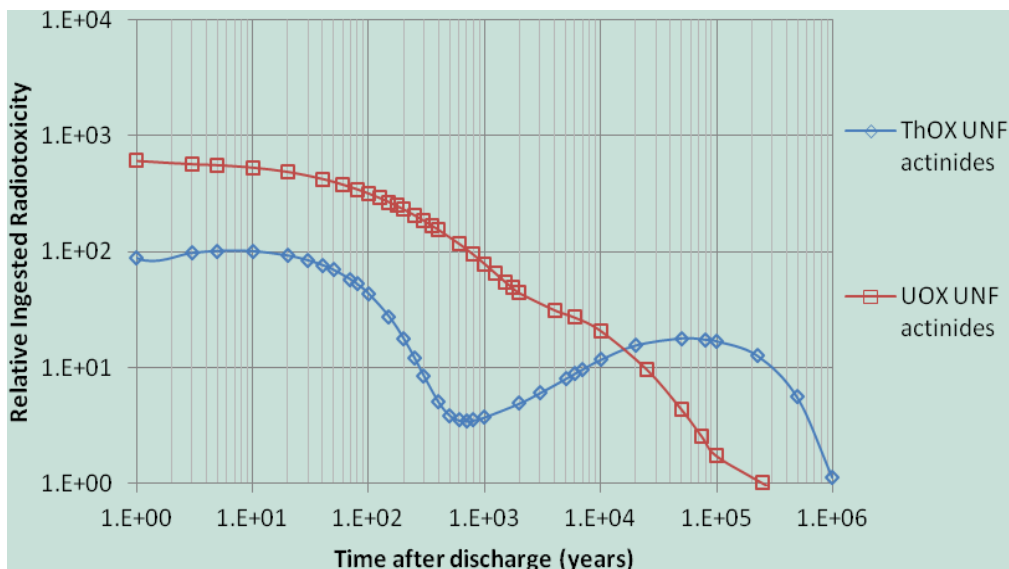


Figure 3: Radiotoxicity of discharged actinides for once through UOX and ThOX fuel assemblies

Decay calculations have then been performed with ORIGEN-ARP [8] to determine the spent fuel overall radiotoxicity as well as the critical contributors (progeny and parent) at the various time ranges for both UOX and ThOX UNF. The radiotoxicity versus time for UNF UOX and ThOX is shown in Figure 3. Note that only the radiotoxicity due to actinides at discharge and their progeny through decay is plotted. Fission products are not considered as they play an insignificant role to achieving our objective. Figure 3 shows that the actinide radiotoxicity for ThOX is notably lower than that of UOX UNF in the 10-10,000 years range. The radiotoxicity for ThOX UNF reaches a minimum at ~1,000 years and then increases following buildup of highly radiotoxic Th-229

from alpha decay of U-233. Due to this behavior, at ~20,000 years the radiotoxicity of UNF ThOX becomes greater than that of UOX. At ~ 100,000 years the ThOX radiotoxicity is finally decreasing.

It should be noted that the spike in ThOX radiotoxicity in the time-range between ten and hundred thousand years could be reduced, or eliminated, by efficiently recovering the U-233 in the reprocessed ThOX fuel, or by burning it more completely (by fission) before the fuel final disposal. (An efficient recovery of U-233 from ThOX UNF is of course a mandatory prerequisite for an effective breeding strategy). In light of the above, the increase in radiotoxicity due to the U-233 decay chain in ThOX in the ten to hundred thousand years range will likely not be manifest once an effective recovery and recycle strategy is devised (as shown further on in this study). The radiotoxicity calculations for both UOX and ThOX UNF is based on the isotope vectors for each fuel assemblies as shown in Table III.

Table . Note that the lower density of thorium results in a higher average discharge burnup to yield the same cycle length of UOX, i.e. 60 vs. 52.5 GWd/tIHM (Gigawatt day/ton of initial Heavy Metal). The assumptions and results for this study are in line with what reported by Todosow in [7]. In particular, the amount of TRUs generated in ThOX fuel is 10E-4 (Pu) to 10E-6 (Am and Cm respectively) magnitude smaller than in UOX.

