

Methods for Fuel Cycle Analysis in Support of a Reduction of the Radiotoxicity of High Level Waste-11356

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ABSTRACT

Growing concerns regarding the disposal of spent nuclear fuel (SNF) may hamper the nuclear renaissance. The industry must therefore provide politically and socially acceptable solutions for the waste disposal problem. It is believed that reducing the intermediate and long-term radiotoxicity of SNF will alleviate many of these concerns and lead to broader public acceptance of nuclear power. Westinghouse is proposing a partition and transmutation strategy to substantially reduce the radiotoxicity of the high level waste in a 300-year time frame with a synergistic application of advanced reprocessing and reactor technologies. The high level waste is the focus of this study because it has the highest level of toxicity for the longest period of time and is the most mobile of the various forms of waste. A steady state material balance based methodology has been developed to support this objective which enables the evaluation of the impact of alternative design choices on the back-end of the nuclear fuel cycle, such as the reprocessing technology used, reactor design, etc. The composition of the spent fuel and various waste streams generated can be calculated and their radiotoxic content characterized. The paper details the methodology developed.

A general steady state mass balance approach was developed for the back-end of the fuel cycle (see Figure 1), where advanced reactors and reprocessing technologies would be in operation. The mass balance is used to calculate the composition of the waste that will be sent from the reprocessing facility to a permanent repository. Multiple scenarios are possible with different combinations of advanced reactors and reprocessing facilities. A software program has been created to streamline the analysis and facilitate the evaluation of these multiple scenarios. Each scenario can be evaluated based on the composition and radiotoxicity of the waste streams resulting from the mass balance.

INTRODUCTION

A major public concern that potentially hampers the progression of nuclear power as a major contributor to energy in America is the disposal of used nuclear fuel. To overcome this barrier, a permanent solution is needed, and currently a long term repository (i.e. Yucca Mountain) does not appear to be a satisfactory solution as the DOE withdrew their application for the geological repository on March 3, 2010 [1]. In addition the political climate in the United States seems to be shifting towards alternatives to the direct disposal of used nuclear fuel. In response to this shift the Blue Ribbon Commission has been formed under the Obama Administration to evaluate solutions for long term storage of nuclear fuel and waste [2]. With the focus shifted to alternative solutions, a closed fuel cycle seems to be a viable option to reduce the volume of high level waste (HLW) and its radiotoxicity. Although reprocessing was

ruled out long ago in the US, it is currently returning to favor, especially with its international success in countries such as France, India, Japan, and Russia [3].

The goal is to design a system (fuel cycle, reactor design, reprocessing) to bring the radiotoxicity of the HLW that originates from the current inventory of used nuclear fuel to that of an equivalent amount of uranium ore within 300 years, a goal the Westinghouse has proposed in WEC paper 11452 [4]. In order to achieve this goal, the reactor and reprocessing systems must be designed to recycle and burn the major long term contributors to the nuclear waste radiotoxicity, typically the Transuranic (TRU) isotopes of plutonium, and the minor actinides (MAs) of americium, curium and neptunium, and long lived fission products (Long Lived Fission Products).

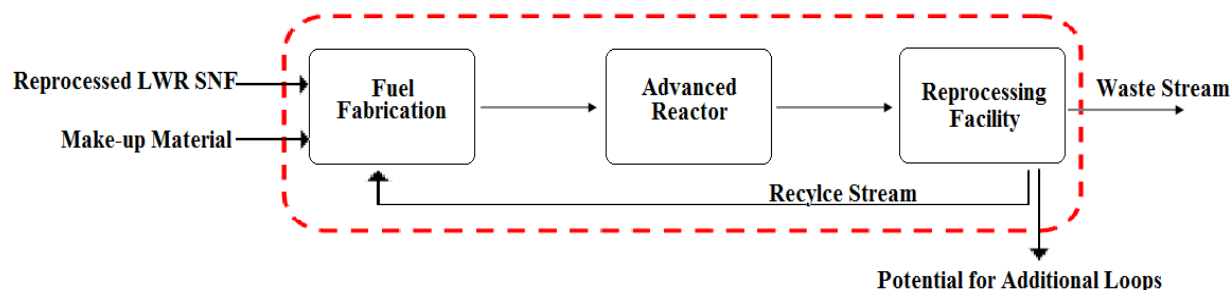
A countless number of combinations of reactor designs and reprocessing technologies are possible and thus an efficient way must be developed to sift through these options in order to focus on meeting the 300 year goal. This paper presents a steady state approach for sorting through the different reprocessing and reactor options to find the scenarios that can achieve the 300 year goal. The approach is an analytical method, taking into account the known capabilities of reprocessing technologies and the impact of various reactor designs on the spent fuel composition.

