Integration of National Laboratory and Low-Activity Waste Pretreatment System Technology Service Providers - 16435

Karthik Subramanian*, Mike Thien*, Dawn Wellman**, Connie Herman*** *Washington River Protection Solutions, LLC **Pacific Northwest National Laboratory *** Savannah River National Laboratory

Abstract

The National Laboratories are a critical partner and provide expertise in numerous aspects of the successful execution of the Direct-Feed Low Activity Waste Program. The National Laboratories are maturing the technologies of the Low-Activity Waste Pretreatment System consistent with DOE Order 413.3B, "Program and Project Management for the Acquisition of Capital Assets," expectations. The National Laboratories continue to mature waste forms, i.e., glass and secondary waste grout, for formulations and predictions of long-term performance as inputs to performance assessments. The working processes with the National Laboratories have been developed in procurements, communications, and reporting to support the necessary delivery-based technology support. The relationship continues to evolve from planning and technology development to support of ongoing operations and integration of multiple highly coordinated facilities.

Introduction

Radioactive and chemical wastes from nuclear weapon production are stored in underground tanks at the Hanford Site, located in the state of Washington. The waste tanks contain a complex and diverse mix of radioactive and chemical waste in the form of sludge, salts, and liquids, necessitating a variety of unique waste retrieval, treatment, and disposition methods. Generically, the tank waste can be characterized as the following:

- Sludge Insoluble materials largely consisting of metal hydroxides and oxides that precipitated when acidic wastes from spent nuclear fuel processing and other activities were neutralized and converted to high pH for storage in carbon steel tanks. The sludge waste makes up the largest component that will be processed via high-level waste (HLW) vitrification into a stable glass form.
- 2. Supernatant Liquid waste with high sodium content and high pH.
- 3. Saltcake A mixture of salts that precipitated from supernatant as the specific gravity was increased by evaporation to reduce tank storage space requirements. Saltcake must be re-dissolved and processed as supernatant waste. The supernatant and saltcake contain the majority of highly radioactive cesium which must be separated and processed with the sludge stream into HLW glass. The decontaminated supernatant will be processed via low-activity waste (LAW) vitrification into a stable glass form.

4. Potential contact-handled transuranic waste (CH-TRU) – A mixture of sludge and saltcake consisting of some 1.4 million gallons in 11 specific single-shell tanks (SSTs). The material in these tanks is being reviewed to determine the potential to transfer to the Waste Isolation Pilot Plant (WIPP) versus being processed on-site into HLW and LAW glass fractions.

In order to begin immobilization of tank waste as soon as practicable, a Direct Feed LAW (DFLAW) flowsheet has been developed. In the DFLAW configuration, tank waste liquids will be provided to the LAW Pretreatment System (LAWPS). The LAWPS will separate the HLW and LAW fractions and provide qualified feed to the Hanford Tank Waste Treatment and Immobilization Plant (WTP) LAW vitrification facility. Successful startup and operation of DFLAW requires the completion of engineering, design, and construction of numerous facilities, flowsheet steward-ship, programs integration across facilities, generation of a series of permits, and development of the regulatory framework to dispose of the waste forms generated.

The National Laboratories (NLs) are an integral part of delivering the DFLAW mission, and are a key technical and technology resource for multiple facets of the DFLAW program. The NLs are involved in the necessary operational base operations technical support, LAWPS technology maturation, performance assessments, flowsheet optimization, and technology roadmap development. The One-System manages a cohesive NL support program through the Chief Technology Office (CTO). The NLs are providing the critical technology maturation activities in support of the LAWPS project, such as resin dump system testing and support to the integrated scale testing. The NLs are developing the critical waste form performance data packages utilized as input to the Integrated Disposal Facility (IDF) performance assessment (PA). These packages provide the performance data for the LAW glass waste form and the low-temperature secondary liquid and secondary solid waste forms for modeling purposes. The NLs are embedded within the flowsheet organization to provide continuous support to flowsheet stewardship and optimization, as well as execute the maturation activities.

National Laboratory Involvement in DFLAW

The NLs are integrated into the DFLAW program in numerous capacities, primarily through direct project support/technology maturation and flowsheet/programmatic optimization. A summary of these activities as related to the DFLAW program are shown in Figure 1.



Figure 1. Integration of National Laboratories with the DFLAW Program and Flowsheet

Management of NL Involvement

The specific NLs performing DFLAW scope are Pacific Northwest National Laboratory (PNNL), Savannah River National Laboratory (SRNL), and Oak Ridge National Laboratory (ORNL). In addition to these NLs, the Vitreous State Laboratory (VSL) at the Catholic University of America (CUA) is also involved with the program. Each of these Laboratories brings specific competencies in support of the DFLAW program and is collaboratively working towards successful deployment of DFLAW.

