Solid Secondary Waste Form Testing Activities for the Integrated Disposal Facility Performance Assessment Support - 17567

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ABSTRACT

Solid secondary waste (SSW) that will be generated at the Waste Treatment and Immobilization Plant (WTP) and from other activities at the Hanford site is destined for disposal at the Integrated Disposal Facility (IDF). Construction of the IDF was undertaken based on a Performance Assessment (PA) performed in 2001. In 2016, a SSW data package was prepared to support the development of an updated PA (2017 IDF PA).

There are many inputs required to support the IDF PA. Data needs were identified based on experience from the previous PAs and from the development of conceptual models and modeling approaches to be applied for the 2017 PA. Interactions with the IDF PA team and the inventory data package developer helped to focus detailed efforts on a few key waste streams, contaminants of concern, and disposal configurations that are expected to be the primary contributors to the dose resulting from SSW disposed at the IDF.

The key waste streams include: High Efficiency Particulate Air (HEPA) filters, ion exchange resins, granular activated carbon, and Ag-mordenite. The key parameters addressed in the 2016 data package include: saturated hydraulic conductivity, moisture characteristic curves, effective diffusion coefficient, distribution coefficient and porosity. The IDF PA team also identified a set of key contaminants that are expected to be the primary concern for the PA calculations (Tc-99, I-129, Cs-137, Sr-90, uranium isotopes (and total uranium), chromium, mercury and nitrate). These contaminants were the focus of the SSW data package.

Specific formulations had not been identified for cementitious materials that will be used to encapsulate or stabilize SSW, and no IDF-specific experiments had been conducted to obtain the data for the PA. Thus, for the 2016 SSW data package, recommended property values were provided for a range of representative candidate materials based on a review of existing literature. However, starting in FY17, IDF-specific experiments will be performed for Hanford SSW expected to contribute most significantly to the dose for the IDF. This paper will summarize the information obtained for the 2016 data package and the plans for further testing to refine key inputs for the IDF PA.

INTRODUCTION

Background

The solid secondary waste is one of the wastes expected to be generated as a result of Hanford Waste Treatment & Immobilization Plant (WTP) operation. Briefly, the Hanford Site, in south central Washington State, had been used extensively for production of defense materials by the U.S. Department of Energy (DOE) and its predecessors. Radioactive wastes were generated over four decades during the processing of nuclear fuel to produce plutonium for the nation's nuclear weapons arsenal. The Hanford River Protection Project (RPP) mission is to safeguard the nuclear waste stored in Hanford's 177 underground tanks and to manage the waste safely and responsibly until it can be treated in the WTP for final disposition.

In order to immobilize the Hanford tank wastes as soon as practicable, Direct Feed Low Activity Waste (DFLAW) operation is planned to be implemented. In DFLAW, low activity waste (LAW) feed will be provided to the LAW Pretreatment System (LAWPS). The LAWPS will separate the HLW and LAW fractions and provide qualified feed to the WTP-LAW Vitrification Facility. The HLW fraction will be returned and stored to the double shell tanks (DST) until HLW WTP vitrification facility is in operation. The vitrified LAW and other WTP-generated waste forms will be disposed in the Integrated Disposal Facility (IDF) located in Hanford.

A variety of low level wastes (LLW) will be disposed in the IDF, as documented in DOE/EIS-0391 [1]. The wastes include those generated by WTP operations (includes the SSW), and the non-WTP-generated waste streams that are estimated in the inventory data package [2]. These wastes are depicted in Figure 1, SSW is highlighted to show where this stream originated. SSW is generated from WTP and Effluent Treatment Facility (ETF) (spent consumables and plant equipment that wore out or failed during the treatment mission).