This project also examines the pros and cons of the Uranium versus Thorium fuel cycle. The use of ^{233}Th is in fact a better choice for the thermal spectrum and can conceivably be used in a breeder reactor [5]. Thorium-based fuels are of particular interest since they reproduce a HLW with a lower radiotoxicity versus conventional U-based fuels [6]. The reduction in radiotoxicity is fostered primarily because ^{232}Th has a different decay chain than ^{238}U and thus it does not as readily transmute into transuranics with significant radiotoxicity impacts [7]. Notably, ^{232}Th can breed fissile ^{233}U in many reactor types and neutron spectra (even though with different efficiencies). Thorium also has the benefit of being widely abundant and results in the formation of very little transuranic isotopes, which are major contributors to radiotoxicity. While closed fuel cycle technology is further advanced internationally, Westinghouse hopes to design a closed fuel cycle that is politically, environmentally and economically favorable by analytically examining different combinations of reactors, reprocessing technologies and fuels.

DETERMINING THE WASTE COMPOSITION

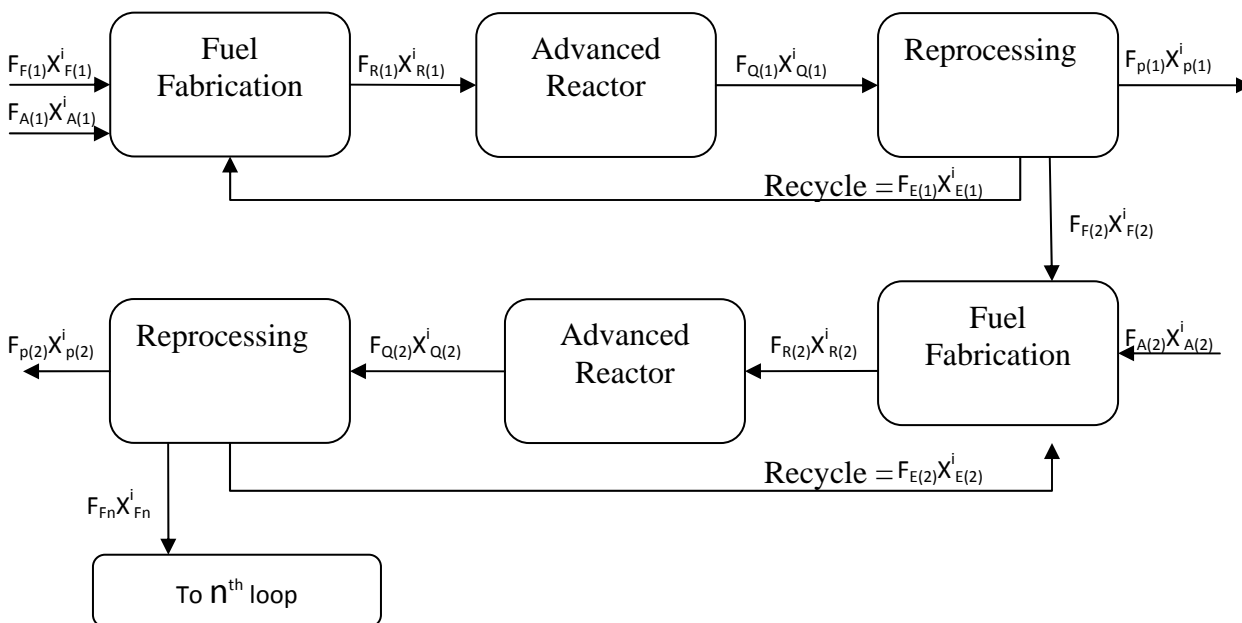
To analytically determine whether or not a proposed system can achieve the 300 year goal, a steady state mass balance is used to find the mass and composition of the waste leaving the system. The "system" being evaluated in the mass balance is the entire back-end of the fuel cycle (i.e. reactor fuel fabrication, advanced reactors, reprocessing facilities). Figure 1 depicts the back-end of the fuel cycle in a flow chart.

