Waste Management Approach for a Potential Future U.S. Closed Fuel Cycle - 16233

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ABSTRACT

The U.S. Department of Energy, Office of Nuclear Energy has chartered an effort to develop technologies to enable safe and cost effective recycle of commercial used nuclear fuel (UNF) in the U.S. Part of this effort includes the evaluation of exiting waste management technologies for effective treatment of wastes in the context of current U.S. regulations and development of waste forms and processes with significant cost and/or performance benefits over those existing. This study summarizes the results of these ongoing efforts with a focus on the highly radioactive primary waste streams.

The primary streams considered and the recommended waste forms include:

- Tritium separated from either a low volume gas stream or a high volume water stream. The recommended waste form is low-water cement in high integrity containers.
- Iodine-129 separated from off-gas streams in aqueous processing. There are a range of potentially suitable waste forms. As a reference case, a glass composite material (GCM) formed by the encapsulation of the silver Mordenite (AgZ) getter material in a low-temperature glass is assumed. A number of alternatives with distinct advantages are also considered including a fused silica waste form with encapsulated nano-sized AgI crystals.
- Carbon-14 separated from LWR fuel treatment off-gases and immobilized as a $CaCO_3$ in a cement waste form.
- Krypton-85 separated from LWR and SFR fuel treatment off-gases and stored as a compressed gas.
- An aqueous reprocessing high-level waste (HLW) raffinate waste which is immobilized by the vitrification process in one of three forms: a single phase borosilicate glass, a borosilicate based glass ceramic, or a multi-phased titanate ceramic [e.g., synthetic rock (Synroc)].
- An undissolved solids (UDS) fraction from aqueous reprocessing of LWR fuel that is either included in the borosilicate HLW glass or is immobilized in the form of a metal alloy in the case of glass ceramics or titanate ceramics.
- Zirconium-based LWR fuel cladding hulls and stainless steel (SS) fuel assembly hardware that are washed and super-compacted for disposal or as an alternative Zr purification and reuse (or disposal as low-level waste, LLW) by reactive gas separations.
- Electrochemical process salt HLW which is immobilized in a glass bonded Sodalite waste form known as the ceramic waste form (CWF).

- Electrochemical process UDS and SS cladding hulls which are melted into an iron based alloy waste form.

Mass and volume estimates for each of the recommended waste forms based on the source terms from a representative flowsheet are reported.

INTRODUCTION

Background and Approach

The U.S. Department of Energy, Office of Nuclear Energy is evaluating options for disposition of commercial used nuclear fuel (UNF). Research is being performed to develop transformational technologies that may be employed as part of advanced, sustainable, nuclear fuel cycles. The Fuel Cycle Technologies (FCT) program is aimed at developing a broad set of options that will enable future decision-makers to make informed decisions about how best to manage UNF.[1]

This work supports the FCT though the detailed evaluation of wastes produced from the advanced nuclear fuel cycles and to prioritize future waste management technology development. The approach taken was to:

1) select a representative, high-performance, fuel cycle and associated chemical flowsheets
2) develop a consistent mass balance for materials within the flowsheet based on split factors and chemical additions for each unit operation (derived from literature and ongoing experimental work within the FCT)
3) select waste forms and processes for each major waste stream generated
4) estimate the composition and loading for waste forms and the scale of the waste process equipment
5) report the results for use in more detailed fuel cycle options evaluation

Generally, the most mature (e.g., demonstrated) waste form/process that meet the current U.S. requirements were selected. In many cases additional waste forms or process that were less technically mature, but, show potential for significant advantages over the most mature technology were also evaluated.

Fuel Cycle

To this end a systematic study was performed to evaluate and screen potential nuclear fuel cycles for future application within the U.S.[2] This study identified four groups of fuel cycles as being most promising for R&D, based on the evaluation criteria used:

- Continuous recycle of U/Pu with new natural-U fuel in fast critical reactors
- Continuous recycle of U/TRU with new natural-U fuel in fast critical reactors
- Continuous recycle of U/Pu with new natural-U fuel in both fast and thermal critical reactors

- Continuous recycle of U/TRU with new natural-U fuel in both fast and thermal critical reactors

All four of these fuel cycles include the continuous recycle of actinides in fast or fast plus thermal reactors. Waste technology described in this paper is therefore focused on wastes generated from continuous recycle of actinides (U, Pu, Np, Am, and Cm).

