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The Savannah River Site Accelerated Clean-Up Mission: Sludge Retrieval and Immobilization

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Abstract

Approximately 130 million liters of sludge/supernate high-level radioactive waste (HLW) is currently stored in underground carbon steel tanks at the Savannah River Site (SRS) in Aiken, South Carolina. The Defense Waste Processing Facility (DWPF) began immobilizing these wastes in borosilicate glass in 1996. Currently, the radioactive glass is being produced as a "sludge-only" composition by combining washed high-level sludge with glass frit and melting. The glass is poured into stainless steel canisters for eventual disposal in a permanent geological repository.

Recent directives from the U.S. Department of Energy have been focused on accelerated clean-up missions for the various sites around the DOE complex. The Savannah River Site has developed a program to meet this new directive. With respect to the DWPF, a "vision case" has been developed which reduces the overall immobilization campaign by several years. This reduction is based on several parameters; two of which are increases in melt rate and waste loading. To support this incentive, the Savannah River Technology Center is focusing on increasing waste loading and/or improving melt rates via strategic glass formulation, changes in acid addition strategies, and reassessing process control models to challenge their extreme conservatism. Glass formulation activities have recently shifted from the strategy of using "generic" frits developed to process a number of different sludge batches, to the concept of developing a specific frit for each sludge batch in order to obtain higher waste loadings and/or improved melt rates. Increases in waste loading have also been realized by the development of a new liquidus temperature model – a parameter which has typically limited waste loadings in previous processing. Coupling the new liquidus model with strategic glass formulations will result in an increase in overall waste throughput. Assessments are also being made on the potential to minimize the number of sludge washing steps to minimize the volume of wash water (ultimately treated by the high-level waste evaporators) without compromising processability, product quality, or waste throughput goals.

In order for DWPF to process the waste faster, retrieval of sludge from the waste tanks must be accelerated. Acceleration of sludge retrieval will focus on developing technologies to reduce the time and cost to retrieve the bulk of the sludge waste from tanks, retrieve residual heels, and retrieve waste from the annulus of tanks that have leaked. Much of the cost for the retrieval infrastructure can be attributed to the elaborate support structure that must be built, spanning the tank top, to support the four mixing pumps and the transfer pump. Previous work has shown through testing of a high capacity Advanced Design Mixer Pump (ADMP) that the number of mixer pumps necessary to mobilize the waste could be reduced by half.

Introduction

The production of nuclear materials at the Savannah River Site (SRS) in Aiken, South Carolina has produced alkaline high level liquid waste that is stored in underground carbon steel waste tanks. Approximately 130 million liters of sludge/supernate high-level radioactive waste (HLW) is currently being stored. The Defense Waste Processing Facility (DWPF) began immobilizing these wastes in borosilicate glass in 1996. Currently, the radioactive glass is being produced as a "sludge-only" composition by combining high-level sludge with glass frit and melting. The glass is poured into stainless steel canisters for eventual disposed in a permanent geological repository. Prior to the receipt of sludge waste in the DWPF it is washed to remove soluble salts.

Recent directives from the U.S. Department of Energy (DOE) have been focused on accelerated clean-up missions for the various sites around the DOE complex. The Savannah River Site has developed a program to meet this new directive.(1) This new plan focuses on risk reduction, which will reduce the cleanup costs and the time it takes to complete the effort. The Expedited High Level Waste (HLW) Processing initiative is one aspect of the overall plan for SRS. This effort will produce approximately 1000 fewer canisters in the DWPF and complete high level waste processing 10 years earlier than scheduled and save an estimated $6.6 billion. This initiative classifies the HLW into 4 components and tailors the treatment to each of those components. The components are:

- Sludge (which contains the majority of the long lived radionuclides)
- Low curie salt
- Low curie salt with higher actinide content
- High curie salt with higher actinide content

The purpose of this segregation is to allow the least costly method of treatment to be used on each stream. The sludge along with the actinides and cesium from the salt processing will be immobilized at DWPF.

