U.S. Department of Energy
Office of Environmental Management

Report to Congress Regarding Technology Development and Deployment Activities

July 2008

Introduction

In the report accompanying the House Energy and Water Development (HEWD) Appropriations Bill, 2008 (110-185), the Committee requests the Department to provide a report to Congress on the Environmental Management technology development plan in the following areas:

1) evaluation of alternative supplemental treatment of low-activity waste (LAW) at Hanford, using a systems-analysis approach;
2) increase waste loading in glass; and
3) improving methods to remove non-radioactive components from the sludge-heels in the high-level waste tanks.

This report provides the Department's response to that request. The response is divided into three major sections. The first section describes evaluations that are underway to refine technology development plans associated with alternative supplemental LAW treatment; the following two sections describe, in turn, the latter technical areas listed in the Committee's request, including the technology needs and the previous technology development work that has been performed.

In the broader context, technology development plans in these areas are consistent with the Department of Energy (DOE) Office of Environmental Management (EM) strategic initiative areas identified in the Environmental Management Engineering and Technology Roadmap issued to Congress in March 2008. The Roadmap was developed based on substantial input and support from the National Research Council of the National Academies and in close consultation with senior DOE field management. The National Academies issued an interim report on February 14, 2008, that endorsed the Roadmap. The Roadmap lays out the long-term plan for technology development in Environmental Management and is, therefore, the primary source for Environmental Management plans regarding waste processing technologies. Waste Processing strategic initiatives include: *Improved Waste Storage Technology, Reliable and Efficient Waste Retrieval Technologies, Enhanced Tank Closure Processes, Next-Generation Pretreatment Approaches, and Enhanced Stabilization Technologies*.

I. Alternative Supplemental Treatment of LAW at Hanford, using a Systems Analysis Approach

A. River Protection Project System Plan at the U.S. Department of Energy - Office of River Protection

The Department completed its revised River Protection Project (RPP) System Plan in May 2008; it establishes a "Reference Case" that provides a description of how the mission could be accomplished and communicates the potential impacts of key issues and uncertainties on the mission. The Reference Case demonstrates how the Office of River Protection could use the Waste Treatment Plant with supplemental LAW treatment to complete the treatment and disposal of Hanford tank waste in a reasonable time frame. This case assumes that the Waste Treatment Plant presently being constructed will perform better than the minimum contractual performance requirements. Key assumptions are evaluated in several scenarios.

The Reference Case continues a description of the baseline planning for waste feed delivery, single-shell tank retrieval, and supplemental treatment with the hot commissioning and ramp-up plans for the Waste Treatment Plant. Further, it includes a path forward for tank waste with high concentrations of transuranic materials that employs disposal at the Waste Isolation Pilot Plant. Identification of those areas that might benefit from resolution of technical issues and uncertainties in the RPP Systems Plan supports the Office of River Protection's risk management efforts and is used as an important input to Technology Development planning, as discussed in the Roadmap (discussed above).

B. Evaluation of Alternative Supplemental Treatment of LAW at Hanford

In response to the HEWD Committee's concern regarding the need for a systems analysis approach to evaluation of alternative supplemental treatment of LAW, the Assistant Secretary for Environmental Management directed that an External Technical Review be performed of completed and planned systems analyses associated with LAW processing at Hanford. Specifically, the review is evaluating these topics: the present systems planning at the Office of River Protection, the path forward for LAW disposition, and options associated with increased LAW melter capacity. The lines of inquiry for this review include the following technical subject matter: the overall River Protection Project flowsheet; variables that impact LAW processing effectiveness, such as aluminum removal and sodium/caustic recycle; and balance of plant considerations (e.g., electricity, cooling capacity). Four independent experts in appropriate disciplines have been recruited to perform this review. An initial on-site portion of the review was completed during the week of March 24, 2008, along with a follow-up trip June 11-13, 2008. It is anticipated that a final review report will be completed in July 2008.

In parallel, the Office of River Protection has directed the Waste Treatment Plant contractor to perform engineering studies that will provide further detailed information regarding the relative feasibility and potential impacts of the options associated with increasing LAW melter capacity. These preliminary studies were completed by the Waste Treatment Plant contractor in late May 2008 and are presently under review by the Department. These reports, along with the revised RPP System Plan (discussed in Section I.A, above) will provide additional insight concerning LAW processing to inform on-going systems planning and analysis.

C. Applicable Engineering Technology Development

Approaches to implementing the technology needs for treatment of LAW at Hanford are in the *Next-Generation Pretreatment Approaches* and *Enhanced Stabilization Technologies* strategic initiatives of the EM Engineering and Technology Roadmap.