Figure 1: Schematic of the Back-End of the Fuel Cycle



The mass balance is performed around the boundaries of the “loop” which is the sequence of fuel fabrication, advanced reactor, and reprocessing facility. It is indicated by the boundaries of the red dotted line in Figure 1. The current mass balance (based on the structure of Figure 1) assumes that the entire system is at steady state. The system has a parallel structure, where the lineup of advanced reactor fuel fabrication to advanced reactor to reprocessing facility is defined as a loop within the system. The system can consist of multiple loops (such as depicted in figure 2); in fact, different reactors and fuels may need to be employed for optimum burning of actinides and Long Lived Fission Products to achieve the 300-year criteria.

Figure 2: Multiple Loop Schematic



Where multiple loops exist, the preceding loop feeds separated isotopes from its reprocessing facility to the proceeding loop’s fuel fabrication where it is incorporated into that loop. The feed material to the system is LWR spent fuel.

The original light water reactor (LWR) spent fuel is reprocessed and partitioned as needed to make a HLW that meets the 300 year goal. Any residuals are sent to the advanced reactor fuel for burning in the advanced reactor loop. A makeup stream of fresh material is provided to

maintain the total reactivity to meet the prescribed cycle energy.

The advanced reactor will be run with the intent of burning MAs (by fission) and transmuting Long Lived Fission Products to reduce the radiotoxicity of the final waste. Advanced reactors of current consideration are listed in Figure 3.

Figure 3: List of Potential Reactors [8]

| Reactor | Spectrum |
|-------------------------------------|------------------|
| Gas Cooled Fast Reactor | Fast |
| Lead Cooled Fast Reactor | Fast |
| Molten Salt Reactors | Fast/Epi-Thermal |
| Sodium Cooled Fast Reactor | Fast |
| Supercritical Water Cooled Reactors | Fast/Epi-Thermal |
| Very High Temperature Reactor | Thermal |

Note that Long Lived Fission Products will only be put into the fast reactor fuel if required to meet the 300 year limit. Many fast reactors appear to be viable systems for burning MAs as they run in the fast spectrum where more MAs are undergo fission than in the thermal spectrum where they tend to be transmuted. Only one stream enters and exits the advanced reactor. The exiting stream is sent to a reprocessing facility where isotopes that do not meet a 300 year criteria are isolated for further irradiation.

The reprocessing facility has only one stream entering but three streams exiting. The first exiting stream is the waste stream, which contains the material that could not be separated via reprocessing or did not need to be separated. The goal for this waste stream is to meet the 300-year criteria. The second stream is the recycle stream. The recycle stream contains the isotopes that were able to be separated via reprocessing and are going to be recycled within the present loop. The final stream is the pass stream. The pass stream would send isotopes that are able to be separated via reprocessing to another loop in the system. In general, an isotope would be passed if it is more amenable to burning in a different system loop's reactor.

