

# **Integrated Waste Management Strategy and Radioactive Waste Forms for the 21<sup>st</sup> Century**

## **Alternative Materials for Radioactive Waste Stabilization and Nuclear Materials Containment**

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March 2007

The INL is a  
U.S. Department of Energy  
National Laboratory  
operated by  
Battelle Energy Alliance



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## Introduction

The U. S. Department of Energy (DOE) Global Nuclear Energy Partnership (GNEP) was announced in 2006. As currently envisioned, GNEP will be the basis for growth of nuclear energy worldwide, using a closed proliferation-resistant fuel cycle. The Integrated Waste Management Strategy (IWMS) is designed to ensure that all wastes generated by fuel fabrication and recycling will have a routine disposition path making the most of feedback to fuel and recycling operations to eliminate or minimize byproducts and wastes. If waste must be generated, processes will be designed with waste treatment in mind to reduce use of reagents that complicate stabilization and minimize volume.

The IWMS will address three distinct levels of technology investigation and systems analyses and will provide a cogent path from (1) research and development (R&D) and engineering scale demonstration, (Level I); to (2) full scale domestic deployment (Level II); and finally to (3) establishing an integrated global nuclear energy infrastructure (Level III). The near-term focus of GNEP is on achieving a basis for large-scale commercial deployment (Level II), including the R&D and engineering scale activities in Level I that are necessary to support such an accomplishment. Throughout these levels is the need for innovative thinking to simplify, including regulations, separations and waste forms to minimize the burden of safe disposition of wastes on the fuel cycle.

## Background

In the U. S., policy for disposal of Spent Nuclear Fuel (SNF) and High Level Waste (HLW) is derived from the Nuclear Waste Policy Act (NWPA) as amended.<sup>1</sup>

Currently, SNF is not reprocessed in the U. S., and most of the HLW inventory comes from past processing of defense related materials. Commercial fuels come from Nuclear Regulatory Commission (NRC) licensees, and, under current policy, are to be disposed by the DOE with defense fuels and stabilized HLW in a geologic repository. The total inventory to be disposed is legislatively limited to 70,000 metric tons of heavy metal (MTHM) until a second repository is available. This mass limit actually refers to the initial uranium charged to reactors from which SNF and HLW are derived. Defense related materials are limited to 10% of that inventory. No repository has yet been licensed, but the Yucca Mountain Facility (YMF) has been the most studied to date, and licensing activities are underway. Under the DOE are three offices that work together to manage the fuel cycle. The Office of Nuclear Energy (NE) supports US nuclear energy programs, including GNEP. The Office of Environmental Management (EM) is responsible for mitigating the risks and hazards posed by the legacy of nuclear weapons production and research, including defense HLW. Finally, the Office of Civilian Radioactive Waste Management (OCRWM or RW) manages and disposes high-level radioactive waste and spent nuclear fuel including designing and modeling, obtaining the license, and operating a geologic repository.

Throughout the past several years, the Advanced Fuel Cycle Initiative (AFCI), which was the predecessor program to GNEP, sponsored extensive R&D efforts related to aqueous-based and pyrochemical separations processes for recycling thermal reactor [i.e. light water reactor (LWR)]

and fast reactor (FR) SNF. The proposed fuel cycle consumes TRU elements and supports growth of carbon-free international nuclear energy markets. Building on the nuclear science and engineering knowledge gained over the last 60 years, the proposed recycling system is not only more sustainable than prior concepts, it will also generate less long-lived waste and reduce the impacts of heat and long-lived radiation on a geologic repository.

The research conducted through AFCI was primarily focused on developing an understanding of the chemistry and performance of the various steps that constitute these processes. This has also resulted in an understanding of the basic characteristics of the waste streams that are expected from the separations activities. However, specific chemical composition and quantities of the waste streams will depend on the separations efficiencies and operational performance of full-scale separations processes, which have yet to be demonstrated and characterized. Nevertheless, an conceptual disposition paths for each of these waste streams, including a waste form, waste processing technology, and storage/disposal scenario, has been proposed. Some key data gaps have been identified relative to the waste forms and waste processing technologies as well as several regulatory challenges.<sup>ii</sup> These gaps will continue to be identified as the concepts evolve, and must be resolved prior to full-scale implementation.