Bulk vitrification technology has been under evaluation for several years and is considered to be at an advanced stage of technology development. Completion of the ongoing site engineering work to prepare a preliminary documented safety analysis, in accordance with DOE Order 413.3A, can provide sufficient technical basis to support the design of a full-scale facility if DOE decides to take this technology further. In addition, tasks have been identified that advance the technology development of potential alternatives to bulk vitrification for supplement treatment of LAW, such as Fluidized Bed Steam Reforming.

Additional activities encompass the identification and development of technologies that allow pretreatment of liquid waste in order to reduce the amount of waste processed. The key tasks in this area that began work in fiscal year 2008 are:

- Continue development of the in-riser rotary microfilter for Savannah River and Hanford deployment. This solid/liquid separation technique provides exceptional performance over the existing baseline technology.
- Complete the testing of resins suitable for use in in-riser ion exchange systems that could be used to treat salt wastes prior to the turnover to the larger projects such as the Waste Treatment Plant at Hanford and the Salt Waste Processing Facility at Savannah River.
- Complete the fabrication of the pilot-scale test apparatus for fractional crystallization, which separates radioactive from non-radioactive solids, as a potential supplemental pretreatment for Hanford.
- Develop advanced mixing models to predict mixing times in large diameter tanks in support of optimized plant operations. This will reduce the wear on large-scale pumps required for tank farm operations.

II. Increased Waste Loading in Glass

A. Technology Need

Current high-level waste immobilization efforts at Savannah River and those planned at Hanford are based on a high temperature vitrification process in which high-level waste is mixed with a pre-fabricated glass frit at Savannah River or glass-forming chemicals at Hanford and melted. The resulting glass product is poured into steel canisters for ultimate disposal in a geologic repository. Development of the capability for increasing the high-level waste loading in glass (i.e., the amount of waste per canister) offers the potential for life-cycle cost savings in the DOE complex, by reducing the total number of canisters to be sent to the geologic repository. However, the cost and schedule of high-level waste treatment also depends on the rate of high-level waste glass production, i.e., the number of canisters produced over a given time frame. Therefore, reducing life-cycle costs for the DOE complex is dependent on both waste loading and glass production rates.

Current estimates for the number of glass canisters to be produced from the Hanford high-level waste range from under 10,000 to nearly 30,000. Similarly, current estimates indicate that the Defense Waste Processing Facility at Savannah River will produce between 6,300 - 7,900 canisters while operating from 1996 through 2028 or 2035.

Currently, there are two primary technical limitations that occur as high-level waste loading in glass is increased:

- crystallization, which can potentially impact melter processing or glass performance, takes place; and
- lower melt rate or production rate (which has actually been observed with the current melters at the Defense Waste Processing Facility).

To support continuous improvement of waste throughput, the next generation of Waste Treatment Plant and Defense Waste Processing Facility glass formulations is needed.

B. Previous Environmental Management Technology Development

Environmental Management technology development activities to improve loading of high-level waste in glass for Hanford and Savannah River sites are presently underway. The results of these activities are highlighted below to link the basis for further technology investments with expected benefits:

- Hanford high-level waste glasses containing higher levels of aluminum (which adversely impact waste loading) were tested by a team of researchers from Pacific Northwest National Laboratory, Khlopin Radium Institute, and the Savannah River National Laboratory. Additional testing with high aluminum-content glasses was performed at the Vitreous State Laboratory, located at Catholic University. Information from these two programs was used to support development efforts for a recent Defense Waste Processing Facility process run ("Batch 4"), supporting Defense Waste Processing Facility processing waste at the highest levels since radioactive operations began.
- Glasses for increased loading were formulated for Defense Waste Processing Facility's next processing run ("Batch 5") in support of the Sludge Mass Reduction task, supported by the Environmental Management Technology Development program.
- A method for controlling glass composition to avoid crystal accumulation was proposed by the Pacific Northwest National Laboratory and proof-of-principle testing was successful. Work to further demonstrate this formulation concept began in fiscal year 2007.
- Initial computer models for qualitative description of composition impacts on Defense Waste Processing Facility's melting rate were developed by Savannah River National Laboratory and are being expanded with additional data at higher waste loadings to make predictive models for waste loading optimization.
- Glasses for higher concentrations of bismuth, phosphorous, and sodium (plus aluminum) were formulated and tested at the Vitreous State Laboratory. It was determined that the bismuth limits are above those previously planned for, which should translate into higher waste loadings for bismuth-limited waste streams when processed at the Waste Treatment Plant. Glass compositions for waste with

bismuth and phosphorus are particularly important at Hanford where the bismuth-phosphate nuclear materials separation process was conducted.

C. Ongoing Environmental Management Technology Development

Consistent with the EM Technology Roadmap strategic initiative for *Enhanced Stabilization Technologies,* a program to accomplish increased waste throughput at Hanford and Savannah River has been implemented. The program will systematically evaluate key waste glass data requirements and integrate with advanced melter technology development based on domestic and international experience.