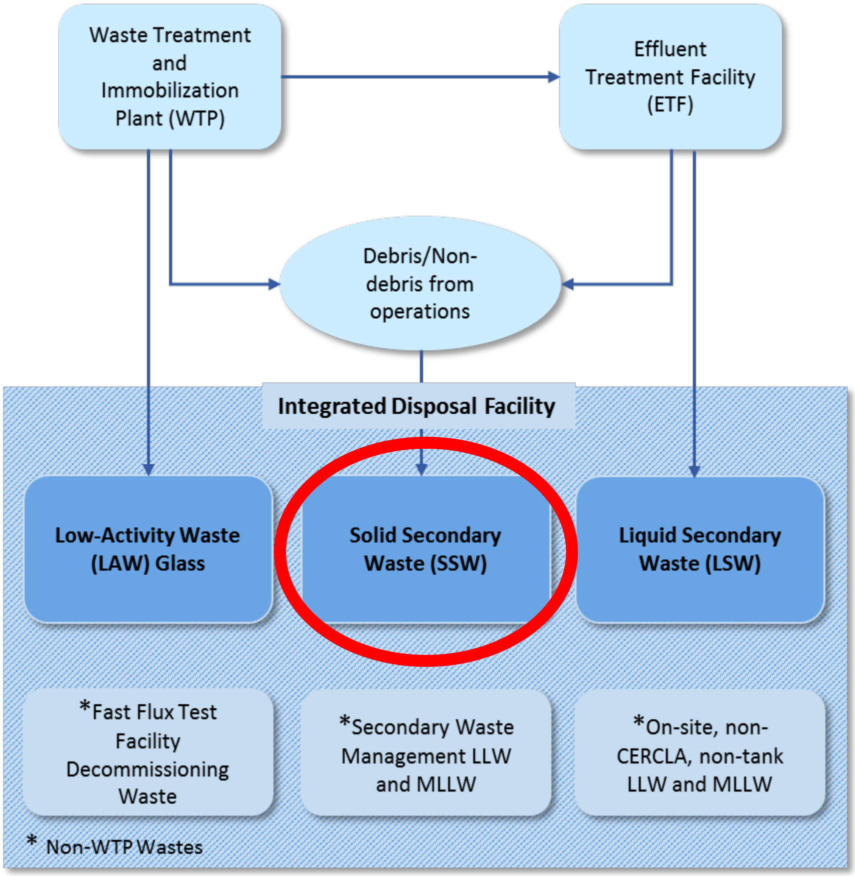

Figure 1. Wastes to be disposed at the Integrated Disposal Facility

Performance Assessment (PA) for IDF Disposal

Before the waste forms of SSW and other LAW or LLW will be disposed in the IDF, DOE must conduct an IDF Performance Assessment (PA). The long-term performance of the IDF to contain contaminants of potential concern (COPC) that have been immobilized in the waste form to be disposed in the IDF is evaluated in the PA. The 2017 IDF PA will be performed in accordance with DOE Order 435.1, Radioactive Waste Management [3]. This PA is an update to the 2001 Immobilized Law Activity Waste PA [4]. The approval of the 2017 IDF PA will support and obtain the permitting of waste disposal at the IDF.

The PA uses integrated numerical models, representing the engineered (the waste forms) and natural systems, to evaluate the long-term performance of the IDF. To run the model, relevant data are needed. Such relevant data are obtained by testing the waste forms.

Purpose and Scope of SSW Form Testing Project

The purpose of the SSW form development testing project is to obtain the relevant data for the IDF PA. The first task is to obtain information relevant to the development of SSW from sites that generate SSW counterpart waste forms in the US and other countries. Formulation, development and testing of waste forms specific to Hanford SSWs will then follow. SSW that will be disposed in the IDF will be stabilized in a cementitious waste form.

The scope of the project is associated with technology maturation and analysis of waste forms for the disposal of secondary solid wastes generated by the DFLAW Program. Waste forms must be compatible with the IDF Waste Acceptance Criteria and Resource Conservation and Recovery Act (RCRA) permit requirements [5]. Work will focus on providing data for use in the 2017 IDF PA which is needed to obtain the waste disposal permit and the subsequent PA maintenance activities.

This paper lays out the conceptual models applied in the IDF PA for the cementitious waste form of SSW, the specific waste streams that are treated as SSW and the work that are done or planned for the development and testing of Hanford SSW cementitious waste forms. Testing activities of waste forms will provide the key parameters or data needed to perform the IDF PA.

CONCEPTUAL MODELING AND PARAMETERS RELEVANT FOR IDF PA

Conceptual Models Applied in the IDF PA

When a waste form has hydraulic conductivity that is much lower than that of the backfill, which is true for the SSW cementitious waste form, subsurface water flow through the near field is expected to be diverted around the waste form. There are two mechanisms of COPC transport; i) diffusion of the radioactive or hazardous constituent within the waste form that constitutes the primary mechanism for constituent release from the waste form into the backfill surrounding the waste form, and ii) advective flow transports the diffused material to the bottom of the facility into the vadose zone.