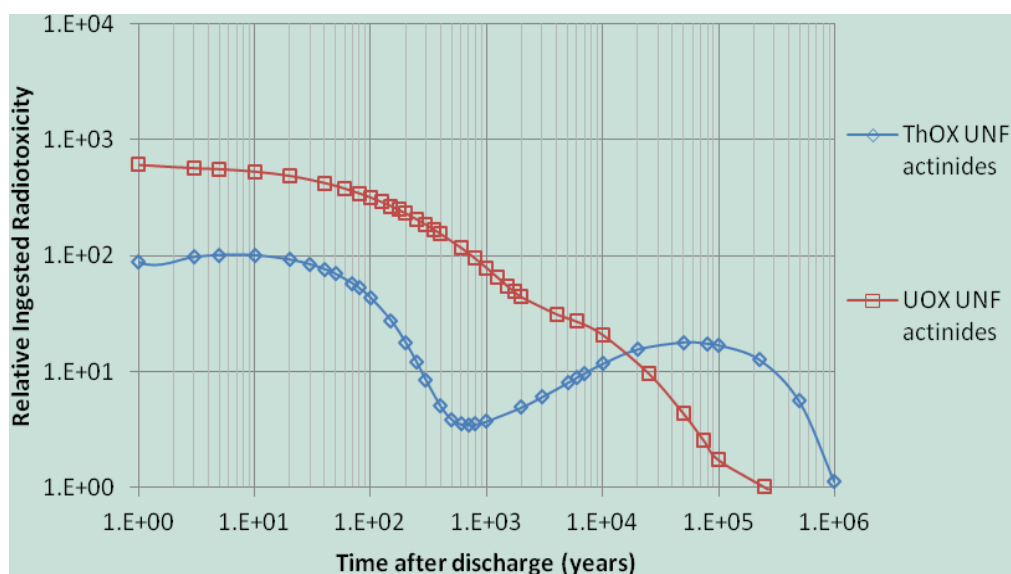


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Table III: Mass flow for UOX and ThOX fuel assembly study

	4.3 w/o of U235 in U		4.3 w/o of U-233 in Th	
Discharge burnup	52.5 GWd/tIHM		60 GWd/tIHM	
isotope	charge	discharge	charge	discharge
Th-232		2.11E-04	4.03E+02	3.87E+02
Pa-231		2.90E-04		3.26E-02
Pa-233		1.36E-05		5.31E-01
U-232		4.49E-04		3.26E-02
U-233		1.21E-03	1.81E+01	7.64E+00
U-234	1.77E-01	7.20E+01		2.21E+00
U-235	2.01E+01	2.31E+03		4.37E-01
U-236		2.82E+03		7.36E-02
U-237		7.23E+00		1.67E-04
U-238	4.47E+02	4.27E+05		6.76E-05
Np-237		3.97E+02		4.61E-03
Pu-238		2.11E+02		1.01E-03
Pu-239		2.82E+03		1.29E-04
Pu-240		1.47E+03		2.58E-05
Pu-241		9.02E+02		1.66E-05
Pu-242		5.75E+02		3.59E-06
Am-241		2.84E+01		2.37E-07
Am-243		1.84E+02		4.44E-09
Cm-242		1.35E+01		5.99E-08
Cm-244		9.59E+01		7.48E-08
Total U	4.67E+02	4.32E+02	1.81E+01	1.04E+01
Total Pu		5.98E+00		1.18E-03
Total Am		2.13E-01		6.89E-07
Total Cm		1.16E-01		1.39E-07

The primary contributors to radiotoxicity of UOX and ThOX UNF are shown in Table IV and Table V respectively together with their percentage contribution to the total radiotoxicity. The primary isotopes responsible for the radiotoxicity of UOX UNF are well known from literature. Therefore we will focus our attention on the isotopes of ThOX UNF.