### Strategy

The strategy is primarily to follow a simple philosophy of integrating the inherent responsibilities for waste management into the rest of the nuclear fuel cycle. When options are available in designing fuel fabrication and separations processes, including scrap recovery and reagent use, the impacts on waste management will be considered, and feedback will be provided from a byproducts and waste disposition perspective. In addition to the GNEP programmatic goals, the IWMS has two distinct mandates:

- 1) No wastes will be generated without ensuring a pathway for safe disposition in the form of recycle, reuse, or safe disposal.
- 2) No long-term storage of unstabilized or liquid wastes will be allowed. Waste storage will only be for the express purpose of process throughput or as a means of treatment to allow it to decay over a prescribed time period to render it safe for disposal.

Currently in the US and internationally the capability exists to process and stabilize all of the waste streams resulting from the aqueous reprocessing and pyroprocessing flowsheets. This general knowledge has provided the underpinnings for the current disposition concept that has been established for GNEP. However, it does not necessarily represent an optimized, or even an efficient basis, even for a single-facility infrastructure. For example, many waste form options exist for fission product (FP) waste streams (borosilicate glass (BSG), iron-phosphate glasses, glass-bonded ceramics, ceramics, etc.), yet BSG has been identified as the HLW form. This may or may not offer the best option when waste loading, durability, and cost are all considered. This potential inefficiency is further exacerbated in the context of a large, integrated domestic nuclear infrastructure complex, and further yet when evaluated from a global perspective. A key challenge is to demonstrate a commercially-viable fuel cycle. This will necessarily drive the GNEP program to demonstrate an optimized waste management strategy that considers the scale and dynamics of complex systems, including fuel fabrication, reprocessing, storage, disposal, and the associated ancillary infrastructure (e.g. transportation) and material flow through the system. Feedback amongst fuel fabrication and recycling and waste and byproduct management is essential to optimize the fuel cycle.

## Current Conceptual Waste Disposition

For all of the waste streams expected to result from aqueous separations and pyroprocessing, an initial waste form, treatment technology, and disposal/storage path have been identified. In most cases, the waste form chemistry and performance and the process technology efficiency have been demonstrated and validated on an engineering-scale. For several of these waste streams, the default disposal pathway is as HLW in a geologic repository. However, currently there is no storage or disposal infrastructure or regulatory framework in place to allow for efficient final disposition that meets the GNEP goals of long-term extension of the geologic repository. Significant work is needed to investigate alternative, more efficient waste forms and waste processing technologies and opportunities for system integration and optimization through targeted systems analyses and trade studies. The waste streams and disposition paths envisioned for GNEP are shown for aqueous reprocessing in Table 1 and for pyroprocessing in Table 2. Note that some of these pathways are not currently available and may require regulatory changes. A more detailed summary of current radioactive waste forms and disposal issues can be found in 2007 Draft Global Nuclear Energy Partnership — Materials Disposition and Waste Form Status Report, February, 2007.<sup>ii</sup>

Many opportunities exist to potentially improve on these concepts, and systems analyses on waste treatment and wastefoms are currently underway. Some examples include:

### *Cesium/Strontium*

The current concept for treatment of the aqueous Cs/Sr product relies on conversion to a powder via a fluidized bed steam reformer. Data to date shows the product is primarily very fine (~10 micron). This material was to be made into a monolith using clay to form a hydroceramic, but this may be reconsidered due to concerns on gas generation due to radiolysis. Reliable fluidized-bed operation and maintenance with concentrated Cs is also a significant safety concern, and options for conversion to a monolithic solid while destroying coincident organic should be evaluated. In the latest pyroprocessing flowsheets the Cs/Sr product from oxide fuels is a glass bonded sodalite containing Cs/Sr and small amounts of halite; from metal fuels the matrix is similar, but the Cs/Sr are not segregated and the La/FP are included. In addition to waste loading, selection of the final waste form will also consider heat transfer, gas generation, container corrosion and resistance to degradation due to radioactive decay (i.e. changes in valence, atomic size, and chemistry) and should pass Toxic Characteristic Leach Procedure (TCLP) for Ba leachability.