The NL involvement within the DFLAW program is managed through the CTO. Management of the NL scope is accomplished through the establishment of a core communication and management team that includes the CTO and a lead for each of the Laboratories. Several tools have been used to institutionalize the NL support in a delivery-focused manner to ensure the successful deployment of DFLAW:

- Embedded NL staff: The CTO has embedded staff from the Laboratories that support the development of technology maturation scope across the scope of DFLAW. These embedded staff provide the necessary day-to-day input to development of scope as well as execution of scope. The early input from the Laboratories in construction of the experimental programs has been critical to program execution. In addition, the embedded staff provide the conduit for reachback into each respective Laboratory for specific technical support.
- NL core leadership: The Laboratories have identified specific leads for Office of River Protection (ORP) support providing a single conduit for the variety of scope each Laboratory is performing. Engaging this core leadership team has been critical in developing and execution of scope. In addition, this team has provided the necessary leadership for corrective actions when necessary.
- Coordinated monthly reporting: The Laboratories are utilizing an earned value management system type tool to report on a monthly basis to ensure program-wide understanding of laboratory involvement. In addition, this provides the necessary focus of cost and schedule to ensure timely delivery of procurements as well as scope execution. The monthly reporting mechanisms culminate in a conference call where scope, cost, and schedule are discussed. This coordination continues to evolve to ensure successful collaboration
- Scope planning enhancements: The planning process for fiscal year laboratory integration and scope typically began in the first quarter for each fiscal year. However, the planning for FY16 planning was accomplished through a two-day workshop held in June of 2015. Although the budget and scope for FY16 were not yet finalized at the time, the collaboration between the Laboratories, ORP, and Washington River Protection Solutions allowed for early planning and procurement of scope when FY16 began in October. As a result of the early collaboration, the scope was planned and all procured early in FY16 in contrast to previous years.

Direct Project Support and Technology Maturation

The direct project support includes significant NL support to the LAWPS, DFLAW flowsheet optimization, and waste form performance data development as inputs to the IDF PA. A detailed technology maturation plan (TMP) has been developed in support of the LAWPS project. The TMP describes the necessary activities to mature the technology in concert with project execution. The flowsheet optimization work being performed by the NL includes specific experimentation to optimize corrosion control requirements for returns to the tank farms to minimize volume influents and conserve tank space. The waste form performance experimentation includes long-term testing of LAW glass and solid and liquid secondary waste forms to determine necessary input parameters for long-term fate and transport modeling for the performance assessment.

LAWPS Technology Maturation

The LAWPS will separate HLW components from LAW components by filtration and ion exchange and store the low-activity solution. From the storage system, the LAW will be fed to the WTP LAW facility for conversion to a solidified waste form. The LAWPS focuses on the separation of HLW from the feed stream to result in LAW. Additionally, the LAWPS allows the potential to draw down double-shell tank storage volume and the WTP to begin vitrifying low-activity tank waste earlier than the current projection.

A diagram of the LAWPS flowsheet is shown in Figure 2.

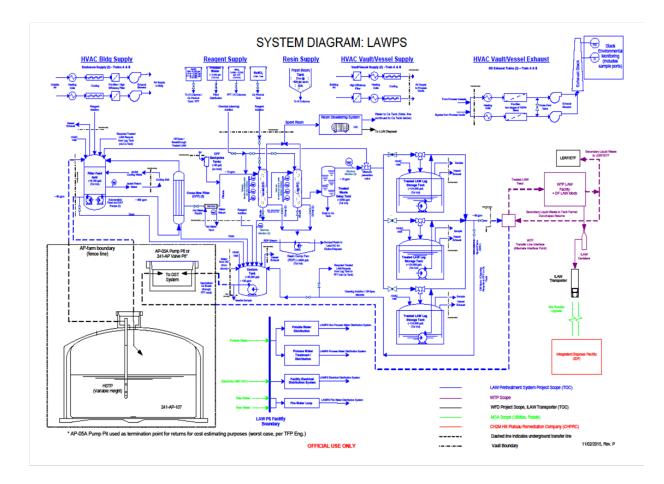


Figure 2. DFLAW System Diagram.

A detailed technology maturation program has been developed to guide the necessary development and experimentation as inputs to the project execution. The specific scope of the LAWPS technology maturation is shown in Table 1.