Table V shows that the main contributors to the long-term radiotoxicity of ThOX UNF are attributable to the generation and decay of U-233. U-233 is the main contributor in the 1,000-year range. Its decay product, Th-229, becomes predominant in the range between 10,000 and 100,000 years (the half life of U-233 is ~160,000 years). The largest contributor in the million year range is Po-210, a progeny of U-234 (U-234 in thorium is mostly produced by neutron capture in U-233). Most of the largest contributors belong to the same decay chain, i.e. they originate by U-234. Hence increasing the destruction by fission in U-233 will positively impact the residual radiotoxicity of ThOX UNF. The largest benefit in the wastes radiotoxicity from recycle should be expected by maximizing the U recovery ratio.

The last column of

Table V shows the total radiotoxicity of ThOX vs. UOX. The reduced amount of TRUs in ThOX fosters radiotoxicity up to 40 times lower than in UOX UNF between 100 and 1,000 years. In the 10,000 years range the radiotoxicity of ThOX is still one half of that in UOX, before the previously noted increasing trend in ThOX prevails.

The specifics (e.g. isotopic recovery ratios) required to meet the radiotoxicity goal for the HLW generated from thorium-based and uranium-based used fuel vectors are discussed in the next section.

Table IV: Percentage contribution to total radiotoxicity at various times after discharge of UOX UNF

Time after discharge (years)									
100		1000		10,000		100,000		1,000,000	
Am-241	39.5%	Am-241	50.3%	Pu-239	49.8%	Pu-239	45.8%	Th-229	25.6%
Pu-238	29.6%	Pu-240	30.3%	Pu-240	44.3%	Po-210	19.9%	Po-210	25.1%
Sr-90	9.9%	Pu-239	16.4%	Am-243	4.1%	Pb-210	11.5%	Pb-210	14.4%
Cs-137	8.2%	Am-243	2.5%	Pu-242	0.8%	Pu-242	7.6%	Pu-242	6.1%
Pu-240	6.2%	Pu-242	0.2%	Po-210	0.2%	Ra-226	4.6%	Ra-226	5.8%
Pu-239	3.1%	Pu-238	0.1%	U-234	0.1%	Th-230	3.4%	Ra-225	5.2%
Cm-244	1.4%	Cm-245	0.1%	Pb-210	0.1%	Th-229	2.4%	Th-230	4.4%

Table V: Percentage contribution to total radiotoxicity at various times after discharge of ThOX UNF

Time after discharge (years)									
100		1000		10,000		100,000		1,000,000	
U-232	64.1%	U-233	34.0%	Th-229	59.9%	Th-229	52.1%	Po-210	39.3%
Th-228	14.4%	Th-229	27.5%	Ra-225	12.1%	Po-210	14.0%	Pb-210	22.6%
Ra-224	13.0%	Ac-227	14.1%	U-233	10.5%	Ra-225	10.5%	Th-229	14.5%
U-233	2.9%	Pa-231	9.1%	Ac-227	3.7%	Pb-210	8.0%	Ra-226	9.2%
Pu-238	1.3%	U-234	5.7%	Po-210	3.0%	U-233	5.2%	Th-230	6.9%
Pb-212	1.2%	Ra-225	5.6%	Ac-225	2.9%	Ra-226	3.3%	Ra-225	2.9%
Ac-227	1.2%	Ac-225	1.4%	Pa-231	2.4%	Ac-225	2.6%	U-233	1.4%
UOX/ThOX	0.10		0.04		0.52		9.10		2.56

Required recovery ratio for reprocessing of thorium and uranium UNF to meet the 300-year waste criteria

In a closed fuel cycle, although the transmutation rate of actinides depends on the specific properties of the systems adopted for transmutation (e.g. fast reactors, accelerator-driven systems etc), the final waste will be predominantly characterized by the reprocessing plants. The efficiency that can be achieved in the separation and recovery of some critical isotopes is a key to reduce the HLW radiotoxic content and meet the 300-year objective.