The Cs/Sr stream is significant in size, and will likely receive intense scrutiny due to the unprecedented strategy for decay storage. Systems studies are also planned for the separations-disposition strategy to evaluate the benefits of separating out Cs/Sr. It is imperative that the studies are creative in evaluating concepts to achieve a reliable waste form that will last 300-500 years and be acceptable for cross-generational management. Hybridization of aqueous and pyroprocessing to separate waste salts, pressing a matrix to form a compacted product up to and including Hot Isostatic Pressing (HIP) to minimize volume and volatility, and perhaps including a transition metal as an electron donor/receptor should all be considered. A novel concept such as encapsulating the dried granular solid Cs/Sr oxide (e.g. rotary calciner product) in a low melting point (<500C) alloy (Zn, Sn, Cu, Al) could maximize heat transfer, provide for radioactive decay and mitigate corrosion and Cs volatility. This study should also include review of the significant body of work already done on converting Cs/Sr to forms to be used for heat and radiation sources.

Table 1. Waste Streams and GNEP Conceptual Disposition for Aqueous Reprocessing

Aqueous Process Streams	Stream Description/Derivation	Envisioned Disposition
Assembly hardware (SS)	Spacers, endcaps, etc. removed prior to chopping	Direct repository disposal as compacted or melted activated metal Evaluate performance assessment for SLB of GTCC
Gaseous Products Kr/Xe and <sup>3</sup> H	Voloxidation releases Kr/Xe and <sup>3</sup> H which are caught on absorber beds	Decay storage of Kr/Xe and <sup>3</sup> H followed by SLB of all absorbers or packaged forms as LLW
Iodine, Carbon-14	Sorption of I on silver zeolite, and <sup>14</sup> C as carbonate	SLB of grouted absorber if Class A/B/C LLW, geologic repository of GTCC
Hulls / Cladding (Zr)	Residuals following fuel dissolution washed to LLW levels using HF and HNO <sub>3</sub> in dissolver	Direct disposal as LLW-SLB, disposal as LLW-GTCC if necessary due to activation
Undissolved Solids	Sludge from dissolver bottom and clarifier solids, containing noble metals and TRU	Melt with portion of metal wastes for repository disposal
Separated LEU	Oxidation of uranyl nitrate solution from UREX to U <sub>3</sub> O <sub>8</sub>	Store as national resource material or SLB of oxide as LLW
Tc on IX resin	Acid side IX of UREX raffinate can be stripped of pyrolyzed to Tc metal	Melt in Zr/SS alloy for repository disposal using portion of cladding and SS hardware
Cs / Sr stream	CCD/PEG Solvent extraction of UREX raffinate, yields Cs, Sr, barium (Ba) and rubidium (Rb)	Stabilize for long-term (300 yr) decay storage and eventual disposal as Class C LLW
TRU stream	Oxidize TRU either Pu/Np and Am/Cm separately or together	Product for FR fuel fabrication
Lanthanides and FP stream	TRUEX raffinate and Talspeak product	Vitrify as glass for repository disposal
Liquid waste (aqueous and organics)	Liquids from several locations in the process including off-gas treatment streams, spent solvents, solvent wash solutions, laboratory returns, and other miscellaneous liquids	Stabilized solids, SLB of stabilized salts as LLW
Miscellaneous Solid debris	Spent equipment, PPE, laboratory and operation solid waste (pipettes, wipes, etc.), after decontamination.	Direct SLB as LLW

Table 2. Waste Streams and Conceptual Disposition for Pyroprocessing

Pyroprocess Streams	Stream Description/Derivation	Envisioned Disposition
Volatile Products (Kr/Xe, <sup>3</sup> H)	Released during chopping process and electrorefining and caught on absorbers	Decay storage of Kr/Xe, <sup>3</sup> H followed by SLB of all absorbers or packaged forms as LLW
Residual metals and UDS	Undissolved metal waste stream from dissolution includes SS hulls, Tc, Zr, and noble metals	Melt as metal waste form for repository disposal
Separated LEU	Deposited on iron cathode as U metal followed by heating to remove adherent salts	Store as national resource material or SLB of oxide as LLW
Cs/Sr	Capture on zeolite from salt bath, contains Cs, Sr, Ba, and Rb	Make glass bonded zeolite for long-term (300 yr) decay storage and eventual disposal as Class C LLW
TRU stream	TRU electrolytically partitioned with some LEU	Product for FR fuel fabrication
Lanthanide, FP, iodine and carbon-14 stream	Zeolite membrane separated FP containing salts	Convert to glass bonded zeolite for repository disposal
Miscellaneous Solid debris	Spent equipment, electrorefining crucibles, PPE, laboratory and operation solid waste	Direct SLB as LLW

