

INEEL/CON-04-02481 PREPRINT

## **High Level Waste Disposal System Optimization**

D. Gombert
J. Roach
M. Connolly
W. Holtzscheiter

February 27 - March 3, 2005

**Waste Management '05** 

This is a preprint of a paper intended for publication in a iournal or proceedings. Since changes may be made before publication, this preprint should not be cited or reproduced without permission of the author. This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, or any of their employees, makes any warranty, expressed or implied, or assumes any legal liability or responsibility for any third party's use, or the results of such use, of any information, apparatus, product or process disclosed in this report, or represents that its use by such third party would not infringe privately owned rights. The views expressed in this paper are not necessarily those of the U.S. Government or the sponsoring agency.

High Level Waste Disposal System Optimization

D. Gombert, J. Roach, M. Connolly, Bechtel BWXT Idaho, LLC Idaho National Engineering and Environmental Laboratory

W. Holtzscheiter, Westinghouse Savannah River Company, Savannah River National Laboratory

#### **ABSTRACT**

The DOE high level waste (HLW) disposal system is based on decisions made in the 1970s. The de facto Yucca Mountain WAC for HLW, contained in the Waste Acceptance System Requirements Document (WASRD), and the DOE-EM Waste Acceptance Product Specification for Vitrified High Level Waste Forms (WAPS) tentatively describes waste forms to be interred in the repository. The final WAC must be written to conform to the licensing specifications issued by the Nuclear Regulatory Commission (NRC) in the actual license. We now know that the current system is far from optimal for disposal of the diverse HLW streams, and proven alternatives are available to reduce costs by billions of dollars. Before applying for the license to receive and possess (the operating license), the governing basis for HLW disposal should be reassessed to consider extensive waste form and process technology research and development (R&D) efforts, which have been conducted by DOE-EM, international agencies (i.e. ANSTO, CEA), and the private sector. Current plants must go forward, but retrofit studies are needed now. This will lead to a more optimized HLW disposal system that will provide for accelerated HLW disposition, more efficient utilization of the YMF, and overall system cost reduction. A comprehensive and collaborative program between DOE-EM and DOE-RW is required to investigate alternative approaches to optimizing the HLW disposal system

#### INTRODUCTION

The high level waste (HLW) disposal system consists of the Yucca Mountain Facility (YMF) and waste product (e.g. glass) generation facilities, either currently located or planned, at the Savannah River, Hanford, and Idaho sites. This system is based on decisions made in the 1970s, and the DOE has tentatively described waste forms to be interred in the *Waste Acceptance System Requirements Document* (WASRD), as well as the DOE-EM *Waste Acceptance Product Specification for Vitrified High Level Waste Forms* (WAPS). We now know that the current system is far from optimal for disposal of the diverse HLW streams, and proven alternatives are available to reduce costs by billions of dollars. These changes are also necessary to meet schedule commitments the DOE has made to host states. Current plants must go forward, but retrofit studies are needed now. Preliminary studies show \$2B can be saved in Idaho alone, using current technology.

Responsibility for management of the HLW disposal system is shared between the U. S. Department of Energy (DOE) Office of Civilian Radioactive Waste Management (DOE-RW) and DOE Office of Environmental Management (DOE-EM). The DOE-RW license application must include technical bases that document the acceptability of waste forms against regulatory performance standards. To facilitate progress on immobilization of HLW, the DOE has described the planned borosilicate glass waste form and package requirements in the WAPS and the WASRD. These documents actually serve two purposes: 1) they describe the envelope for acceptable HLW products as part of the technical bases in the license application, and 2) they provide tentative waste acceptance criteria (WAC) for products from existing and planned vitrification facilities. The WAC is only tentative, because the final WAC must be written to conform to the licensing specifications issued by the Nuclear Regulatory Commission (NRC) in the actual license. This governing basis for HLW disposal should be reassessed to consider extensive waste form and process technology research and development (R&D) efforts, which have been conducted by DOE-EM, international agencies (i.e. ANSTO, CEA), and the private sector; as well as support development of performance based waste acceptance criteria (PBWAC) for YMF. This will lead to a more optimized HLW disposal system that will provide for accelerated HLW disposition, more efficient utilization of the YMF, and overall system cost reduction. The proposed program requires close collaboration among DOE-EM, DOE-RW, and a team that includes DOE national laboratories, international agencies, and the private sector, undertaking an integrated effort to meet the following goals:

- 1. Reevaluating and revising the assumptions in the DOE-EM and DOE-RW documents that control the waste form/package/disposal program.
- Qualifying additional protective, but more efficient, waste matrices by matching the matrix to the waste instead of forcing all wastes into borosilicate glass (BSG). This does <u>not</u> include developing new waste forms, only qualifying existing matrices to demonstrate protection equal to or better than the baseline (Environmental Assessment [EA]) glass.
- 3. Demonstrating existing technology that can produce the alternative waste matrices better than the current baseline (i.e. ceramic-lined, electrode-type, joule-heated melters [JHMs]). This does <u>not</u> include developing new technology concepts. The emphasis is on demonstrating existing technology that is clearly better (reliability, productivity, cost) than current technology, and to justify its use in future facilities or for retrofit at time of planned change-out.
- 4. Preparing the technical bases to support inclusion of additional acceptable waste forms for HLW disposal in the final application for the License to Receive and Possess.

### **BACKGROUND**

The YMF will be a long-term repository for spent nuclear fuel (SNF) and stabilized HLW from fuel reprocessing. The combined performance of the waste matrix, the engineered

waste package(s) and facility design, and the effects of the hydrogeologic interaction at the site govern the repository performance as a system for sequestering radionuclides from the biosphere. This system has been the focus of technical analysis for nearly three decades, with emphasis on system performance in protecting the public from radiological hazards. However, the system has not been optimized for HLW disposal. While DOE HLW/SNF inventory represents about 10% of the repository space, and only about 5% of the activity is in the HLW, there is potential for significant cost-savings in optimizing the HLW processing strategy. HLW processing is costing billions of dollars, thus the potential savings from even small-percentage cost savings are significant. A key limitation on a comprehensive engineering analysis to optimize HLW disposal is the DOE self-imposed requirement in the WAPS that all HLW be converted to BSG. The DOE has also standardized on using JHMs, but currently available technology should be more durable and covers a broader operating envelope. The DOE complex technical community knows this to be an inefficient approach for many wastes, but institutional barriers limit action to change the system.

While the U.S. Environmental Protection Agency has identified vitrification as the "Best Demonstrated Available Technology" for HLW, it does not designate BSG as the only acceptable formulation; hence no regulatory driver exists to restrict the acceptable waste form for HLW to BSG. Borosilicate glass was chosen because it is a very stable material, capable of hosting a wide variety of elements in its amorphous matrix. Both DOE-RW and DOE-EM have adopted BSG as the only qualified waste form for HLW disposal, though other forms are used for plutonium and SNF. Although other waste forms are not specifically precluded, BSG is specifically defined in the key documents that govern HLW disposal at YMF, including the *Waste Acceptance Product Specification for Vitrified High Level Waste Forms (WAPS)* (DOE/EM-0093 Rev. 2), and the *Waste Acceptance System Requirements Document (WASRD)* (DOE/RW-0351P Rev. 4).

# Specific examples include:

#### From the WAPS:

- GLOSSARY FOR THE WASTE ACCEPTANCE PRODUCT SPECIFICATIONS High-Level Radioactive Waste (HLW) the highly radioactive material resulting from the reprocessing of spent nuclear fuel in defense or commercial facilities, including liquid waste produced directly in the reprocessing operation. For purposes of this document, <u>HLW is vitrified borosilicate glass that has been cast in a stainless steel canister.</u>
- Acceptance Criterion The consistency of the waste form shall be demonstrated using the Product Consistency Test (PCT) [3]. For acceptance, the mean concentrations of lithium, sodium and boron in the leachate, <u>after normalizing for the concentrations in the glass</u>, shall each be less than those of the benchmark glass described in the Environmental Assessment for selection of the DWPF waste form [4].