Process Overview

The SRS High Level Waste System integrates a series of facilities that accept, treat, and immobilize the waste (Figure 1). There are four types of underground waste storage tanks at SRS. The Type I and type II tanks are categorized as "high risk" because they do not meet current secondary containment and leak detection standards. Type III tanks meet current requirements while Type IV are closer than Types I and II but do not completely meet requirements. The SRS waste is broadly characterized as "sludge waste" or "salt waste". The sludge consists of metal hydroxides, carbonates, and oxides. Sludge generally contains the radionuclides plutonium, uranium and strontium and is approximately 8% of the waste volume and 55% of the waste radioactivity. The salt waste is soluble and is supernate or saltcake (previously dissolved salts that have now crystallized out of solution). Incoming liquid waste to the Tank Farm is evaporated to make better use of tank space.

Waste in the tanks is mobilized using long shaft mixer pumps supported by trussed bridgework. Sludge is mobilized using four 45 ft long 150 hp mixer pumps and transferred using a long shaft transfer pump. Installation of the infrastructure is time consuming and expensive taking as much as approximately 4 years to prepare and costing

approximately $20M for each tank. After the waste is mobilized and removed, the tank is evaluated to determine how much waste is remaining and what actions yet need to be taken to prepare the tank for closure. Many tanks contain internal obstructions that inhibit mixing in the tanks and hold up slurry solids. Adding water and again mixing to mobilize the waste may retrieve additional waste. Waste that is caked together may not mobilize and will be left in the tank as heels. No standard procedures exist to remove heels. To date nine tanks have leaked and contain radioactive waste in the annulus. Techniques to retrieve waste from the annulus have been considered but none have been deployed.

Figure 1. High Level Waste System

Prior to transfer to the DWPF, the sludge portion of the high level waste is washed to remove soluble salts. Several wash cycles are usually performed to produce a sludge that is acceptable for DWPF processing and to minimize the number of canisters that will be produced. Two Type III tanks (Tanks 40 and 51) are used as the feed tanks to DWPF. A wash cycle consists of the addition of inhibited water, agitation, settling and decanting of the wash water. Typically the wash water is sent to the evaporator system in order to maintain adequate tank space.

The salt waste will be processed using an integrated approached. The low curie salt waste will be treated and disposed of at the Saltstone facility (grout waste form). Low curie/high actinide salt waste will be processed to remove the actinides and sent to DWPF and the low curie material to Saltstone. High curie salt waste will be processed to remove the cesium using Caustic Side Solvent Extraction. The cesium stream will be fed to DWPF and the low level fraction to Saltstone. The salt solution is not yet available for vitrification, thus, DWPF is currently operating with a sludge-only flowsheet.

The sludge is transferred into the DWPF Sludge Receipt and Adjustment Tank (SRAT) (11,000 gallon tank) and then neutralized with nitric acid. Formic acid is also added to the SRAT to control redox and reduce mercury. The excess formic acid reduces the mercuric oxide in the sludge to elemental mercury. The elemental mercury is then steam stripped

from the SRAT into a holding tank from which it is later pumped and decontaminated. The SRAT product is transferred to the Slurry Mix Evaporator (SME) where a borosilicate glass frit is added and the slurry is concentrated to produce melter feed. The SME is the hold point in the process where feed acceptability is determined as part of the Glass Product Control Program.(2) Compositional analyses of the SME samples are used to determine the acceptability of the melter feed. The acceptability of the melter feed is determined using glass property models and statistical algorithms which take into account analytical uncertainty to ensure the glass product will be acceptable and that processing constraints (such as viscosity and liquidus) are met. These models define an "operational window" within which the melter feed composition must reside to be acceptable.