The challenges related to increasing the loading of high-level waste in glass, while maintaining adequate properties and processing rates, have been divided into nine tasks. Those tasks which began work in fiscal year 2008 are as follows:

- Broader Hanford high-level waste glass formulations are aimed at expanding the formulations for Waste Treatment Plant to cover more of the wastes currently stored at Hanford. This activity is exploratory in nature. Glasses are formulated and tested for individual representative wastes. Once a successful glass is developed, it is tested at melter scale to determine processing rate.
- Formulation of high-waste loading Defense Waste Processing Facility glasses is aimed at formulating glasses with high waste throughput for Savannah River wastes. Glasses spanning a range of loadings are formulated, fabricated, and tested. Melting rate tests are performed on the successful glasses to determine the optimal waste throughput rate.
- Crystal tolerant glass formulations are aimed at developing a new formulation method that replaces conservative crystal fraction limit with predicted crystal accumulation rates. The method, if successful, could result in a step function increase in waste loadings.
- The Next Generation Melter glass formulations task is aimed at developing glass formulations that can take advantages of the distinct capabilities of new melter technologies.
- Experimental methods for assessing the melting rate of Hanford high-level waste glasses are being developed at the Pacific Northwest National Laboratory. These methods will be used to improve melting rate of highly waste-loaded Hanford high-level waste glasses.
- A glass property database is under development at the Pacific Northwest National Laboratory and the Savannah River National Laboratory to expand the knowledge of high-level waste glass compositions. The database will be used to expand glass property models at the Waste Treatment Plant and the Defense Waste Processing Facility to allow for a broader range of waste compositions to be treated and higher waste loadings to be processed.

III. Improving Methods to Remove Non-Radioactive Components from Sludge Heels in High-Level Waste Tanks

A. Technology Need

The removal of non-radioactive components of sludge offers several advantages that would be beneficial to the Hanford and Savannah River sites. An aspect of the need to treat the heels from tank sludge is that heels represent mostly hard-to-retrieve material at the bottom of each tank. The tank heel contribution to radioactive waste loading issues is typically not that significant; the methodology used to remove the non-radioactive sludge components can result in a substantial impact on the waste loading in glass. If these sludge components can be removed using a leaching process (the chemical removal of soluble constituents), the volume of high-level waste can be significantly reduced, thus shortening the operating life cycle significantly. For example, the current aluminum leaching process in use in the Savannah River tank farms is estimated to reduce the operating life cycle by about 7 years and decrease the number of high-level waste canisters to be disposed in the geological repository by about 1000. At Hanford, the leaching processes under development have the potential to reduce the operating life cycle of the Waste Treatment Plant by up to 20 years and substantially decrease the number of canisters to be disposed in the geological repository.

The primary constituents targeted for removal by leaching are aluminum and chromium. Too much aluminum can increase the sludge viscosity, glass viscosity, and reduce overall process throughput. Chromium leaching is necessary to prevent formation of crystalline compounds in the glass, but is only needed at Hanford because of differences in the sludge waste chemistry. Improving glass formulations to increase tolerance of aluminum and chromium (discussed above) is another approach to decrease high-level waste glass volume.

There are several technical issues associated with the leaching processes for either aluminum or chromium. Disposal of the aluminum-and-chromium-rich secondary waste streams produced by leaching must be addressed at both Hanford and Savannah River. As previously mentioned for Hanford, the aluminum leaching process presently uses large amounts of sodium hydroxide to leach the aluminum; the development of a caustic recycle process is ongoing and requires additional work to reduce identified technical risks. The removal of chromium is complex and presents additional potential technical challenges; additional technology development is needed to understand the impacts to the Waste Treatment Plant.

B. Previous Environmental Management Technology Development

For several years, the Office of Waste Processing has been investigating methods for the recovery of the inventory of sodium hydroxide that would be required to perform the Waste Treatment Plant caustic leaching process. The use of specifically designed ceramic membranes has been examined to separate sodium from radioactive sodium-based waste, in order to reduce the weight and volume of the waste to be treated and disposed. If successfully demonstrated, this process will allow clean caustic (sodium hydroxide) to be reused, resulting in substantial projected savings on radioactive waste processing. Additional work is ongoing to ensure the successful integration of the ceramic technology, or other caustic recovery technologies, into the Waste Treatment Plant.

Additionally, an Environmental Management-sponsored workshop was held in 2007 to discuss the status of a number of chemical processes to leach constituents from high-level waste sludges at both Hanford and Savannah River. This workshop examined the high-level waste process flowsheet for each site, discussed the status of knowledge of the leaching processes, communicated the research plans, and identified opportunities for synergy to address knowledge gaps. This workshop resulted in a number of leaching projects to be considered for further technology development consistent with the Environmental Management Engineering and Technology Roadmap are discussed further below.