#### From the WASRD:

- Durability and Phase Stability of Vitrified HLW A. <u>The standard vitrified HLW form shall be borosilicate glass</u> sealed inside an austenitic stainless steel canister(s) with a concentric neck and lifting flange.
- Product Consistency
  - 1. The Producer shall demonstrate control of waste form production by comparing production samples or process control information, separately or in combination to the Environmental Assessment benchmark glass using the Product Consistency Test or equivalent.
  - 2. For acceptance, the mean concentrations of lithium, sodium, and boron in the leachate, <u>after normalization for the concentrations in the glass</u>, shall be less than those of the benchmark glass.

The WAPS and WASRD restrict the envelope of acceptable waste forms for HLW even further by specifying an outdated version of the Product Consistency Test (PCT), as delineated in ASTM C-1285-94 which cannot be obtained through ASTM since it has been superseded by ASTM C-1285-02. PCT measures individual constituents dissolved from the glass into the solution, e.g. boron, sodium, and lithium. The current version of PCT has been expanded to include glass and glass ceramics and provides the protocol determining the appropriate constituents to be measured. A similar protocol should be established for waste forms that do not contain a glassy phase. Though it is commonly stated that HLW forms need not be glass, DOE-RW and DOE-EM regulations and guidance form the de facto WAC understood by the sites immobilizing HLW, and these documents are written around BSG glass. By specifying an outdated version of the PCT standard, the documents do not support testing alternative matrices.

The WAPS and the WASRD form a tentative concept for the WAC as expected by the DOE based on the decision to standardize on BSG. The actual WAC will be established based on the licensing specification appended by the NRC to the final license to receive and possess nuclear materials at the repository. The licensing specification will be based on the technical bases provided in the license application. The technical bases that document the acceptability of a waste form against the regulatory performance standards can still be expanded over the next few years to include additional waste forms.

Similarly, the DOE HLW programs revolve around JHMs, the standard technology producing much of the world's glass. The designs for processing HLW are much more expensive, using highly corrosion resistant refractories and high-nickel alloy electrodes, but the technology is essentially the same. This technology was designed to process consistent glass chemistry, in large quantities, within the temperature range typical to common glasses used by industry. Many HLW applications push the envelope for the technology to the extreme.

• The chemistry of HLW is inconsistent.

- The temperatures required to incorporate refractory elements into BSG (i.e. Al, Cr, and Zr) in significant quantities exceed the normal operating range of the materials available for these melters.
- Many HLW compositions are highly corrosive and contain significant volatile species.
- Maintenance in a highly radioactive environment is very expensive.
- Melter lifetime is very limited; even state-of-the-art facilities are ill equipped to dismantle and dispose of the large, highly contaminated spent units.

The benefits and limitations of BSGs and JHMs are well known, well researched and widely documented. There are, however, alternatives for both, including iron-phosphate glasses, glass-ceramics and hydroceramics (waste forms); and cold-crucible melters and hot-isostatic presses (processing technologies). The knowledge gained from over 30 years of R&D within the DOE complex, international agencies and the private sector is largely being disregarded, even though independent evaluations funded by DOE-EM have recommended these alternatives be considered.