Once the melter feed material in the SME is determined to be acceptable, it is transferred to the Melter Feed Tank (MFT) and then fed to the joule heated melter. One SME/MFT batch produces approximately 7 canisters of glass. The DWPF melter has four resistance heaters in the vapor space and two pairs of electrodes in the molten glass. The feed slurry is introduced from the top of the melter and forms a crust, or cold cap, on the surface of the melt pool as the water is evaporated and removed via the off-gas system. The cold cap melts from the bottom and forms the borosilicate glass matrix. The nominal glass melt pool temperature is 1150ºC.

The glass is removed from the melter near the bottom through a side riser and pour spout. A vacuum is drawn on the pour spout to pour the glass. After a canister is filled, a temporary seal is installed to prevent free liquid from entering the canister during the decontamination process. Decontamination of the canister surface consists of blasting an air-injected frit slurry against the canister. The frit slurry from the decontamination process is used in the next SME batch as part of the required frit addition. After smear testing to ensure no radioactive surface contamination is present, the canister is then welded closed and transferred to an interim storage building.

Production Status

Processing of the first two radioactive sludge batches (approximately 950,000 gallons of sludge) has been completed in the DWPF. The third sludge batch is currently being processed. Production was suspended in November 2002 in order to replace the first DWPF melter after 6 years of radioactive operation. Glass production is expected to resume in April 2003. Table I summarizes the processing of the initial sludge batches.(3)

Table I. Summary of Sludge Batch Processing

Sludge Batch	Dates Processed	Volume of Sludge (kgal)*	Number of Canisters	Average Waste Loading (wt% waste oxides)^	Average Glass Pour Rate (lbs/hr)
1A	3/96 – 9/98	351	495	25	146
1B	10/98 -11/01	460	726	25	161
2	12/01 -	600 (projected)	470 (projected)	27.5	159

*Total solids is typically 16-19 wt%.

^Waste loading is the percentage of waste oxides in the glass

As with any complex technical facility, difficulties were encountered during the initial years of operation. The composition of the sludge and melter feed material remained fairly constant during the processing of sludge batch 1A. Sludge batch 1B contained approximately eight times more Sr-90 than the first sludge batch. Some processing difficulties were encountered with sludge batch 1B that weren't seen with sludge batch 1A. Sludge batch 1B appeared to have different rheological properties (e.g. higher yield stress). There were have several cases where line pluggage occurred. Also, the region of melter feed acceptability, as defined by the glass product and processing models, was smaller for sludge batch 1B, thus, making it more difficult to select the correct amount of frit to add to the SME.

Sludge batch 2 processing was initiated in December 2001. This sludge batch has a significantly higher uranium content that the previous batches and the batch has presented unique processing problems. This sludge batch appears to retain air causing pump trips and heat transfer problems during chemical processing. Also, sustaining the average melt rate has been difficult. A research and development program has been developed to understand these issues. Additionally, an improved frit composition (Frit 320) has been developed for sludge batch 2.(4) Current plans are to implement this new frit to improve melt rate (up to approximately 175 lbs/hr) and increase waste loading (up to approximately 35 wt%) following the melter outage. The development of this frit was the initiation of the effort to accelerate immobilization efforts as will be discussed later.

Accelerated Cleanup

As previously discussed, SRS has developed an accelerated cleanup plan in response to recent directives from the DOE.(1) This plan puts the emphasis on risk reduction in order to reduce processing time and reduce costs. SRS is adopting a Safe Mission Essential concept and applying it to its programs and activities. This will appropriately align requirements with work to be completed. The Expedited HLW Processing initiative allows for:

- Faster reduction of waste inventory
- Accelerated risk reduction for waste removal from high risk tanks
- Ability to meet Federal Facility Agreement commitments
- Increased tank farm flexibility (ability to handle emergent issues)
- A graded approach to salt processing will be used by utilizing multiple approaches

A key aspect of this initiative is utilizing a graded approach to salt processing. This strategy processes low curie salt at Saltstone, creates an Actinide Removal Process to remove actinides from the low curie/high actinide stream, uses a small scale Caustic Side Solvent Extraction to process high curie salt, and then tailors the high curie salt waste processing capability depending on success of early low curie salt disposal.