This section shows the potential HLW wastes and associated radiotoxicity from the reprocessing of uranium-based and thorium-based fuel. In particular, HLW from the reprocessing of the following vectors are considered:

- UOX, i.e. oxide fuel with U-235 fissile and U-238 fertile
- ThOX, i.e. oxide fuel with U-233 fissile and Th-232 fertile
- UMOX RG- Pu, i.e. oxide fuel with U tails admixed with Reactor Grade (RG) Pu
- TMOX RG-Pu, i.e. oxide fuel with Th-232 admixed with Reactor Grade (RG) Pu
- UMOX WG- Pu, i.e. oxide fuel with U tails admixed with Weapon Grade (WG) Pu
- TMOX WG-Pu, i.e. oxide fuel with Th-232 admixed with Weapon Grade (WG) Pu

All results have been produced based on unit assembly depletion calculations performed with SCALE, and using ORIGEN to perform the out-of-core decay and HLW radiotoxicity calculations.

UOX vs. ThOX Waste Radiotoxicity

Figures 4 and 5 show the radiotoxicity for the reprocessing wastes of both UOX and ThOX fuel assuming various actinide recovery ratios. These plots show the separation efficiency needed to generate an acceptable waste, i.e. waste that meets the 300-year goal. For meeting this criteria the wastes relative radiotoxicity must be below 1, (solid horizontal line in the figures) at and after the 300-year mark (solid vertical line in the plots).

For this preliminary analysis we assumed comparable separation efficiencies for the reprocessing streams of UOX and ThOX UNF. For UOX UNF, TRUs (Pu, Np, Am and Cm) need to be recovered at a very high recovery ratio (> 99 %) in order to generate waste that complies with the 300-year goal. On the contrary, ThOX UNF requires a comparatively smaller efficiency (99%) for fewer key elements (U and Pa). This is true under the assumption of having unlimited top-up U-233 available, due to the reduced TRUs generation in this case. As we will see T-MOX fuel (Th-232 admixed with Pu) will require efficient recovery over all the range of actinides, including TRUs.

Keeping the above caveat in mind, it appears that while in theory the 300-year goal is achievable for both uranium- and thorium-based fuel, the latter fuel seems more promising from the perspective of relaxing some of the specifics on the reprocessing. However, this potential advantage can be lost once the actual reprocessing of UNF thorium-based fuel is undertaken due to practical constraints (thorium fuel is more difficult to dissolve due to high chemical stability, plus the reprocessing technology for uranium based fuel is better known and established than for thorium).

Figure 5 reveals that Pa recovery substantially lowers the residual radiotoxicity of the wastes from 100 years onwards. The relevant isotope in this case is Pa-231 which is highly toxic and alpha decays to Ac-227 with a half-life of 32,760 years. Due to the long half life of Pa and some of its progeny its contribution to radiotoxicity spans thousands of years. The efficient recovery of Pa from spent ThOX UNF and its destruction from the final fuel vector is therefore an important issue.