Focus on a specific waste form and a particular technology limits the overall efficiency of the HLW disposal system. Decisions made in the 1970s are driving additional costly requirements that offer no benefits in terms of enhanced environmental or worker protection. Other factors that further complicate this situation is that the treatment systems must not only address the wide variety of HLW streams at Hanford and Savannah River, as well as other waste streams generated during accelerated closure activities, but must also address sodium bearing waste and calcine waste at Idaho, which are dramatically different in chemical composition and physical form. It can be expected to ultimately include wastes that will be generated in meeting the nation's future energy needs. We know today that waste forms other than BSG are acceptable choices to immobilize key radioactive and hazardous components in existing and future waste streams (e.g. through a technical down selection process, *plutonium* was to be immobilized in a *ceramic* waste form). As large volume waste streams are addressed (e.g. Idaho National Engineering and Environmental Laboratory (INEEL) calcine), it is imperative to match the waste form to the waste stream characteristics. As an example, INEEL calcines contain components that are difficult to incorporate in a BSG waste form. It has been demonstrated that waste loadings exceeding 50% can be achieved using a cold crucible induction melter to generate a glass-ceramic waste form. If calcine is mandated to be immobilized in a BSG, the resulting waste form is neither economical nor an optimally performing waste form. Requiring a single host matrix for a variety of wastes, some of which are nearly insoluble in the BSG matrix (e.g. phosphorous and sulfur) results in lower waste loading, that leads to greater waste volume, and, in turn, higher processing and operations costs at both the treatment and disposal facilities. These overall HLW disposal system inefficiencies cost more money and take more time. Higher waste processing and disposal efficiency can be realized by performing the engineering analyses and trade-studies necessary to select the most efficient methods for processing the full spectrum of wastes across the DOE complex.

Evaluation of additional waste processing technologies and waste forms without development of new waste form qualification procedures, without expanding the technical bases for waste forms acceptable in the repository, and without changing the DOE governing documents will not result in an optimized HLW disposal system. A comprehensive and collaborative program between DOE-EM and DOE-RW is required to investigate alternative approaches to optimizing the HLW disposal system

#### PROPOSED PROGRAM

This program will result in deployment of more efficient waste processing technologies and additional waste forms to be described in more comprehensive technical bases for use in the future permit application for the repository. To realize the maximum benefit in schedule and cost reduction both activities must be conducted in parallel. This will require close coordination between DOE-RW and DOE-EM as well as strong collaboration among the participating National Laboratories, international agencies and private sector entities, together with the DOE operating facilities that comprise the HLW disposal system. Three primary thrust areas constitute this program; together they offer near-term, mid-term, and long-term benefits. The initial scope is to collaboratively define the three concepts in specific tasks, assigning scope to where the work can be accomplished most effectively.

### **Technical Bases for Additional Waste Forms**

The ultimate goal, and long-term benefit, of this program is to support and achieve more comprehensive technical bases for the YMF license, allowing acceptance of additional waste forms, thus providing DOE cost-effective alternatives for processing the various HLW inventories throughout the DOE complex. This is predicated on developing acceptable understanding and methodology for advancing technical bases that allow a variety of material forms to be qualified for HLW disposal in the YMF. If the program is properly integrated with DOE-RW efforts, and the system optimization research, testing, and validation are collaboratively conducted between DOE-EM and DOE-RW, the parallel effort should support, not detract from, the licensing activities.

The current Total System Performance Assessment (TSPA) for the repository indicates that the HLW matrix and the corrosion rate of the SNF will not significantly impact overall protection of the biosphere. Thus, specific waste form performance requirements should be evaluated by conducting a sensitivity analysis on the TSPA to determine which of the waste form parameters are controlling. Analyses will use the currently established modeling codes to estimate the overall impacts of waste form and waste package durability on the repository performance. The results will focus the efforts to develop the technical bases for additional waste matrices. In the interim, through collaborative efforts, the governing DOE-EM WAPS and DOE-RW WASRD can be modified in a coordinated approach to ensure consistency, while not impacting the ongoing license application process. These interim administrative steps, which are critical to the phased benefits of this program, must be carefully orchestrated to ensure success.