Flowsheet Assumptions

To evaluate the wastes generated from a continuous recycle nuclear fuel cycle assumptions need to be made on the reactors, fuels, and separations flowsheets being deployed. One reasonable example set of assumptions was selected for this purpose. A homogeneous recycle of the uranium (U) and transuranic (TRU) elements resulting from the reprocessing of light-water reactor (LWR) uranium oxide (UOX) fuel is used as feed for advanced metal fuel fabrication. The metal fuel is irradiated in a sodium cooled fast reactor (SFR) and the used fast reactor (FR) fuel is reprocessed electrochemically. The recovered U/TRU from electrochemical separations is recycled to metallic FR fuel. Waste streams from the aqueous and electrochemical (echem) reprocessing are treated and prepared for disposition. Off-gas from the separations and waste processing are also treated. Figure 1 gives the upper level flowsheet box flow diagram.

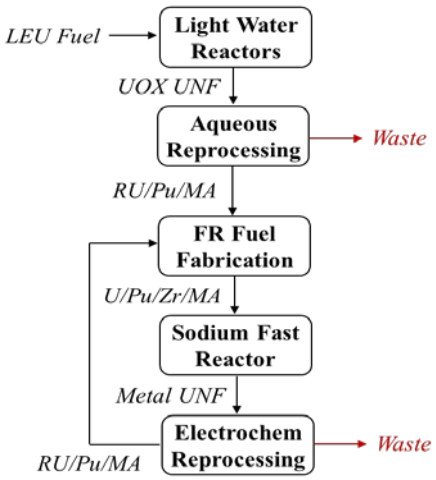

Figure 1. Overall Block Flow Diagram.

Waste compositions and masses are estimated using the following assumptions:

1. Nominal annual throughput of 1000 metric tons of initial uranium (tU)/y LWR fuel is processed with aqueous reprocessing and 20 metric tons of initial heavy metal (tHM)/y FR fuel is processed electrochemically.
2. Used LWR fuel is based on 5% enriched UOX fuel in Zircaloy-4 cladding irradiated for 50 gigawatt days (GWd/tU) in a pressurized water reactor (PWR) and cooled for 5 years before reprocessing (although longer times were used to estimate the impacts of heat on waste).

3. Used FR fuel is based on a U, 20 mass% Pu, 10 mass% Zr, and minor actinides (MA, primarily Np, Am, and Cm) sodium bonded metallic fuel in HT-9 cladding irradiated for 100 GWd/tHM in a SFR operating with a conversion ratio of 0.75 and cooled for 2 years before reprocessing.
4. Separation and decontamination efficiencies of:
    a. Minimum of 99% recovery of U, Pu, and MA, individually, from the used fuel.
    b. Minimum of 99% Separation of actinides from lanthanides (LN).
    c. Maximum of 1% contamination of MA product stream with LN.

The assumed process flowsheets for aqueous and electrochemical reprocessing are given in Figure 2.