Sludge processing is currently limited by several aspects:

1. Time and cost of bulk sludge removal,
2. Time and water required for sludge washing,
3. Melter throughput and waste loading in DWPF.

The preparation of a sludge batch for acceptance by DWPF is a costly and time

consuming process.

Accelerating sludge preparation is necessary to meet the acceleration mission. This effort involves incorporating newly developed waste removal methods for sludge removal. Expediting sludge processing through DWPF involves maximizing waste throughput by balancing an increase in waste loading in the glass and melt rate of the feed through the DWPF melter. This necessitates improved glass property models and glass frits tailored specifically for the sludge waste coming to DWPF.

Sludge Retrieval

Much of the cost for the retrieval infrastructure can be attributed to the elaborate support structure that must be built, spanning the tank top, to support the four mixing pumps and the transfer pump. Previous work has shown through testing of a high capacity Advanced Design Mixer Pump (ADMP) that the number of mixer pumps necessary to mobilize the waste could be reduced by half. The ADMP, however, still requires that a substantial superstructure be built over the tank top. Further work on new waste retrieval system that would be less costly and more rapidly deployed will be necessary to meet the objectives of the acceleration mission.

Within the last two years sludge transfers were made from two high risk tanks. Sludge was suspended and transferred from Tank 8. This sludge is now part of Sludge Batch 2, the current batch being fed to DWPF. Innovative removal techniques, such as the use of centrifugal slurry pumps submerged in the tank with waste supernate rather than fresh water, were used to remove sludge from Tank 19 that minimized the volume of fresh water added to the tank. Tank 19 is now ready for closure.

Increasing Waste Throughput

Maximizing the amount of waste per canister and maximizing the rate at which the waste/frit melts are desired objectives. However, in some cases these objectives can be competing. Thus, the balance between melt rate and waste loading will be carefully considered. For recent frit development efforts for Sludge Batch 2 every effort was made to indirectly develop frit compositions in terms of melt rate, however, the concern is that targeting the maximum waste loading allowed by current model predictions may not necessarily lead to optimum melt rate or waste throughput. In fact, it was shown that for the Frit 320 based system, increased waste loading can have a negative impact on melt rate above some critical waste loading value.(5) The concept that reduced melt rates at higher waste loading is unacceptable should be tempered with an evaluation of the total waste throughput. More specifically, during an assessment of the impacts of waste loading on melt rate, decisions on frit selection or targeted waste loading should not be made solely on the relative melt rate. The decision should consider the total sludge throughput per unit time. Note that currently there are no models that can be used a priori to gain insight into the melt rate of various systems. Assessment for melt rate have been made using an integrated testing methodology that spans from laboratory-scale melts to slurry-fed, mini-melter demonstrations.

There are several parameters to be considered when maximizing waste throughput. Recent efforts on an improved liquidus correlation have led to increased waste loadings for future sludge batches. While a methodology for frit development has been defined that will allow maximum waste throughput by balancing waste loading and melt rate.

Improved Liquidus Correlation

DWPF must balance several glass property constraints to determine an acceptable glass composition region in which to operate. These glass properties include durability, liquidus, homogeneity, and viscosity. The liquidus temperature constraint is needed to prevent bulk crystallization from forming in the melter and blocking the riser or impacting the joule heating. The liquidus temperature is the minimum temperature at which the glass melt and primary crystalline phase are in equilibrium. The liquidus temperature constraint has traditionally limited the amount of waste that can be put in each canister (i.e. waste loading - at SRS, waste loading is defined as the HLW oxide fraction of the final glass).