For the 2017 IDF PA model, two waste form release modeling approaches are being considered for the cementitious waste forms that will be disposed at the IDF.

i) Diffusion-Limited Release Rate Model. In the diffusion-limited release models, diffusion of the solute through the waste form occurs only in the liquid-filled pore space of the waste form, and partitioning of constituents between the solid and liquid phases is included in the release model.

ii) Advection-Diffusion Source Term Release Model. This is a multi-dimensional transport model that accounts for both diffusion and advection through the cementitious waste form as the properties and integrity of the waste form change over time.

Data Needs: Parameters Needed for the Conceptual Modeling

The key parameters that are required to perform either or both the models are the following:

A. Hydraulic properties of waste forms and environmental media

1. Saturated hydraulic conductivity (K_s), Porosity (n), Dry bulk density (ρb)

2. Moisture characteristic curves for unsaturated moisture transport

   Moisture retention properties of samples can be determined using laboratory measurements of pressure head and moisture content. Curve fitting will be used to estimate the parameters in the van Genuchten equation defining the moisture retention curve.

B. Effective diffusion coefficients of waste forms
Effective diffusion coefficient (D_e) is a physical property, not dependent on species-specific sorption and/or solubility. Popular experimental techniques for measuring diffusion coefficients are ANSI/ANS 16.1 method (ANSI/ANS-16.1-2003 reaffirmed 2008) [6] and EPA-1315 [7].

C. Contaminant speciation, partitioning, and solubility. The basic philosophy used to develop and to parameterize the models describing the interactions between waste forms and environmental media involved using: 1) basic mechanistic studies primarily reported in the literature to provide guidance for the conceptual geochemical models, and 2) empirical studies, preferably with site-specific materials and conditions, to provide input values to help quantify the conceptual models. The three parameters used to describe solute interaction with the solid phase cementitious waste forms are:

1. Distribution coefficient (K_d values). The K_d value is the simplest construct describing contaminant sorption to cementitious materials. The Kd value is the ratio of the contaminant concentration sorbed to the solid phase divided by the contaminant concentration in the liquid surrounding the solid phase.

2. Apparent solubility concentration (K_s). In addition to the K_d construct, the K_s can be used to describe COPC partitioning between the solid and aqueous phases, especially within disposal sites and within waste forms. K_s values are used for conditions where the concentrations of the COPCs exceed the solubility of an assumed solubility-controlling mineral phase. Such conditions

are identified based on a combination of thermodynamic calculations and laboratory microscopy and wet chemistry studies involving COPC of varying concentrations [8].

3. Apparent diffusion coefficient (D_a). The D_a construct is a combination of hydrology and geochemistry as compared to *De*, which does not include geochemistry. It provides a measure of desorption, as opposed to (ad)sorption, which typically is the rate limiting step.

Additional parameters can be obtained to represent the geochemistry of SSW cementitious waste forms, the 2016 SSW data package [9] has further information and discussions.

SPECIFIC SSW STREAMS ADDRESSED IN THE IDF PA

The waste streams for the 2017 IDF PA are summarized in RPP-ENV-58738 [10] and described in detail in the IDF PA inventory data package, RPP-ENV-58562 [2]. Figure 1 shows the different waste streams resulting from operations of the WTP that will be treated and disposed at IDF. SSW is generally classified into two categories: debris waste (defined in Washington Administrative Code section 173-303-040 [11] as waste with a particle size greater than 60 mm) and non-debris waste (waste with a particle size less than or equal to 60 mm).

Non-debris waste includes small particulates that can potentially be blended into the cementitious material (e.g., ion exchange resins, granular activated carbon). Debris includes larger materials (e.g., components, filters, etc.) that will be encapsulated in a cementitious material. The IDF PA team identified a set of key contaminants that are expected to be the primary concern for the PA calculations (Tc-99, I-129, Cs-137, Sr-90, uranium isotopes (and total uranium), chromium, mercury and nitrate). The inventory of contaminants were estimated for each projected SSW and it was established that the most critical contaminants that need to be accounted and investigated are Tc-99 and I-129. Four specific SSW streams were identified to contain considerable amounts of Tc-99 and/or I-129:

Carbon Adsorber Beds

SSW inventory data from the Hanford Tank Waste Operations Simulator (HTWOS) shows that the LAW melter spent adsorber beds and Ag-mordenite (see below) are major contributors of I-129. The carbon adsorber beds are part of the LAW off-gas treatment system and contain activated carbon for mercury and halide (Hg, F) removal as well as I-129 abatement. Carbon adsorber beds are considered non-debris mixed LLW (MLLW), which from a treatment perspective, contain potentially problematic amounts of Hg and I-129. Although treatment may remove some of the hazard, the conservative recommendation for disposition of this waste is to dispose it at IDF. The beds would be transported to a local offsite treatment facility where they would be repackaged into suitable disposal containers with a stabilization grout/cementitious[a] material for RCRA metals and Category 3 radioactive waste containment using a Hanford approved formulation that meets regulatory criteria. The option to consider the carbon adsorber beds as an encapsulated waste form not blended in a cementitious matrix is also addressed.