Table 1 Summary of NL Scope for LAWPS Technology Maturation

National Laboratory	Scope
PNNL	 Bench-scale cross-flow filter performance testing with low solids
	 Integrated scale testing simulant development
	 Ion-exchange column hydrogen generation and retention analysis
	4. Expanded isotherm testing for increasing

	sodium molarity to ion exchange feed
SRNL	1. Technology maturation plan development
	2. Cross-flow filter media corrosion testing
	 Resin dump tank proof-of-concept development
	 Ion-exchange column performance modeling
ORNL	Confined swelling pressure testing sRF resin

Waste Feed Qualification and Flowsheet Optimization

The NLs are embedded within the flowsheet organization of One-System, in part, to construct a waste feed qualification program for DFLAW as well as develop and execute the experimentation for flowsheet optimization.

The feed qualification scope matures the DFLAW integrated feed qualification laboratory-scale processes to qualify batches to be processed through DFLAW. The scope potentially involves demonstrating laboratory-scale processes to show they can be used to project key plant performance metrics. The DFLAW Integrated Flowsheet includes solids separation by cross-flow ultrafiltration, cesium ion exchange, and vitrification of the treated LAW. The intent of feed qualification process demonstrations is to show key plant process performance metrics are expected to be satisfied in processing the staged feed. Therefore, this work scope will involve filtering of an actual LAW feed sample at laboratory-scale to demonstrate ultrafiltration. The filtered LAW sample will then be decontaminated by cesium ion exchange

The primary research and development scope for flowsheet optimization is to evaluate the corrosion controls needed to allow transfer of DFLAW effluents to the Hanford Double Shell Tank (DST) system and evaporation of these effluents in the 242-A Evaporator. These effluents include effluents from LAWPS as well as effluents from the WTP LAW facility. The returns from LAWPS include the solids slurry from cross-flow filtration, chemical cleaning of the filters, and elution cycle effluents from ion exchange including concentrated cesium eluent. These streams are expected to be handled by the current corrosion control protocols for the DST system. The WTP LAW return stream will be generated by condensation and scrubbing of the LAW melter off-gas stream. A portion of this stream, which will contain substantial amounts of chloride, fluoride, ammonia, and sulfate ions, and potentially minor concentrations of mercury, may be returned to the tank farms for storage and evaporation. The initial DFLAW flowsheet planned on utilizing the Savannah River Site correlations to calculate the necessary quantity of inhibitor for a given chloride or sulfate concentration. This program is focused on tailoring the controls for Hanford-specific streams that expand the corrosion control envelope, minimizing the necessary inhibitor additions for tank space conservation. These corrosion control experiments are being performed primarily

by SRNL in conjunction with commercial providers. The remaining efforts focus on identifying technical gaps or opportunities for improvements within the DFLAW flowsheet.

Waste Form Performance Data Package Development

Much of the waste from DFLAW operation will be disposed of in the Integrated Disposal Facility, located at the Hanford Site. The main mission of the IDF is for the disposal of a variety of wastes as documented in the Final Tank Closure and Waste Management Environmental Impact Statement for the Hanford Site (TC&WM EIS-0391). Such wastes include the WTP and the non-WTP generated waste streams. WTP-generated wastes include the LAW glass, LAW melters, secondary solid waste and the Effluent Treatment Facility (ETF)-generated secondary solid waste.

A DOE-Disposal Authorization Statement required by DOE Order 435.1 and a Resource Conservation and Recovery Act permit need to be issued before the IDF can operate as a waste disposal facility. A PA must be prepared in support of these regulatory documents. The PA will also underpin the Waste Incidental to Reprocessing Evaluation for LAW glass disposition.

The fundamental inputs to the PA are the waste form characteristics that will be used for long-term fate and transport modeling of contaminants to prove compliance with requirements. The primary waste form characteristics for the IDF PA are performance of the LAW glass and the cementitious waste forms for the liquid and solid secondary waste primarily from the vitrification process and inclusive of all secondary wastes generated from the DFLAW flowsheet

Glass Waste Form Characteristics

The ultimate goal of LAW glass program for IDF PA support is to provide the PA modeling team with relevant data of LAW glasses that are expected to be produced in the WTP LAW facility. Glass waste form data collection and modeling to support the IDF PA has occurred over the past 19 years. The results from these efforts have been summarized in several reports and data packages. These documents discuss the results collected on a variety of LAW glass compositions ranging from the initial glass composition, LD6-5412, selected by the Tank Waste Remediation System program, to the current WTP baseline glasses (e.g., LAWA44, LAWB45, and LAWC22) and up to the currently and the still ongoing development of newly proposed enhanced/advanced glasses.

The IDF PA will employ modeling software to determine the anticipated performance of waste forms. To provide key input parameters for near-field reactive transport modeling, well-constrained and interpretable experiments have been performed to isolate and parameterize the key mechanisms of glass corrosion. When glass contacts with water, three glass corrosion mechanisms are postulated to occur that are applicable to the IDF: kinetic rate law, ion exchange, and the effects of secondary phase formation.