An effort was initiated in 1998 to reduce the uncertainly in the initial liquidus correlation by producing an additional statistically designed set of glasses for use in the development of an improved model. This new data set, along with some other additional glass data, was used to develop an improved semi-empirical liquidus correlation.(6) The correlation has been implemented in DWPF and will be utilized in the Spring of 2003. This new model has been shown to yield higher waste loadings for current and projected sludge batches. These waste loading improvements are shown in Table II. The waste loading improvements were calculated using the sludge compositions and three existing frit compositions. The table demonstrates the improved waste loading but also shows that the frit composition can also have a significant impact on waste loading.

Table II. Projected Waste Loadings (wt% waste oxides) for New and Old Liquidus Model and Various Frit Compositions

	Frit 165	Frit 165	Frit 200	Frit 200	Frit 320	Frit 320
Sludge Batch	Old Model	New Model	Old Model	New Model	Old Model	New Model
1A	25.8	35.2	26.4	30.7	26.9	37.5
1B	28.4	37.3	29.0	32.7	29.6	39.7
2	29.9	36.3	30.5	31.6	31.1	38.6
3 (projected)	28.4	35.6	29.0	30.9	29.5	37.9
4 (projected)	30.2	38.4	30.8	33.6	31.4	40.8

Frit Development

The Savannah River Technology Center is currently developing a frit composition for use with Sludge Batch 3. A paper study has recently been completed that evaluates new frit compositions (7). This effort was focused on Sludge Batch 3 but provides a unique, but technically sound methodology to guide current and future frit development activities. Several key criteria, listed below, have been developed that will provide the technical basis for selecting a frit for a particular sludge batch. The criteria include:

1. Maximizing the projected operating window size over the anticipated sludge

composition region - This provides operational flexibility in targeting waste loadings to meet processing goals. This is strictly an ease-of-processing goal targeted to provide as large of a compositional operating window and as much flexibility as possible.

2. Providing a frit that is robust or insensitive to anticipated sludge composition variation - A viable frit should not only be able to process the nominal composition anticipated but should also be able to process (i.e., be robust or tolerant to) realistic variations of that composition while still maintaining adequate processing and product characteristics.
3. Improving or maintaining high waste loadings – Adjusting the frit composition may allow for increased waste loadings in the same manner as improving the liquidus temperature model.
4. Improving or maintaining high melt rates - Currently, no models exist that can be used a priori to gain insight into the melt rate of various systems. Therefore, this assessment will be made by using a testing methodology that was previously shown to be effective for SB2.(8).
5. Providing a composition that can can be commercially manufactured as a prefabricated glass or frit - DWPF's current program uses a frit rather than batch chemicals. However, this criterion may be modified in the future as the potential benefits of batch chemicals are evaluated.

Given that the five key criteria can be competing, the basis for not only developing but ultimately selecting a frit is complex. The selection process should not be made based on a single criterion but a collection of criteria that provide insight into the economics of processing a sludge batch. A balanced approach should be utilized in both the development and selection of a frit.

The model-based assessments performed for Sludge Batch 3 indicate that judicious selection of the frit can yield processable and durable products at attractive waste loadings for all washing scenarios. The washing scenarios or strategies vary from a few wash cycles to a large number (up to 12) of wash cycles. The results provide support for the concept of developing specific frits for specific sludges to optimize operating windows and waste throughput. Given this, an aggressive washing strategy (large number of washes) may not be required to assure processability or product quality as long as alternative frits are considered (assuming there are no other glass- or process-related restrictions such as anion solubility, H_2 generation, redox control, or rheological control issues).

An integrated strategy has been developed that will lower the risk of introducing a feed into DWPF that although on paper is very attractive (in terms of waste loading) results in a very difficult feed to process (in terms of melt rate). The strategy includes assessments of the five key criteria as noted above. More specifically, model-based assessments provide insight into the operational window sizes as a function of frit and sludge compositions over a waste loading interval of interest. These model-based assessments are supplemented by assessments of melt rate and frittability to provide a technical basis for establishing a baseline flowsheet. In fact, this strategy should provide the basis for developing a decision matrix in which optimum waste throughput could be targeted.