Ion Exchange Resin

Ion exchange resins and HEPA filters (see next section) are the largest sources of Tc-99 for SSW. After being dewatered at WTP, the IX resins (Category 3 non-debris MLLW) would be transported in High Integrity Containers (HICs) offsite for treatment. At the treatment facility, the resin would be repackaged into suitable disposal containers blended with a stabilization grout for RCRA metals and Category 3 radioactive waste containment using a Hanford approved grout formulation meeting regulatory criteria.

HEPA Filters

The current assumption is that non-woven glass paper (borosilicate microfiber) HEPA filters would be used and they could be either MLLW or LLW debris depending on their location/function within the WTP facility. All HEPA filters (both Category 1 and Category 3) are expected to be sent to an offsite treatment facility in carbon steel 55-gallon drums where they will be compacted into “pucks” at an approximated compaction ratio ranging from 5:1 to 10:1. Multiple pucks would be placed into suitable disposal boxes and macroencapsulated with grout to meet LDR requirements for RCRA constituents. This macroencapsulation process would meet Category 3 stabilization requirements, which exceeds Category 1 requirements.

Ag-Mordenite Cartridges

Silver impregnated adsorbers (e.g., mordenite) are designed to capture I-129 from off gas systems, and thus similar to the carbon adsorbers, can be one of the primary sources of I-129 in the IDF inventory. The Ag-mordenite waste stream is expected to be non-debris MLLW similar to the carbon adsorber beds, and may include problematic concentrations of Hg and I-129. Although treatment may remove some of the hazard, the conservative recommendation at this time is to assume disposal at IDF without removal of COPCs. The Ag-mordenite would be transported to a local offsite treatment facility where they would be repackaged into suitable disposal containers blended with a stabilization grout for RCRA metals and Category 3 radioactive waste containment using a Hanford approved grout formulation that meets regulatory criteria.

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[a] Grout or cementitious material refer to formulations that consist of ordinary Portland cement, blast furnace slag and fly ash. The two terms are used interchangeably in this paper.

ONGOING and PLANNED WORK FOR SSW CEMENTITIOUS WASTE FORM

Previous Work Done

Stabilized SSW cementitious waste forms have been included and analyzed as part of the 2017 IDF PA. However, there has not been a development and testing program in place to collect data on Hanford SSW to be disposed in the IDF. Useful data is available from other programs such as cementitious barriers or grout development for tank closure at Hanford and other sites. Hence, information available from published literature was reviewed, surveyed and compiled in a data package for the 2017 IDF PA [9]. There is a considerable amount of data in this data package hence, statistical analysis or recommendations for the parameter values are listed for the IDF PA use.

Ongoing and Planned Work

1. Gathering of Information from International/Europe Counterparts of SSW

Washington River Protection Solutions, LLC (WRPS) has subcontracted a UK-based company, Different By Design (DBD LLC), to perform information gathering and literature review on counterparts of Hanford SSW in the UK, Europe and even in Asia. Information gathered are waste form performance, stability, grout formulations and specific treatments for the counterparts of the four specific SSW stream mentioned above. The information gathered will be used in the development and testing of Hanford SSW.

2. Development and Testing of Hanford-specific SSW Cementitious Waste Forms

WRPS has subcontracted Savannah River National Laboratory (SRNL) to perform the development of waste form formulations and testing to address performance requirements and waste form characteristics of SSW. This work is consistent with Level 1 activities “Preliminary Waste Form Screening” and “Waste Form Development” contained in the Hanford Site Secondary Waste Roadmap [PNNL, 2009]. Results from this work will be used as part of maintenance to provide site-specific data for the material property values currently being used in the IDF PA. In addition, SSW treatment is intended to provide the following benefits:

i. Provide structural stability
ii. Reduce void space in waste packaging
iii. Compressive strength 150 psi
iv. Compatible with container

v. Maintain waste form temperatures < 160°F
vi. Meet RCRA land disposal requirements
vii. Radiation stability (preservation of compressive strength following exposure)

This list of benefits aligns with the draft waste acceptance criteria applicable to the waste streams being considered (Draft document is not publicly available).