## Matching an Optimal Matrix to the Waste Chemistry

The potential benefits of matching waste chemistry to the host matrix and using currently available technologies have been documented in several studies <sup>2,3,4,5,6,7</sup> conducted by DOE over the past several years. Matrix chemistry may include multiple phase BSG, non-BSG (i.e. aluminosilicate glass, iron phosphate glass), glass-ceramics, and ceramics, as long as standards are met for durability and stability. If a collaborative effort between DOE-EM and DOE-RW were initiated based on performance requirements, near term benefits could be realized by DOE operations such as the Defense Waste Processing Facility (DWPF) at the Savannah River Site, as well as the Waste Treatment Plant, currently under construction at Hanford.

Waste-matrix performance validation will require durability testing, as well as an understanding of the interaction of chemical species with the waste package and the geology that would result during long term waste form degradation. These data will be necessary to feed into the TSPA model that DOE-RW uses to predict behavior and transport of contaminants in the YMF subsurface. Fortunately, the basis for most of this testing exists. It is primarily *what* is measured that must change in order to evaluate other materials (e.g. boron is measured to determine the durability of a BSG, whereas the appropriate analysis for an iron-phosphate glass may be iron). This flexibility to measure the appropriate constituents to evaluate alternative waste matrices must be built into the waste/package/disposal system performance requirements documents.

While the bulk of the waste-matrix testing and validation would be conducted by DOE-EM, this effort must be coordinated with and fully supported by DOE-RW. This is necessary for two reasons. First, DOE-RW must modify the WASRD to allow consideration of alternative materials. Second, DOE-RW must be intimately involved in understanding the matrix durability and long-term performance characteristics such that the TSPA models can validate that the facility requirements are met. This combined effort will support development of the technical bases needed for additional waste forms.

# Alternative Waste Processing Technologies Demonstration

Efforts to optimize the DOE HLW disposal system must include investigation and implementation, as appropriate, of alternative process technologies to produce the waste forms more effectively, which will provide both mid-term and long-term benefits to the DOE. This is because some of the additional waste forms that have been shown to be the most beneficial for specific HLW inventories cannot be produced effectively, if at all, in the current baseline technology, which is the JHM.

While the JHM is a proven technology, it also has significant limitations due to temperature and corrosion constraints, particularly in regard to processing the widely varied and challenging chemistry of the DOE HLW inventory. Acceptance of additional glass compositions might provide some near term benefits to the existing baseline JHMs (for example, iron phosphate glasses for high alumina and high zirconia waste streams could potentially double the effective waste loading, and provide for much lower operating temperatures than BSG); however, implementation would most likely require significant facility and system modifications. Alternative melter technologies have been

preliminarily shown to offer potentially greater improvements in cost-effectiveness and system optimization than can be achieved through continued use of JHMs.

For example, the operational life of a JHM is relatively short (i.e. 5 to 7 years) and this can be further reduced when processing waste that is particularly aggressive, or operating at higher temperatures to enhance waste loading. Replacement of a JHM is costly, and significantly impacts the DOE accelerated clean-up schedule. Conversely, the cold crucible induction melter (CCIM) technology is smaller, modular, and has the potential to provide much longer operational life. It has also been shown to provide a greater operating envelope with the ability to effectively process a broader range of waste forms. For an operating facility such as DWPF, retrofit of the JHM with CCIM technology, could potentially allow for processing of challenging waste streams with higher waste loadings, while not affecting the operational life of the melter. Iron phosphate glass, which is uniquely suited for certain DOE HLW inventories, is ideal for processing in the CCIM technology. Also, this technology is particularly advantageous for operation at elevated temperatures (i.e. 1350°C versus 1150°C), thus providing benefits over the baseline JHM for higher waste loading in BSG, while offering longer operational life, and simpler decommissioning. The CCIM has been cited in numerous reports (referenced above), as providing some of the greatest potential benefits to the DOE in immobilization of HLW.