*Technetium*

The Tc waste form from both aqueous and pyroprocessing is to be a metal alloy. The difference in the aqueous and pyroprocessing flowsheets is that aqueous processing uses capture on ion-exchange resin followed by pyrolysis, where pyroprocessing captures the Tc along with SS fuel hulls, and other noble metals in a much larger stream. It is now believed that decontamination of the activated and contaminated Zircaloy LWR hulls is unlikely to achieve Class C LLW limits, so there will probably be a large metal waste stream (including SS hardware) from aqueous as well. This evaluation should include potential for and value of higher waste loading. (I.e. is higher Tc waste loading needed if all of this metal is GTCC waste anyway, vs. does the Tc/TRU content preclude an alternative surface disposal for the activated metal?) Also, Tc is readily oxidized, and mobile as anionic pertechnetate, thus the alloy should contain a more active metal (Zr) to protect the Tc from oxidation, but how much is necessary has not been quantified. Ramifications of incorporating the undissolved solids (UDS) from aqueous processing should be evaluated as well. It is not yet known if the noble metals in the UDS will cause any difficulties with the Tc/SS/Zr alloy. Redox control during melting should also be evaluated to minimize production of dross. One concept is skipping the resin pyrolysis step, and adding the loaded resin directly into the hull/hardware scrap during melting to act as a reductant. Inclusion of small amounts of carbon should not degrade the waste form, and this could result in a more simple process.

*Undissolved Solids/Hulls/Metal Hardware*

In addition to evaluating the UDS impacts to the Tc waste form described above, this evaluation will consider the merits of volume reduction by compaction of hulls and hardware vs. melting. Compaction could yield approximately 60-70% volume reduction whereas melting could yield essentially theoretical density, but melting requires more energy (generally not a significant cost) and could volatilize contaminants (could be quite costly). This study should seek out data to

bracket expected composition of UDS from LWR fuel dissolved in nitric acid w/wo using hydrofluoric acid.

#### *Lanthanides/Balance of Fission Products*

Of all the streams separated in the UREX+1a aqueous fuel processing, the residual lanthanides and fission products remaining after completion of the other key separations are most likely to be considered high-level waste. The U.S. precedent for HLW treatment is conversion to borosilicate glass (BSG) in a joule-heated melter (JHM). Pyroprocessing product results in a glass bonded sodalite containing small amounts of halite. This waste is relatively innocuous after the GNEP separations and could be a candidate for disposal as GTCC. An analysis will be done comparing the expected waste form to national and international standards for low and intermediate level wastes and wasteforms besides BSG. The analysis will consider keeping the La and FP streams separate as well as combining. BSG made in a JHM was chosen as the initial concept because it is the DOE baseline for defense HLW, but this analysis will also consider current technologies used worldwide (cold-crucible induction melters) and other waste forms such as iron-phosphates or HIP products that could provide higher waste loading.

#### *Off Gas: I, <sup>3</sup>H, Xe/Kr, <sup>14</sup>C*

Iodine and carbon-14 must be sequestered essentially indefinitely, but <sup>3</sup>H, Xe/Kr can be managed in decay storage. Capture methods will be evaluated including parameters such as absorber selectivity, efficiency, regeneration effectiveness, and conversion to final waste forms. Initially selected capture technologies include silver-zeolite for iodine, molecular sieve for tritium, caustic scrub for carbon-14, and zeolite (mordenite, faujasite) for Xe/Kr. Whether these isotopes are stripped and stored as compressed gases, stabilized in grout (<sup>3</sup>H, <sup>14</sup>C), or stabilized in place (grouted or collapse of zeolite structure) has yet to be defined. This evaluation will consider the large body of historical data and present the reasoning for why a particular method or methods are chosen. The study will also provide some feedback on the capture efficiency to be expected based on testing to date and what data is needed.