Two stages are used in the model-based assessments to assess various frit/sludge combinations in terms of the projected operational windows and the tolerance to compositional sludge variation: the Nominal Stage and the Variation Stage. The Nominal

Stage utilized a nominal composition representing a potential washing scenario as outlined in Figure 2. In general, candidate frit compositions are assessed with respect to their ability to provide a relatively large operational window based solely on a specific nominal composition – no sludge variation was accounted for in this phase. Assessments are made using predictions from models currently implemented in DWPF over the waste loading interval of interest (25 – 60 wt%). The property predictions assessed included those for liquidus temperature, viscosity, durability, and homogeneity. An increased risk would result with respect to processability or product quality if a decision were based solely on this assessment without accounting for compositional variation. The intent or focus of the Variation Stage assessment is to gain insight into the robustness of candidate frits with respect to compositional variation. Figure 3 provides an overview of the Variation Stage assessment. Again, the major difference between the two stages is the fact that the Variation Stage builds into its assessment an anticipated variation of ± 10% around the nominal composition.

Application of this model-based paper study assessment from the first phase of the frit development for Sludge Batch 3(7) identified the key conclusions summarized in Table 3. The significance or merit of each conclusion is also summarized in Table III.

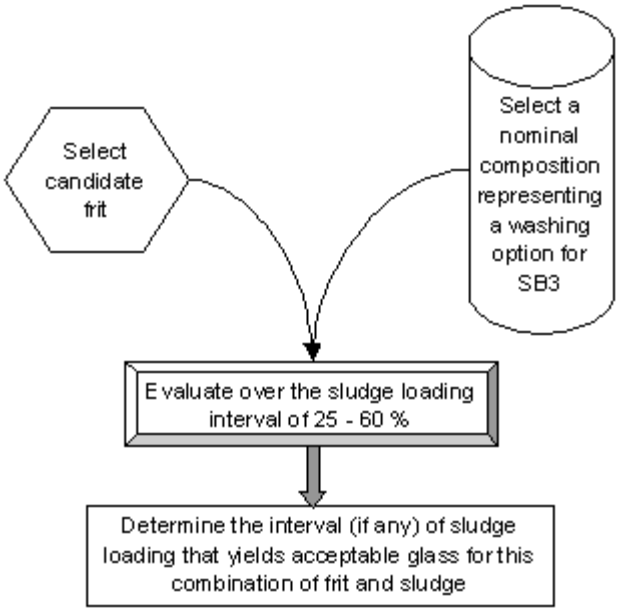
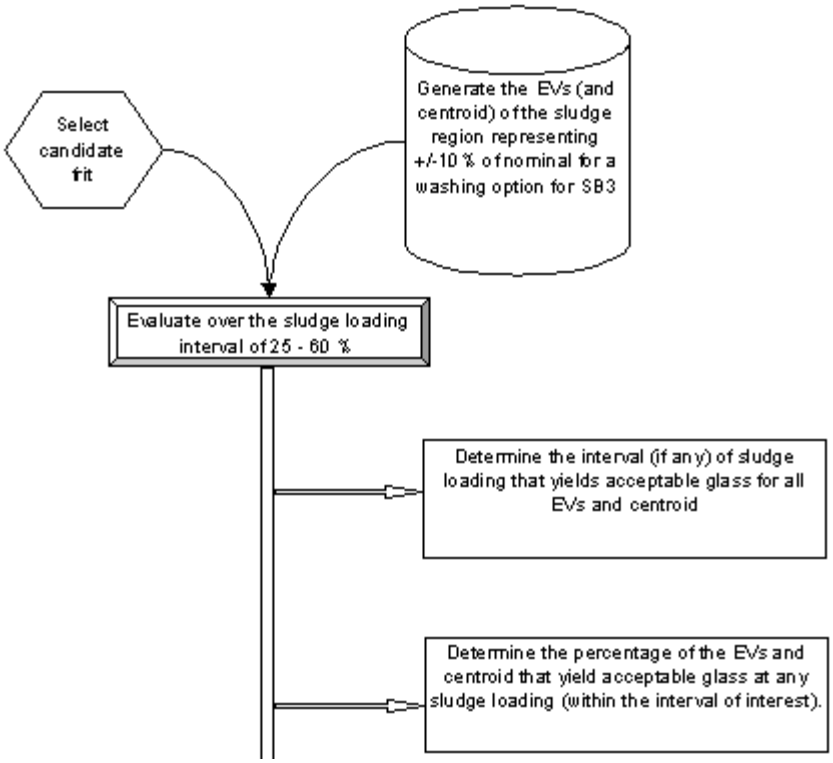