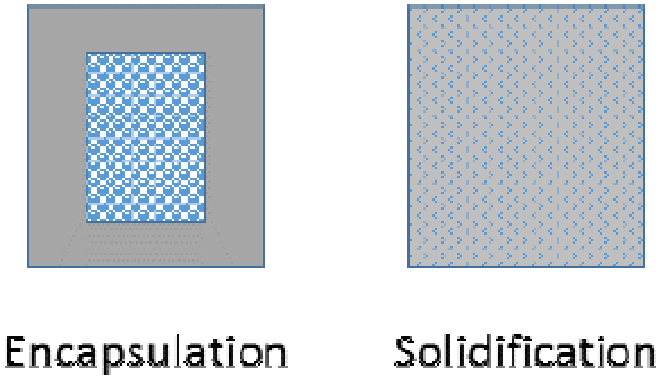
Waste form testing to support the IDF PA will be conducted in three stages, with each stage building on results from the previous set of tests. The stages are:

1. Evaluate grout/cementitious formulation options for waste forms

2. Test waste forms – physical and hydraulic performance

3. Test Waste Forms – chemical performance

SSW stabilization have two configurations; solidification and encapsulation. Solidification may be applied for non-debris wastes of particulate size that would be mixed into a grout or cementitious media. In this case the properties of the waste form will represent the integrated mixture of waste and solidification media (e.g., ion exchange resins blended in a cementitious matrix). Encapsulation can be applied for waste (may be debris or non-debris) with major voids filled by a clean encapsulating media and a specified thickness of encapsulating media completely surrounding the waste. The necessary thickness of the encapsulating media around the waste would be determined in an iterative manner based on results of the PA. Illustration of encapsulation and solidification is shown in Figure 2.