Additionally, implementation of alternative technologies in planned or existing facilities, while providing potential mid-term benefits in overall waste volume reduction and schedule acceleration, will also provide potential long-term benefits through elimination or reduction of decontamination and disposal costs for spent melters. Similarly, alternative waste forms such as high-density ceramics, glass-ceramics, and mineralized ceramics have been investigated and demonstrated to offer significant potential benefits, both in waste loading and overall operations costs, for many DOE applications. However, as with iron phosphate glasses, these waste forms cannot be effectively produced using the baseline JHM technology. Hot isostatic pressing (HIPing) and fluidized bed mineralization appear to be viable technologies that can produce these cost-effective alternative waste forms, but they have not been investigated to determine their true feasibility for implementation within the DOE. In the long-term, validation of these alternative technologies, and development of quantitative operational data, will be crucial to implementation of advanced fuel cycles in support of the next generation nuclear power plants for the nation.

As with validation of alternative waste matrices, efforts to investigate alternative process technologies must be focused, based on the programmatic strategy, on only those approaches that provide clear and significant benefit, while offering realistic opportunities for implementation into DOE facilities and systems. Significant expertise and capability has been developed within the DOE, international agencies (i.e. CEA and ANSTO), as well as private industry (e.g. AREVA) in waste processing technologies. Collaborations with industry and international agencies need to be strengthened and leveraged to realize the maximum benefit of the proposed approach. Feasibility studies

for retrofit opportunities of existing facilities, as well as implementation in future facilities, require the support of private industry.

# Matching Schedule to the YMF License Process

Additional technical bases to support additional HLW matrices can still be added to the application for the license to receive and possess nuclear materials at the repository without impacting the licensing schedule. The current application for construction authorization need not be impacted at all. Proposed program activities must be initiated in fiscal year 2005 to support this schedule. This approach permits the DOE-RW YMF opening strategy and this program to proceed in parallel while allowing development of the PBWAC approach without affecting the YMF 2010 opening date.

### CONCLUSION

This proposed HLW disposal system optimization program, while aggressive in scope and overall impact, can be successful through close coordination between DOE-EM and DOE-RW. It offers an exceptional opportunity for the extensive expertise within DOE, international agencies, and the private sector to effectively implement a program with a focused end point that meets the DOE-EM accelerated cleanup initiatives, while yielding cost savings that could potentially exceed \$2B. Effective and comprehensive integration of all programmatic activities, at all appropriate levels within DOE-EM and DOE-RW, is critical to program success. In addition, several of the alternative technologies that will be investigated appear to offer potential benefits to the DOE Office of Nuclear Energy efforts to develop the next generation nuclear power plant with an advanced fuel cycle. Advantages of these technologies include greater processing flexibility; lower capital costs, smaller footprint, and reduced decommissioning efforts. As the Generation IV Nuclear Reactor and Advanced Fuel Cycle Initiatives mature, opportunities will exist to embody these technologies in future advanced fuel cycle system designs.

#### REFERENCES

- 1. R. A. Day, et al, Glass Ceramics in a Cold Crucible Melter: The Optimum Combination for Greater Waste Processing Efficiency", WM'03, February 23-27, 2003, Tucson, AZ
- 2. J. M. Perez, et al, "Basis for Requesting a Change to the HLW Repository Requirements for the Production of Immobilized High Level Waste for the River Protection Project", October 2002
- 3. R. L. Treat, et al, "Technical Evaluation of Hanford HLW Vitrification Process Alternatives: Report of the Independent Project Evaluation Team", July 2003
- 4. J. Ahearne, et al, "High Level Waste Melter Review Report", TFA-0108, July 2001
- 5. J. M. Perez, et al, "High Level Waste Melter Study Report", PNNL-13502, July 2001
- 6. D. Carl, "Optimization Study No. 6: Alternative Melter Technologies for Future Replacement and Facility Upgrades", 24590-WTP-RPT-G-01-003, Rev 0, April 2002

7. C. A. Kouts Letter to P. M. Bubar, "Cost Impact on Geologic Disposal from Reducing the Number of Hanford HLW Canisters", July 10, 2003**FOOTNOTES**