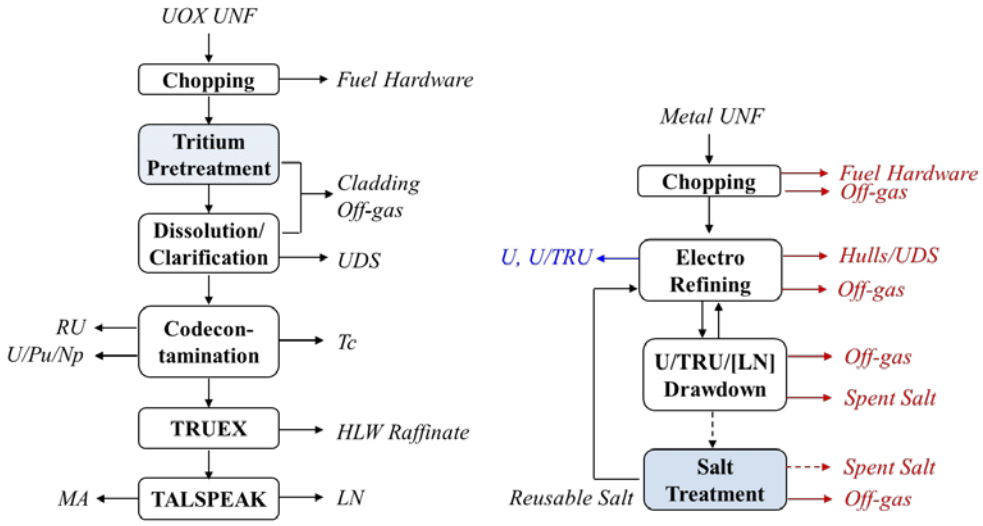

Figure 2. General Aqueous (a) and Electrochemical (b) Separations Process Flowsheets

These processes are described in more detail along with the waste compositions and flow rate in Vienna et al. 2015.[3]

RESULTS

The waste streams considered in this study include:

- Tritium separated from either a low volume gas stream or a high volume water stream,
- Iodine-129 separated from off-gas streams in aqueous processing,
- Carbon-14 separated from LWR fuel treatment off-gases,
- Krypton-85 separated from LWR and SFR fuel treatment off-gases,
- An aqueous reprocessing high-level waste (HLW) raffinate waste,

- An undissolved solids (UDS) fraction from aqueous reprocessing of LWR fuel,
- Zirconium-based LWR fuel cladding hulls
- Stainless steel (SS) fuel assembly hardware,
- Electrochemical process salt HLW, and
- Electrochemical process UDS and SS cladding hulls.

Also evaluated, but not described in this paper are the broad range of secondary wastes from reprocessing facilities [4, 5] and wastes from decommissioning and decontamination of nuclear fuel cycle facilities including reactors, fuel fabrication, and fuel reprocessing plants.[2]

The characteristics and selected forms and processes for each of these streams are briefly described below. More detailed analyses can be found in Vienna et al. 2015.[3]

Tritium

Four options were considered for H-3 management. The first is a thermal pretreatment step to separate most of the tritium from the LWR fuel prior to dissolution. The tritiated water would be captured from the low-volume off-gas stream and immobilized. The second is the management of high-volume water streams exiting the plant that would be directly immobilized. The third option would be to optimize the use of water in the plant to reduce the volume of tritiated water and immobilize only the highly concentrated tritiated water. The fourth option is to isotopically enrich H-3 in the high-volume water stream (the example used in this study was combined electrolysis catalytic exchange [CECE]). Immobilization in low water cement (water: cement ration of 0.3) was found to be the most mature technology for treating the tritiated water stream. Although the 12 year half-life of H-3 suggests that a high durability waste form is not needed, evidence for exchange between ground water and the H-3 in the cement will likely require disposal in a high-integrity container (HIC) to meet U.S. regulatory requirements. The resulting masses and volumes of waste produced are summarized in Table 1.

Radioiodine

U.S. regulations require a high degree of capture for I-129 from the reprocessing facility. The most mature material for adequately separating the iodine from the various gas streams (dissolver, vessel, cell, and melter) is silver functionalized mordenite (AgZ). The AgZ would be immobilized by sintering with a low-temperature glass frit to form a glass composite material (GCM).[6, 7, 8] A relatively new iodine getter was developed with roughly 4× the iodine capacity, higher resistance to aging in plant operating environments, and a simpler waste process – silver functionalized silica aerogel (AgAero).[9] If AgAero is employed,

then the final waste form is a fused silica glass encapsulating nano-particulate AgI.[10, 11] Estimating the mass and volume of waste forms produced for either option requires the accounting for loss of capacity of the getter for aging and the competition of other halogens for available iodine binding sites. The resulting masses and volumes of waste produced are summarized in Table 1.