Encapsulation Solidification

Figure 2. Illustration of the two configurations of SSW stabilization

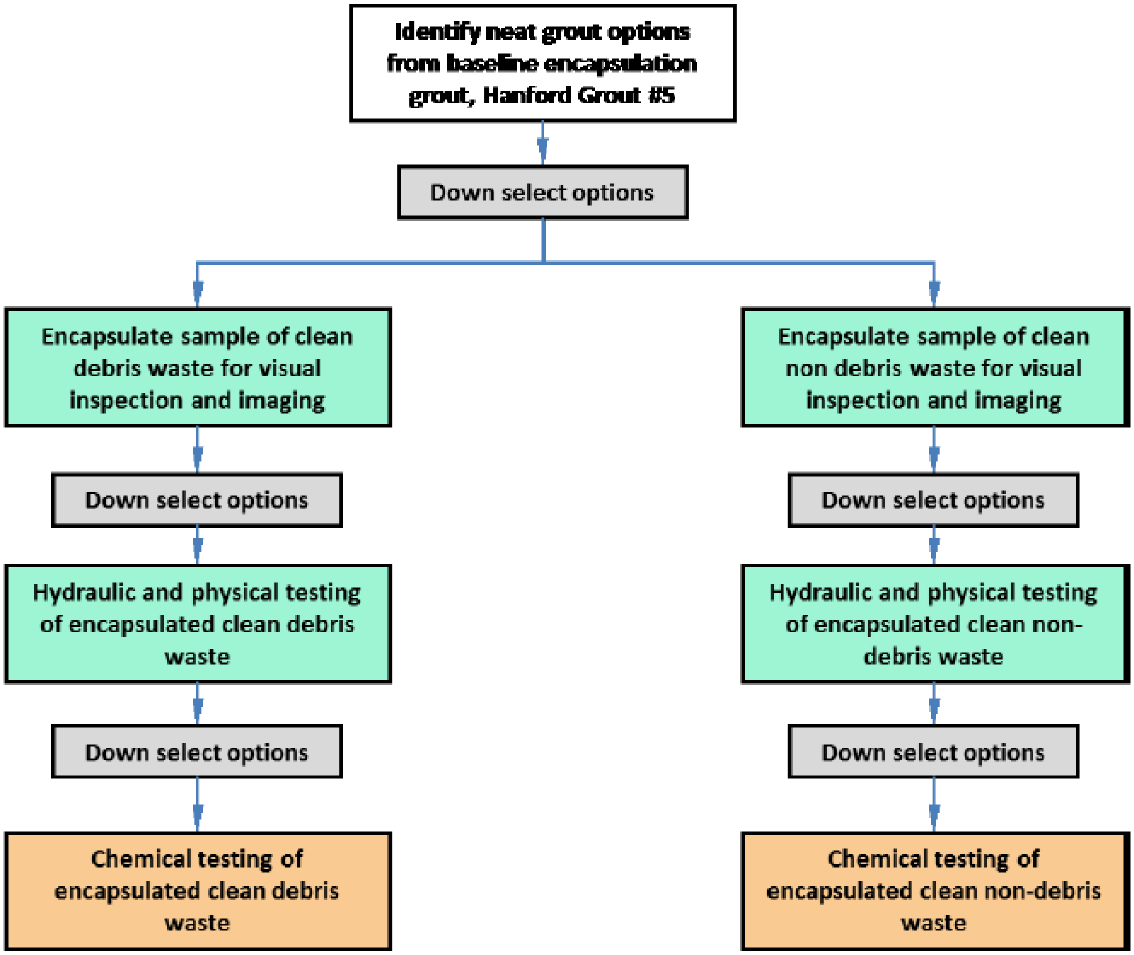


Figure 3. Strategy for SSW Cementitious Waste Form Development and Testing.

Figure 3 illustrates the strategy for developing SSW form. The range of grout options to be initially tested will be down selected to a few options to be used in subsequent testing with simulant SSW. Specific waste forms will be tested in two stages, with the first stage being Physical and Hydraulic Performance and the second stage being Chemical Performance to obtain the key parameters listed above. Some waste form formulations may be eliminated for chemical performance testing depending on results from physical and hydraulic testing. The initial emphasis of waste form testing is planned to address ion exchange resins because initial IDF PA analysis showed considerable inventory of some COPCs. The next SSW to be considered for waste form development will depend on the results of the final 2017 IDF PA, the SSW that shows significant amount of COPCs will be given priority.

Results from physical and hydraulic performance testing will be used to identify waste forms to undergo additional testing to determine transport parameters to support the IDF PA. The waste forms consist of three components, debris or non-debris waste, grout, and pore water. A series of sorption tests are being considered to determine the K_d for selected grout and the K_d for the waste. The first sorption tests could involve two components, either loaded waste and water or water containing COPC and grout. Following these tests, all three components will be incorporated into a waste form to determine leaching properties. This approach will provide information necessary to optimize the waste form and input data for the PA model. In addition, the diffusion coefficient of selected grouts will be determined.

The debris and non-debris wastes for IDF is expected to include cation, anion, radioactive and hazardous COPCs. These COPCs exhibit a wide range of behavior depending on the chemical environment they are in. Tests will be conducted under a range of scenarios to simulate different geochemical conditions that maybe encountered during the expected lifetime of the IDF, (e.g. pH and redox). This approach to leach testing of using multiple chemical conditions is similar to the approach recommended in the USEPA LEAF methods. The LEAF approach “ is to challenge the waste material to a broad range of experimental conditions known to affect constituent leaching, with the intent to derive characteristic or fundamental intrinsic parameters that control leaching” [12].

Performance testing of ion exchange resins is scheduled for FY17 under Test Specification for the Low-Activity Waste Pretreatment System Full-Scale Ion Exchange Column Test and Engineering-Scale Integrated Test. Spent material from the full-scale IX resin testing would be useful in preparation of waste forms in this study as it would closely resemble spent IX resin that is expected to be disposed of as SSW in the IDF. An effort has been made to obtain spent IX resin for use in SSW work. SSW form development will begin in FY17, but testing is likely to extend into FY18.

CONCLUSIONS

Solid secondary waste form development and testing is still on its initial implementation and no previous data is available for the Hanford-specific SSW. By working with the IDF PA team, national laboratories and international company, information had been gathered to bring about a systematic plan. From the IDF PA initial analysis, specific SSW stream were identified. Information from other sites relevant to the development of SSW are gathered from the US and international locations, these information will be utilized to devise formulations and testing methodologies. Finally, national laboratories are delegated to perform the development and testing, utilizing their expert personnel and state of the art equipment.

The data to be acquired in the SSW development and testing project will be utilized for the IDF PA, and will also provide the appropriate stabilization configurations for the disposal of the SSW.

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