C. Ongoing Environmental Management Technology Development

Approaches to implementing the technology needs for removing non-radioactive constituents from the highly radioactive sludges at Savannah River and Hanford are in the *Next-Generation Pretreatment Approaches* strategic initiative. The purpose of the technology development program is to develop engineered solutions that more effectively separate inert materials and LAW from high-level waste, such that only the high-level waste fraction is stabilized for geologic disposal. Among the key challenges being addressed at this time is the development of technology solutions that would allow for the removal of large amounts of aluminum from high-level waste sludge at Savannah River and Hanford in order to reduce the burden on the high-level waste vitrification facilities. Additionally, a significant fraction of predicted sludge batches at Hanford are limited by the chromium content. Technologies are needed to advance the understanding of chromium-oxidants and their impact on downstream processing.

The key tasks in this initiative area in fiscal year 2008 are:

- Develop an understanding of aluminum leachate stability to improve the utility of caustic recycle.
- Continue studies on sludge mass reduction aimed at developing in-tank aluminum leaching alternatives for Savannah River and Hanford.
- Develop and evaluate technologies for increasing boehmite (a challenging chemical phase of aluminum) dissolution as a means to reduce the aluminum burden in high-level waste.
- Continue to develop the Continuous Sludge Leaching process, a promising near-tank aluminum removal technology, presently entering engineering scale development.

References

1. U.S. Department of Energy, Savannah River Site Office, *Life-cycle Liquid Waste Disposition System: Liquid Waste Planning Process, Revision 14,* LWO-PIT-2007-00062, 2007.
2. U.S. Department of Energy, Office of River Protection, *River Protection Project System Plan*, ORP-11242, Revision 3, May 2008.
3. U.S. Department of Energy, Office of Environmental Management, *Engineering and Technology Roadmap*, March 2008, Washington, DC
4. Certa, P.J., T.M. Hohl, A.M. Johnson, S.L. Orcutt, R.S. Wittman, and D.S. Kim, 2005. *Sensitivity of Hanford Immobilized High-level Waste Glass Mass to Chromium and Aluminum Partitioning Assumptions*, RPP-20003, Revision 1, CH2M Hill Hanford Group, Richland, Washington.
5. Gillam, J.M., H. Shah, M. Rios-Armstrong, Sludge Batch Plan, CBU-PIT-2005-00144, Revision 1, August 7, 2006.
6. Gillam, J.M., Aluminum in Waste Tank Sludge: History and Status, CBU-PIT-2006-00068, July 11, 2006.
7. Hamm, B.A., J.M. Gillam, and H.B. Shah (2007). *Savannah River Site Liquid Waste Planning Process: Sludge Batch Plan*, CBU-PIT-2005-00144, Revision 2, U.S. Department of Energy, Savannah River Office, Aiken, South Carolina.
8. Kim, D.S., D.K. Peeler, and P. Hrma (1995). "Effects of Crystallization on the Chemical Durability of Nuclear Waste Glasses," *Ceramic Transactions* 61, pp. 177-185, American Ceramic Society, Westerville, Ohio.
9. McCabe, D.J., R. A. Peterson, J. A. Pike, and W. R. Wilmarth, "Aluminum and Chromium Leaching Workshop Whitepaper," WSRC-STI-2007-00168, April 25, 2007.
10. National Research Council of the National Academies, *Technical and Strategic Advice for the Department of Energy, Office of Environmental Management's Development of a Cleanup Technology Roadmap: Interim Report*, dated February 14, 2008. http://www.nap.edu/catalog.php?record_id=12165
11. Raszewski, F.C., T.B. Edwards, and D.K. Peeler (2008). *Enhanced DOE High-Level Waste Melter Throughput Studies: SRNL Glass Selection Strategy*, WSRC-STI-2007-00652, Savannah River National Laboratory, Aiken, South Carolina.
    Vienna, J.D., P. Hrma, A. Jiricka, D.E. Smith, T.H. Lorier, I.A. Reamer, and R.L. Schulz (2001). *Hanford Immobilized LAW Product Acceptance Testing: Tanks Focus Area Results*, PNNL-13744, Pacific Northwest National Laboratory, Richland, Washington.
12. Wilmarth, W. R., D.T. Hobbs, W.A. Averill, E.B. Fox, and R.A. Peterson, "Review of Ceramatec's Caustic Recovery Technology," WSRC-STI-2007-00366, Revision 0, July 20, 2007.

Note: Citations were not included in the report for reader ease; the above sources were used to develop this report.