Carbon

There is a potential that C-14 must be captured to meet U.S. regulations for aqueous reprocessing radionuclide releases.[12] In that case, C-14 (in the form of CO_2) would be captured by caustic scrubbing of the head-end off-gas stream. The carbonate would be precipitated in the form of $CaCO_3$ and cemented. The resulting masses and volumes of waste produced are summarized in Table 1.

Krypton

Krypton-85 must be captured when processing fuel younger than 30 years (from discharge) in the U.S.[12] The most technically mature technology for Kr capture is cryogenic distillation.[13] However, that technology is relatively expensive to implement and poses challenges in overcoming safety requirements. Several other technologies have been developed and demonstrated at some scale. The most promising for this application is the use of solid sorbents that can operate anywhere from +25°C (for metal organic frameworks) to -80°C (for zeolites).[13, 14, 15, 16, 17] Once captured, the Kr-85 can be stored in pressurized gas cylinders, deposited in metal waste form by sputtering , or sintered into a zeolite by hot-isostatic pressing (HIP). The resulting masses and volumes of waste produced are summarized in Table 1.

High Level Waste

The high-level waste (HLW) raffinate either from codecontamination process or from the combined transuranic extraction raffinate and the MA-lanthanide separations process product can be treated alone or combined with the undissolved solids (UDS) from dissolver solution clarification and the soluble fraction of the Tc. This leads to several potential combinations of waste/streams and forms. The most mature technology and approach is to combine the HLW, the UDS, and the Tc into a single HLW stream and vitrify this stream in alkali-borosilicate glass.[18] The resulting waste loading depends strongly on the amount of decay heat in the HLW. Therefore, estimates of glass masses and volumes were estimated for fuel that is cooled five years and for fuel cooled fifty years before reprocessing (Table 1). Some advanced waste forms show promise for immobilizing the HLW, particularly when the UDS+Tc stream is treated separately. This is due to currently uncertain,

but, restrictive concentration limits of noble metals in glass melters (assumed to be 3 wt% $PdO+Rh_2O_3+RuO_2$ for the purposes of this study [19]). In the case of separate treatment of UDS+Tc stream, metal alloy waste forms are assumed.[20, 21, 22, 23]

Glass ceramics waste form show significant promise for the HLW stream. They can tolerate significantly higher heat than borosilicate glass (with T_g near 950°C), crystals effectively immobilize those components that are sparsely soluble in borosilicate glass (e.g., MoO_3 and lanthanides), and the waste form can be fabricated by the same technology used for borosilicate glass.[24, 25] The second potential waste form is Synroc [26, 27] that can be fabricated by either a sol-gel, calcination, HIP process or by a melt-cast process.[27, 28, 29, 30, 31, 32] Synroc is not sensitive to decay heat and so the mass and volumes are estimated only once, while the masses and volumes for glass ceramics are estimated for both 5 year and 50 year cooled fuel in Table 1.

Hulls and Hardware

The last waste stream considered for aqueous reprocessing is the zirconium based hulls and stainless steel (SS) hardware. The most mature technology that are expected to meet programmatic needs is the super compaction process where the metals are sorted and loaded into drums that are pressed at room temperature to form a roughly 66 % dense metal puck as deployed at the AREVA LaHague Plant. In this case both zirconium and SS are combined into a single stream. An alternative approach is the purification of Zr using chlorination process that would enable either Zr reuse (for nuclear applications) or low-level waste disposal if more cost effective.[33, 34] In the case of Zr purification, the SS hardware would be sorted and that hardware requiring deep geologic disposal would be compacted. The resulting masses and volumes of waste produced are summarized in Table 1.

Electrochemical Salt HLW

The electrochemical process assumed in this study is performed in molten LiCl-KCl eutectic salt. That salt is used until it is no longer-efficient due to build-up of impurities from the fuel and bond materials. Currently the most mature technology for treating this salt waste is the ceramic waste form (CWF) developed to immobilize the salt from EBR-II fuel treatment wastes.[35, 36, 37] A higher waste loaded version of this same CWF has long been postulated [38] and recently been fabricated and tested. Finally, a tellurite-based waste glass was proposed to immobilize salt HLW.[39, 40] These waste forms are summarized in Table 2.