#### *Future Analyses*

In parallel to validating the concepts for waste treatment technologies and wasteforms, Level II strategy analyses and limited testing will also be initiated to provide supporting data. For example, all streams from processing SNF could be potentially classified as HLW under current regulations. In the U. S., this is a functional rather than characteristic designation in that all wastes derived from fuel processing are designated HLW, regardless of their radioactivity, chemistry or the risk they pose to human health or the environment. This makes the geologic repository the default disposal pathway for all waste streams. However, to accomplish some of the GNEP goals, particularly extending the life of the YMF to at least the rest of this century, some radionuclides must be managed separately, such as recycle of TRU elements as fast reactor fuel. Thus, several key regulatory and policy changes must be made to maximize the benefits of advances in technology. The IWMS, in its role as the primary interface point for DOE-NE, RW, and EM, will help to identify the technical and regulatory/policy strategic opportunities for implementing such changes.

Other processes such as separations and fuel fabrication will be analyzed to evaluate how their evolution affects waste management. The Level I and II analyses will provide feedback to improve integrated operation and overall plant efficiency. For example, ferrous sulfamate is currently the preferred reductant to achieve high separations efficiencies of Pu in the aqueous reprocessing flowsheet. However, this adds ~10% iron into the process stream that eventually feeds into the residual mixed fission product stream. The treatment concept for this waste stream is vitrification into a BSG form. The higher iron content could significantly reduce the waste



loading in BSG. Consideration of alternative glass compositions, such as an iron phosphate glass, may resolve this waste-loading issue, but the waste form would then require qualification for disposal in YMF. An investigation of the comparative benefits of a less efficient or more costly reductant versus qualification of an alternative waste form should be conducted to determine the optimal solution. This is an example of the type of potential benefit that can be realized through an integrated waste management system development process.

The practicality of current US radioactive waste regulations which include a mixture of functional and characteristic designations must also be evaluated. For example, high level waste is designated functionally, as the wastes derived from fuel reprocessing. However, with the additional separations envisioned under GNEP, this definition may become obsolete, because what was once lumped as HLW will now be fractionated into specific streams for beneficial reuse, decay storage, and disposal. The residuals that are to be direct disposed as HLW may be better regulated simply as greater than class C or LLW-GTCC, for which regulations already exist. Similarly the definition of TRU wastes, those DOE wastes containing at least 100 nCi/g TRU elements, may be obsolete in the GNEP commercial environment because the definition is strictly limited to defense related materials. The designation "TRU waste" has no legal meaning for commercial wastes, and again, the actual waste definition defaults to GTCC. Perhaps, for GNEP, both HLW and TRU designations can be eliminated, and all wastes can simply be classified characteristically based on the health and environmental risks they pose due to their composition, and simply be regulated as Class A/B/C and GTCC.

Other evaluations will include:

- Evaluate benefits of Cs/Sr separations versus leaving in or combining with the lanthanide/mixed FP stream. While the potential benefits of keeping the short-term heat pulse from Cs/Sr out of the repository is well documented (REFERENCE WIGELAND), it is not obvious that the regulatory structure or siting of a dedicated facility for "decay storage" will be straightforward. It may be advantageous to potentially simplify separations by eliminating the Cs/Sr recovery, thereby leaving the Cs/Sr in the HLW form, and storing the HLW in a dedicated area in or near the repository for long enough time to reduce the heat load satisfactorily.
- The Cs/Sr stream content of the fuel has been proposed for segregation to remove the short-term heat pulse to the repository. For that same reason, this material should be considered as a significant source of energy that could be used for beneficial purposes. Cesium produces a relatively high-energy gamma radiation that must be shielded, but if designed correctly, the decay heat produced could be used to provide passive cooling for waste, as a source of heat to produce steam, or other beneficial purposes.
- Consider hybrid operations that combine the best attributes of aqueous and pyroprocessing to eliminate or combine some waste streams. Candidates include 1) treating the fission product contaminated chloride salt wastes using an aqueous separation and 2) combining the technetium from aqueous with the pyroprocessing metal waste form.
- Evaluate Zircaloy and stainless steel wastes for possible decontamination or surface disposal as GTCC under a performance assessment considering the integrity of the metal itself as a durable waste form similar to decommissioned pressure vessels already disposed at Hanford. National implementation of fuel recycling with expanded use of nuclear energy will create a significant market in which contaminated stainless steel and

Zircaloy could potentially be reused. This market could be large enough to warrant dedicated contaminated metal processing. This concept will also be evaluated.