The following are parameters or set of data that are relevant for the IDF PA:

- Kinetic rate law parameters: At present, a kinetic rate law, based on Transition State Theory, is considered to best describe the network hydrolysis and matrix dissolution of glass. These parameters are obtained from the analysis of data produced in the Single-Pass Flow Through Test (SPFT), typically performed to ASTM Standard C1662. Ion exchange rate, which is an important parameter to quantify in addition to the rate law parameters, is also obtained from SPFT data analysis.
- Secondary phase reaction networks: As cations migrate (via ion exchange) and/or the components of the dissolved glass (via corrosion) accumulate and reach saturation, secondary phases form. The secondary phases can either slow or accelerate glass corrosion. The effects of secondary phase formation is another process that is taken into account when modeling long-term behavior of glass in a disposal system. Key secondary phases can be identified from data collected from Product Consistency Test (ASTM C1285), Pressurized Unsaturated Flow Test, or Vapor Hydration Test (ASTM C1663).

The 2015 Immobilized Low-Activity Waste (ILAW) Glass Data Package for the IDF PA encompasses the kinetic rate law parameters and the secondary reaction networks of glasses that fall under the 'baseline' correlation. To improve the throughput of the WTP, the glass compositions have to expand toward higher alkali and sulfate contents, which means that the enhanced/advanced ILAW glass correlation will likely be applied in the WTP operation. In line with this, 'enhanced/ advanced' glasses are subject to the IDF PA and entail testing. PNNL and VSL are performing the necessary IDF PA data package development testing in concert with the DOE-ORP led glass composition program to support implementation of the advanced glasses into WTP LAW.

Cementitious Waste Forms Characteristics

The secondary liquid wastes treated in the Hanford ETF are planned to be solidified and immobilized in cementitious waste forms and disposed of in the IDF. Hanford ETF currently treats secondary liquid waste streams including evaporator condensates from the 242A Evaporator, Environmental Restoration Disposal Facility and IDF leachates, laboratory wastes, and groundwater liquid wastes. The concentrated brine from the secondary treatment train within the ETF is currently fed to a thin film drier, producing a powdered salt waste form for disposal in drums. The ETF is also expected to treat secondary liquid waste from WTP operations, including DFLAW immobilization. Whether the solidification of the secondary liquid wastes is done on-site or off-site, a cementitious waste form that meets requisite performance characteristics is necessary. The development of cementitious waste form has begun using the Cast Stone formulation, after which modifications will be applied based on the waste stream composition. Modifications can include 1) adding or modifying reagents composition such as increasing/decreasing blast furnace slag (or addition of lime to enhance the retention capability of the waste form for contaminants of concern (COC), 2) adding getters that will capture specifically targeted COCs such as technetium (Tc-99) or iodine (I-129), and possibly other ways to improve the capabilities of the waste form to effectively immobilize the secondary liquid waste stream. The model used for cementitious waste form performance for previous PAs was a diffusion-controlled release model. Key data or values for the diffusional release model need to be determined for each cementitious waste form formulation, such as the porosity, effective diffusion coefficient, distribution coefficients and saturation.

The IDF PA is considering a modeling approach that will take into account the chemical changes to the waste form such as oxidation, carbonation, and sulfate attacks. This approach will also be based more on the fundamental properties of the waste form materials and how they change over time. The secondary waste cementitious waste form development program is measuring waste form properties to support the diffusional release model. The efforts are being done in collaboration with the PA team and will support data acquisition activities for the PA modeling approach.

The waste form qualification activity is composed of work to 1) demonstrate that the waste form will effectively immobilize and stabilize the targeted waste stream (through contaminant leach rates, compressive strength), 2) demonstrate the solidification process with waste forms prepared from actual and/or spiked radioactive wastes, and 3) provide long-term waste form performance data and information on degradation and release mechanisms to support the IDF PA to meet DOE Order 435.1 disposal requirements. Properties of the individual dry blend materials, fresh properties of each mix, and the properties of the cured (or hardened) samples are being characterized. These properties will predetermine or screen the cementitious formulations that will be subjected to further tests to obtain the IDF PA-relevant data.

Summary

The NLs are integrated into the DFLAW program in numerous capacities, primarily through direct project support/technology maturation and flowsheet/programmatic optimization. Management of the NL scope is accomplished through the establishment of a core communication and management team that includes the CTO and a lead for each of the Laboratories. Each of these Laboratories brings specific competencies in support of the DFLAW program and is collaboratively working towards successful deployment of DFLAW. The program is committed to the integration of the needed technical expertise of the NLs in the complex technical scope for the tank waste disposition mission at Hanford.