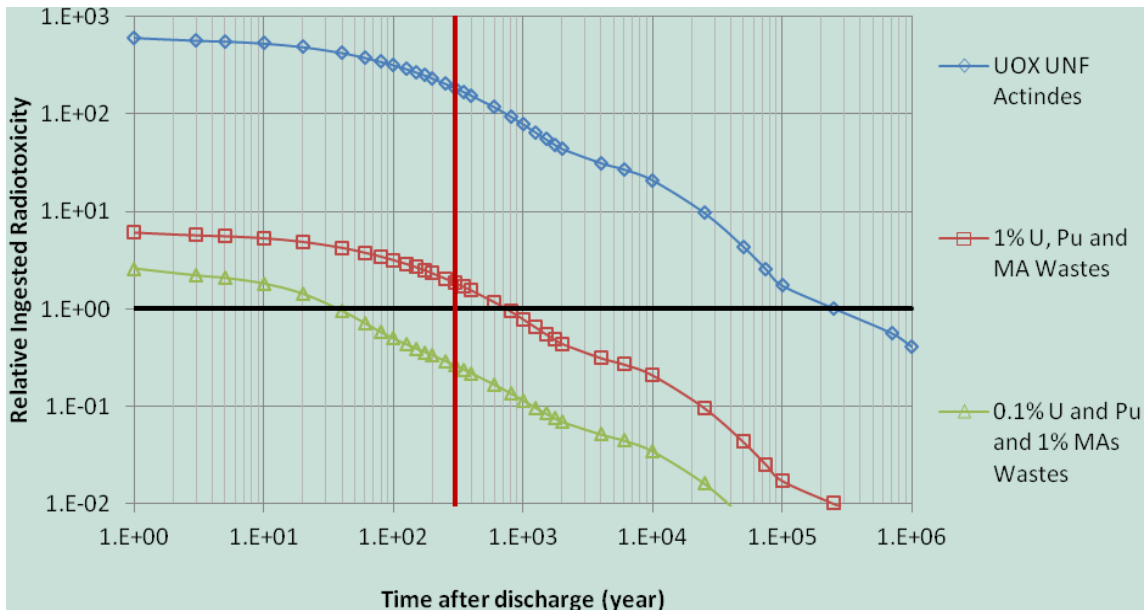


Figure 4: Radiotoxicity of waste streams at various recovery ratios for UOX UNF

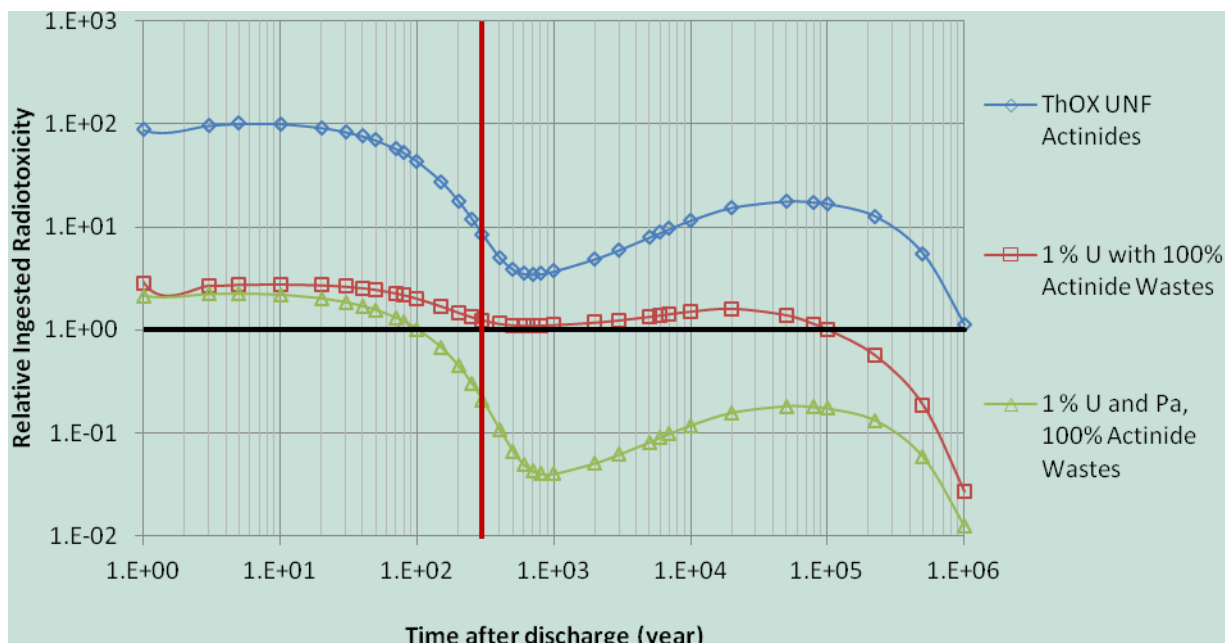


Figure 5: Radiotoxicity of waste streams at various recovery ratios for ThOX UNF.

U-MOX vs. T-MOX with reactor-grade and weapon-grade Pu

The radiotoxicity of U-MOX and T-MOX UNF and reprocessing wastes is compared in this section. Admixing Th with fissile Pu (or TRU) is one of the options to start-up a thorium-based fuel cycle economy and to make up for any deficiency in the amount of in-bred U-233. The isotopic vector for reactor-grade (RG) or weapon-grade (WG) Pu is shown in table VI. The RG-Pu vector is coming from separation and recovery of Pu from standard PWR UOX UNF under typical burnup conditions, yielding a ~65 w/o fissile Pu (54 w/o Pu-239 + 11 w/o Pu-241) in Pu. As known, WG-Pu coming from dismantled nuclear weapons is predominantly Pu-239 (94%) with a small amount of Pu-240 (6%).