- A significant expansion of nuclear energy using both aqueous and pyroprocessing will generate some wastes that cannot be readily or efficiently vitrified. Thus expansion of the technical bases for a HLW repository license to include additional HLW forms other than BSG based on mechanistic understanding of waste form degradation and how radionuclides are released should also be evaluated. Argonne National Laboratory and Idaho National Laboratory have started this process for the ceramic and metal waste forms from pyroprocessing, but the basis for the modeling will probably have to be evaluated to determine if available performance data can be used to adequately characterize these waste forms.
- The current Waste Isolation Pilot Plant (WIPP) repository capacity and license are restricted to *defense* wastes. Commercial wastes exceeding 100 nCi/g would be greater than Class C (GTCC), and would require disposal in a geologic repository. Disposal of this waste in the YMF could possibly preclude meeting the GNEP goal of reducing the amount of TRU destined for the YMF by 99%. This disposal path should be reevaluated, including consideration of the 100 nCi/g limit, and disposition of wastes contaminated to greater than background or naturally occurring radioactive material (NORM) levels (10nCi/g) but less than 100 nCi/g. Most TRU is to be recycled as fuel, but significant TRU will probably be uneconomic to recover from equipment, operating wastes (i.e. rags, bags, PPE, etc). There must be a disposition pathway for commercial wastes, and the issues of long term heat generation and toxicity must be addressed for the geologic disposal facility.
- In the U. S. radioactive iodine in concentrations greater than Class C must be disposed in a geologic repository or some equivalent manner that mitigates dose over the very long half life of <sup>129</sup>I. One option to consider is managing this material in a similar manner to TRU wastes, but not just because of the long half-life. The WIPP repository for TRU wastes in the U.S. is built into a salt deposit, the residual of an ancient sea. This sea salt contains significant nonradioactive iodine that would provide isotopic dilution to any radioactive iodine eventually leached from the waste form. This type of synergy amongst waste chemistry, waste form and waste disposition should be considered in new strategies for future wastes.
- Designation of a routine disposal pathway for GTCC LLW not requiring a case-by-case performance assessment requires significant regulatory analysis. Thermal and radio-toxicity issues must be considered.
- Voloxidation is a developmental concept at this time, but failing efficient separation of volatile FPs prior to chemical or electrolytic dissolution of fuel, I, <sup>3</sup>H, and <sup>14</sup>C will contaminate many streams internal to the processes, complicating waste management later. High efficiency separation of these FPs at the head-end will simplify capture and waste disposition. Some of these processes have not been in active development for over 20 years. AFCF will be an excellent test bed to proof test and verify performance of advanced offgas treatment trains for use in Level II.
- Consideration of the concept of “decay storage”: secure storage facilities to allow problematic radionuclides such as Cs, Sr, tritium, and noble gases to decay to LLW

limits. These materials must be stored for several hundred years isolated from the biosphere, and protected against unregulated use.

#### Conclusions

A closed fuel-cycle has long been sought to support implementation of nuclear energy for peaceful uses in a sustainable, environmentally responsible manner. The GNEP concept offers one solution for a proliferation resistant fuel cycle, and also offers many opportunities to reconsider how radioactive wastes are managed. New strategies may require advances in waste form materials and how they are characterized. The concepts described here are examples of studies to be done.

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#### References

<sup>1</sup>. The Act consists of the Act of Jan. 7, 1983 (Public Law 97-425; 96 Stat. 2201), as amended by P.L. 100-203, Title V, Subtitle A (December 22, 1987), P.L. 100-507 (October 18, 1988), and P.L. 102-486 (The Energy Policy Act of 1992, October 24, 1992). The Act is generally codified at 42 U.S.C. 10101 and following.

<sup>ii</sup> Gombert II, D. “2007 Draft Global Nuclear Energy Partnership — Materials Disposition and Waste Form Status Report”, GNEP-WAST-AI-TR-2007-00013.