To determine whether a chosen system meets the 300-year criteria, one must find the mass and composition of the waste product for the whole fuel cycle. Eventually the mass of all the isotopes in the waste product will be input into ORIGEN ARP 5.1, which calculates the radiotoxicity evolution over time to compare against the 300-year objective. To find the exiting mass and composition (by isotope) a mass balance must be performed around the system.

If a conversion ratio can be defined for every isotope i , a mass balance can be defined around each n th loop and then the waste product of all loops can be summed to get the total waste of the system. For ease of variables in the derivation, a few ratios will be defined. These are the conversion ratio, R , which is the mass flow (by isotope) out of the reactor over the mass flow into the reactor, and Q , the recycle ratio, which is the mass flow in the recycle stream over the

mass flow out of the reactor.

This can be modeled to apply to a series of loops, where the formula applies to the n^{th} loop:

$$F_{pn}X_{pn}^i = \frac{R_n(1-Q_n)(F_{An}X_{An}^i + F_{Fn}X_{Fn}^i)}{(1-Q_nR_n)}$$

$F_{pn}X_{pn}^i$ = Waste stream for isotope i, loop n

R_n = Conversion ratio for isotope i, loop n

Q_n = Recycle ratio for isotope i, loop n

$F_{An}X_{An}^i$ = Make-up feed stream composition for isotope i, loop n

$F_{Fn}X_{Fn}^i$ = Feed stream from previous loop for isotope i

This formula can then be applied to each loop of the system and the $F_{pn}X_{pn}^i$ for each isotope i are summed to find the total waste product from the system. Note the waste product from the original LWR not used in the advanced reactor fuel must also be taken into account. The total mass (by isotope) of the waste will be evaluated in ORIGEN to find radiotoxicity and determine if it meets the 300 year criteria.

CONVERSION RATIOS

The conversion ratio across the reactor is defined to be the ratio of the mass of the isotope exiting the reactor over the mass of the isotope that entered. It is necessary that the reactor be modeled with an appropriate core physics simulation tool to attain accurate masses of the isotopes. Also, it should be considered that these conversion ratios vary over time (i.e. cycle number) until an equilibrium cycle is established for every isotope of the system. In addition, the cooling time of the fuel (e.g. 5 years) needs to be accounted for. ORIGEN can be used to account for decay during this cooling period. The fuel vector coming out of the reactor is therefore passed to ORIGEN which will give the vector before reprocessing. This final vector divided by the initial vector will give the R for each isotope.

Note that in addition to the recycle strategy and cycle number, these R values are dependent upon the reactor and fuel type. Based on flux spectra from the literature, the reactor types that look most promising will be selected. Accordingly, core physics simulations for each selected reactor type and fuel type (e.g., uranium versus thorium, metal vs. oxide, etc) will be performed.

The R values are a good indication of feasibility of different reactors for reducing the presence of actinides and Long Lived Fission Products in the waste. An R value under 1 is indicative of a reactor that will remove an isotope, and thus ideally a reactor would have R values below 1 for

MA and Long Lived Fission Products. It must be noted that if an MA or Long Lived Fission Product had an R value of less than 1, but was transmuted into a longer lived waste product, then this cancels the benefit and must be taken into account. R values also provide interesting insights on fuel type choice (uranium versus thorium-based fuels). In a similar manner to comparing reactor types described above, a fuel is certainly promising if it has a great deal of R values under 1 (especially for minor actinides and other problem isotopes). It is currently expected that thorium will have significantly lower R values for Pu and MAs than uranium, which provides justification for consideration of thorium for fuel in the back-end. It has been shown in previous studies that thorium will create less TRUs than uranium in similar reactor scenarios and that this leads to lower radiotoxicity [9] in the final waste. Tommasi [10] determined that, "americium and curium would be produced in negligible amounts" and that "the waste radiotoxicity associated with Th-U fuels is lesser than that of U-Pu fuels up to some 10^4 years after disposal". It should be noted that Thorium provides no advantage as far as Long Lived Fission Products and may create other "problem" isotopes [7]. An adequate simulation campaign of various reactor models and implementation strategies will enable better quantification of the potential radiotoxicity improvement due to switching from uranium to a thorium-based fuel cycle.