Table VI: Isotopic composition of reactor-grade and weapon-grade Pu

	Pu-238 (%)	Pu-239 (%)	Pu-240 (%)	Pu-241 (%)	Pu-242 (%)
RG-Pu	2.6	54.0	25.3	10.8	7.3
WG-Pu	0.0	94.0	6.0	0.0	0.0

The methodology adopted to calculate the radiotoxicity of the U-MOX and T-MOX HLW wastes from reprocessing of discharged fuel is the same as that previously reported. The calculated relative radiotoxicity for each fuel type is reported in the Figure 6 and 7.

The radiotoxicity of U-MOX and T-MOX RG-Pu (and, similarly, WG-Pu) is similar up to ~20,000 years. This is unlike the UOX vs. ThOX comparison, where ThOX radiotoxicity in the intermediate to long-time range was shown to be substantially lower than in UOX (compare Figure 6 and Figure 3). The reason is that in T-MOX the initial Pu is directly or indirectly (by neutron capture and subsequent decay) responsible for most of the discharged fuel radiotoxic content. The negative impact on radiotoxicity due to further TRU fertilization in Pu can be seen comparing the radiotoxicity of U-MOX RG-Pu against U-MOX WG-Pu (see plots in Figure 7, the behavior for T-MOX RG Pu vs. T-MOX WG-Pu would be similar). More parasitic neutron captures take place in RG-Pu with respect to WG-Pu due to the degraded Pu vector characterizing the latter. Note also that T-MOX fuel radiotoxicity plot surpasses, and departs, from that of U-MOX in the interval between 10E+5 to 10E+6 years. This is the net

effect of the long-term spike in radiotoxic behavior previously discussed for ThOX combined with the general increase in radiotoxicity at lower times due to the presence of TRU in T-MOX.

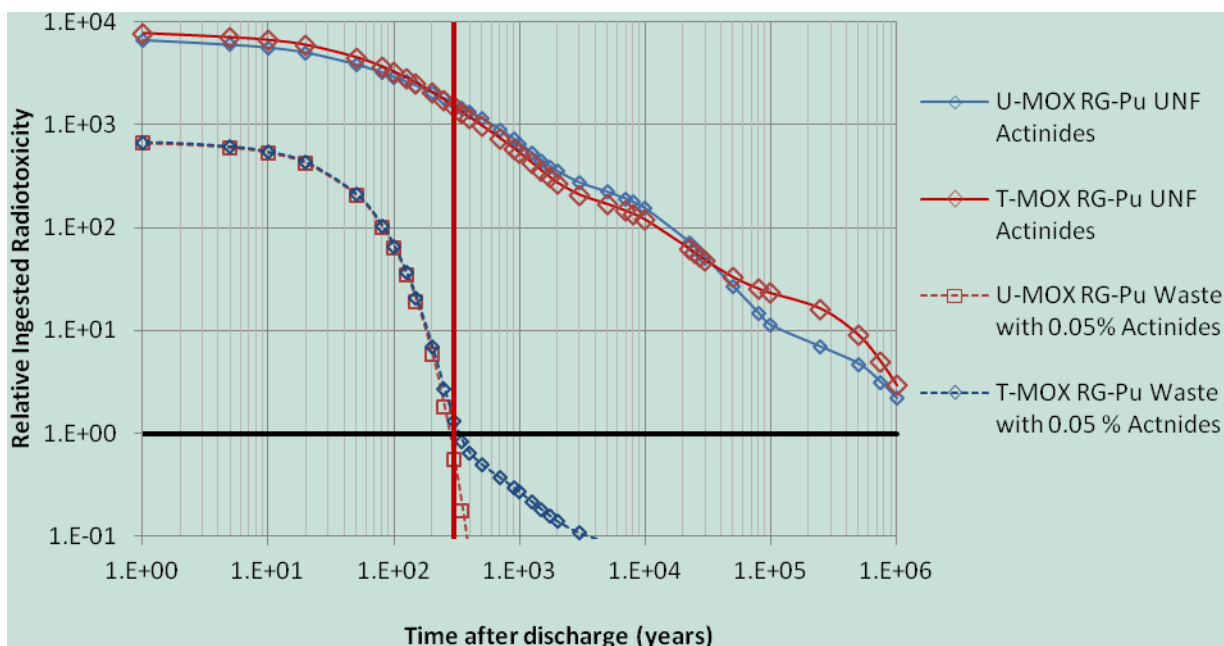


Figure 6: Ingested radiotoxicity for irradiated RG-Pu MOX and RG-Pu T-MOX fuel assemblies and HLW streams from reprocessing.