Other researchers have proposed a number of potential waste treatment options in which the chlorine is recycled and only the chlorine free waste components are immobilized.[41, 42, 43, 44] These approaches were not evaluated in this study.

Electrochemical Hulls and Noble Metals

The last waste stream specifically considered is the SS hulls and undissolved solids (those FR fuel elements more noble than U) from electrochemical processing. The most mature waste form for this stream is the melted metal waste form that has been deployed to treat EBR-II treatment wastes.[45, 46, 47] The resulting waste form mass and volume is given in Table 2. No other waste forms or processes were considered in this study.

SUMMARY AND CONCLUSIONS

A detailed study was undertaken to evaluate the wastes to be produced from an example closed nuclear fuel cycle in the U.S.[3] A mass balance was estimated for the example processes considered to implement this fuel cycle and used to evaluate waste management options. Reference and alternative waste forms and associated processes were selected for each major waste stream. The masses and volumes of waste forms are summarized in Tables 1 and 2. These results are useful for planning the impacts of closed fuel cycle implementation in the U.S. and to help focus additional research in the area.

Table 1. Estimated Reprocessing Waste Masses and Volumes for a 1000 tU/y Aqueous Used Fuel Reprocessing Facility (50 GW/tU burnup LWR fuel).

Stream	Waste form	Mass, kg/tU	Vol, L/tU	Comment
Tritium				
Pretreatment off-gas	low water cement	0.07	0.035	water:cement ratio of 0.3
High volume water	low water cement	16 700	8770	water:cement ratio of 0.3
Optimized water	low water cement	1000	526	water:cement ratio of 0.3
Isotope enriched water	low water cement	16.7	8.77	water:cement ratio of 0.3
Iodine				
AgZ	glass composite material	104	25.5	total halogen:I ratio of 3
AgAero	fused silica-AgI	4.65	1.55	total halogen:I ratio of 3
Krypton	Low pressure gas	0.646	3.7×10^{-3}	50 atm
	High pressure gas	0.646	1.1×10^{-3}	163 atm
	Zeolite	0.646	2.8×10^{-3}	HIP
	Metal matrix	0.646	7.8×10^{-4}	Cu matrix

Carbon	Cement	81	43	water:cement ratio of 0.3
High level waste				
HLW+UDS+Tc	Borosilicate glass	401	154	5 y cooled fuel
HLW+UDS+Tc	Borosilicate glass	302	116	50 y cooled fuel
HLW Only	Glass ceramics	214	70	5 y cooled fuel
HLW Only	Glass ceramics	76	25	50 y cooled fuel
HLW Only	Titanate ceramics	173	38	
UDS+Tc	e-metal	9.71	0.81	
UDS+Tc	Fe-metal	19.4	2.43	
Hulls and Hardware				
Hulls + Hardware	Supercompacted	301	67.6	66% volume
Hardware only	Supercompacted	47	8.9	66% volume
Zr recycle	Recycled			wastes TBD

Table 2. Estimated Reprocessing Waste Masses and Volumes for a 20 tU/y Electrochemical Used Fuel Reprocessing Facility (100 GW/tHM burnup SFR fuel).

Stream	**Waste form**	**Mass, kg/tHM**	**Vol, L/tHM**	**Comment**
HLW Salt	CWF Nominal	2,700	1,280	25 mass% BSG, 0 mass% Halite
	High-loaded CWF	1,740	980	20 mass% BSG, 2.5 mass% Halite
	TeO_2-based glass	3,920	810	Density 4,830 kg/m^3
Ducts, Plenums, etc…	Supercompacted	3,310	630	66% volume
Hulls and noble metals	Melted Fe-metal	530	67	
Krypton	Low pressure gas	0.688	3.94×10^{-3}	50 atm
	High pressure gas	0.688	1.17×10^{-3}	163 atm
	Zeolite	0.688	2.98×10^{-3}	HIP
	Metal matrix	0.688	8.31×10^{-4}	Cu matrix

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