REPROCESSING RATIOS

The reprocessing ratio, M, is the ratio of the amount of an isotope that can be physically separated from the waste product over the amount of isotope before separations. This project is not necessarily attempting to define a new reprocessing technique and so all processes and values were obtained from literature. The M value is important in the derived formula for the waste product as the Q value is equivalent to the M value when an isotope is being recycled. This is due to the fact that when an isotope can be separated from the waste stream by the reprocessing technology an either or situation exists. The separated isotope can either be recycled within the loop or passed on to the subsequent loop. Thus if the isotope is recycled within the loop then the reprocessing ratio (M value) becomes the recycle ratio Q. While if the isotope is passed on to the subsequent loop, then the M value is also used to determine the amount that can physically be passed to the next system. The remaining part of this section on M values entails specific reprocessing technologies that are currently available and their M values found in literature.

Plutonium Uranium Reduction and Extraction, also known as PUREX, is an aqueous process that separates just uranium and plutonium. The process is a liquid-liquid extraction using tributyl phosphate as the extractant. The main issue with PUREX is the proliferation risk as Plutonium and Uranium are in separate pure streams. Although unlikely to be used in the future it has been proven in industrial use in the US, UK, France and Russia [11]. Derivations of PUREX are more promising for the future, especially the UREX+ suite developed at Argonne National Lab. These derivations are much more appealing as they eliminate many proliferation concerns because there is never a pure plutonium stream. They also allow for separations of more isotopes of MAs and Long Lived Fission Products reducing the heat load and radiotoxicity of the waste a great deal more than could be achieved with PUREX alone.

The reprocessing ratio for each can be found in Table 1.

Table 1: PUREX & UREX Reprocessing Ratios (M Values) [12]

| Element | Purex | Urex+1a* | Urex+2 | Urex+3a | Urex+3 |
|-------------|-------|----------|---------|---------|--------|
| U | .999 | .99997 | .999994 | .99999 | .9996 |
| Pu | .999 | .00005 | .998 | .9993 | .996 |
| Am | 0 | .003 | 0 | 1 | .988 |
| Cm | 0 | 0 | 0 | 0 | 0 |
| Np | 0 | 0 | .872 | 1 | .703 |
| Cs | 0 | .9985 | .997 | 1 | - |
| Tc | 0 | .955 | .801 | 0 | .944 |
| Sr | 0 | .999 | .986 | .999 | 0 |
| Lanthanides | 0 | .9997 | 0 | 0 | 0 |
| Rare Earths | 0 | 0 | 0 | 0 | .272 |

*Urex+1a values from 2006 testing

Dry reprocessing also exists as an option for reprocessing of spent nuclear fuel. Atomic International Reduction Oxidation process, also known as AIROX, is a dry oxidation-reduction process [13]. It can be used for reprocessing dry oxide fuels. It removes cladding and some volatile fission products, such as Kr, Xe, I, and ³H, however it does not remove any actinides. Benefits include its simplicity, especially compared to aqueous reprocessing. It is also more proliferation resistant than aqueous reprocessing as it only uses gaseous and solid materials [13] and never produces a pure Pu stream. Table 2 presents the reprocessing ratios (M Values) for the AIROX process. It most likely will also be a cost effective process for conditioning fuels in batch intervals, especially in fast reactors, where there is a huge buildup of Fission Products, that might decrease the clad and fuel performance.