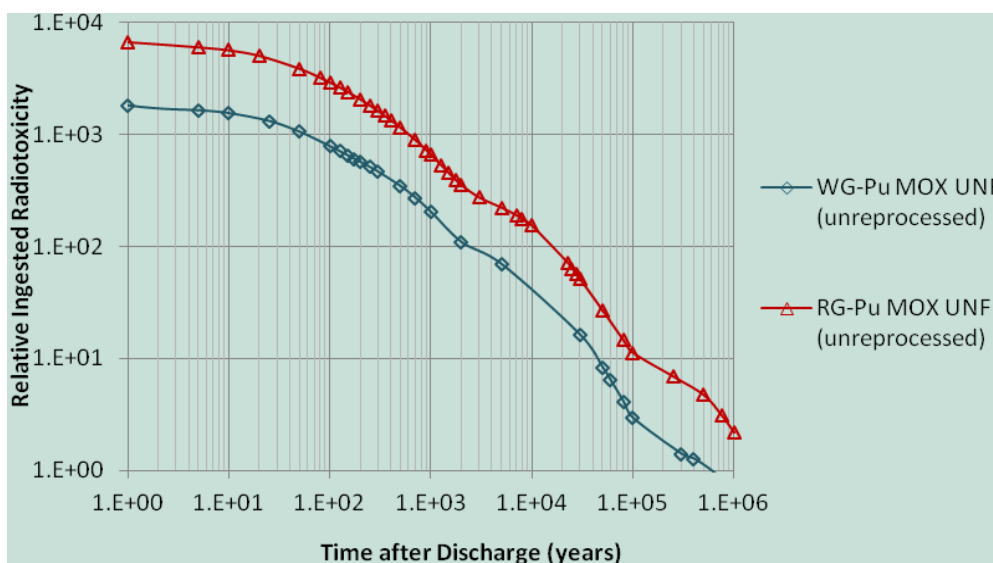


Figure 7: Ingestion radiotoxicity for irradiated WG-Pu and RG-Pu MOX fuel assemblies

From the bottom plots in Figure 6, one can presume that used U-MOX and T-MOX fuels can theoretically achieve the 300-year objective. However, the recovery ratios imposed on the reprocessing are extremely tight, with full recovery of actinides with an efficiency of 99.95%. This might not be technically feasible or economically realistic. Thus recycling Pu (or TRU) in a thermal reactor will make further recovery and recycle (in thermal or,

more likely, fast reactors) extremely expensive and will likely generate unfavorable HLW. This may be a strategic consideration to keep in mind before embarking in a MOX program.

CONCLUSIONS

This paper analyses the requirements for the separation and recycle of UNF to achieve an “acceptable” HLW, i.e. one that reaches radiotoxicity comparable to that of the equivalent open-cycle natural uranium in 300 years. Only HLW generated by reprocessing losses for first, thermal recycle fuel has been investigated. The effect on HLW of multiple recycling of fuel in current and advanced reactors need to be assessed in future studies.

From this preliminary analysis it is shown that thorium-based fuel holds more promises for generating acceptable HLW to the extent that U-233 top-up fissile can be made available. If Pu instead of U-233 is used, either for a shortcoming in the supply of U-233 or to dispose of legacy waste UOX UNF, the HLW radiotoxicity generated is substantially higher, comparable to that of MOX fuel.

The insights gained in this study on radiotoxicity for first thermal recycle fuel with various fissile and fertile materials will guide the development of a multiple tier system, with a mix of current and advanced reactors, serving the mission of generating acceptable HLW.

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