Table 2: AIROX Reprocessing Ratios (M Values) [13]

| Isotope | Recovery Ratio |
|---|----------------|
| ³ H | 1 |
| C ¹⁴ | 1 |
| ⁸³ Kr, ⁸⁴ Kr, ⁸⁵ Kr, ⁸⁶ Kr | 1 |
| ¹²⁹ I, ¹²⁷ I | 1 |
| Xe | 1 |
| ¹³³ Cs, ¹³⁴ Cs, ¹³⁵ Cs, ¹³⁶ Cs, ¹³⁷ Cs | .9 |
| ¹⁰¹ Ru, ¹⁰² Ru, ¹⁰⁴ Ru, ¹⁰⁶ Ru | .9 |
| Cd | .75 |
| Te | .75 |
| In | .75 |

Pyroprocessing is a promising option for reprocessing. It is based on basic electrochemical behavior where the spent fuel is dissolved in a molten salt electrolyte [14]. The Uranium, Plutonium, MAs and some other fission products are dissolved and deposited on cathodes.

Pyroprocessing is very proliferation resistant because the Plutonium cannot theoretically be separated as it has a very similar deposition potential to other TRU elements and uranium. Another benefit of pyroprocessing is its ability to reprocess both oxide (with some difficulty) and metal fuels. Currently oxide fuels are being used in LWRs within the US, but if advanced reactors were to be employed which use metal fuels, the ability of pyroprocessing to convert between the two would be valuable [15]. A major benefit may also exist in the potential for cost reductions. It firstly is a simple process which in turn is cost effective. Secondly it can be located on reactor sites, eliminating the need for transport of SNF. Transport of SNF itself adds cost and proliferation concerns. Unfortunately, consistent and accurate reprocessing values could not be found in literature.

FLUOREX exploits both fluoride volatility as well as solvent extraction [16]. Fluoride volatility utilizes the ability of uranium to combine with fluorine to form UF_6 . The uranium hexafluoride and other volatile fluorides that are created are separated and sent to uranium clean up and recovery to be isolated as pure components. The main elements targeted are Uranium, MAs: Cm, Am, Np, and Long Lived Fission Products: I, Tc, Cs, Sr, ^{85}Kr [17]. By eliminating cesium and strontium, the heat load on the repository is reduced greatly. Another benefit is that fluoride volatility does not have the ability to separate out pure plutonium and is thus better for proliferation concerns. Table 3 has the reprocessing ratios for the fluoride voloxidation process.

Table 3: Fluoride Voloxidation Reprocessing Ratios (M values) [17]

| Isotope | Recovery Ratio |
|-----------|----------------|
| U | .99 |
| Pu | .99 |
| 3H | 1 |
| ^{14}C | 1 |
| ^{85}Kr | 1 |
| ^{129}I | 1 |
| Cs | .98 |
| Tc | 1 |
| Ru | 1 |
| Rh | .8 |
| Te | .9 |
| Mo | .8 |

Supercritical CO_2 extraction is another potentially viable reprocessing option [18]. This extraction utilizes HNO_3 -TBP in supercritical CO_2 as the solvent, which can form complexes with U, Pu, and other important elements, including some Long Lived Fission Products, from SNF. The advantages of supercritical CO_2 extraction are that supercritical CO_2 is readily available and inexpensive. It is nontoxic and environmentally friendly. Finally it reduces

flammability hazards compared to other aqueous processes because it does not involve organic or aqueous solvents [18]. However, it operates at high pressures which may make remote operation difficult. Table 4 presents the reprocessing ratios for Supercritical CO₂ Extraction.

Table 4: Supercritical CO₂ Extraction Reprocessing Ratios (M values) [18]

| Isotope | Recovery Ratio |
|---------|----------------|
| U | .94 |
| Pu | .86 |
| Eu | .94 |
| Am | .86 |

CONCLUSION

If the nuclear industry is to continue to grow, a waste solution will need to be provided that is both economically viable and has public support. Introducing a system of advanced reactors and reprocessing technologies is an attractive option if it has the potential to reduce the radiotoxicity of spent nuclear fuel to that of the original uranium ore within 300 years. However it is costly and exhausting to manually determine the outcomes of the multitude of possible schemes that exist for closing the fuel cycle. For preliminary examination, an analytical method that can be easily formatted in a program such as Microsoft Excel © is efficient and useful for comparison purposes.

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