Figure 2. Evaluation Strategy Utilized in the Nominal Stage Assessment of Sludge Compositions

Table III. Major Conclusions and Merits from Frit Develoment Efforts

Major Conclusions	Merit
Compositional frit adjustments can be made to compensate for various waste stream compositions resulting from the washing strategy pursued. A cost-benefit analysis should be performed to fully understand the advantages and disadvantages of selecting a targeted washing scenario.	The ability to compositionally compensate for the degree of washing via frit development efforts provides an incentive to further evaluate the potential to integrate a "low washed sludge" into the overall flowsheet. Less washing of the sludge has advantages in terms of significant time and labor savings in the pretreatment/retrieval operation. A reduced washing campaign should also result in less water being sent to the

	evaporators.
The concept of developing alternative frits for specific sludges (in an attempt to maximize throughput and/or operational flexibility) was observed numerous times during these assessments.	Modifying frit compositions for each sludge batch will allow waste throughput to be maximized for each sludge batch.
Historically, most of the previous sludge batches processed through DWPF have been liquidus temperature limited (in terms of the maximum waste loading achievable). An interesting outcome in this assessment was the fact that frit development efforts could take advantage of the high alkali content of the sludge, driving or forcing the low washed sludges to be durability limited.	Working with a durability limited system may be beneficial from a waste loading and melt rate perspective since currently the durability property has the largest amount of flexibility until the limit is met.



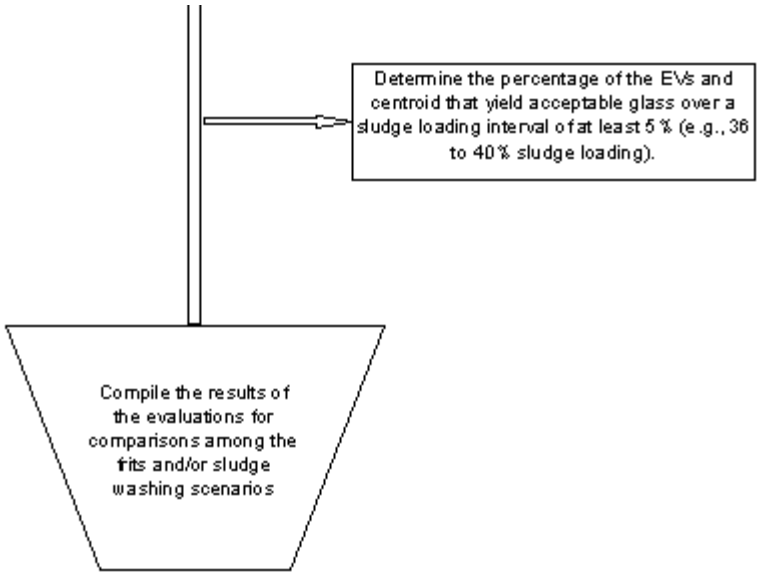


Figure 3. Evaluation Strategy Utilized in the Variation Stage Assessment of Sludge Compositions

Other Property Model Improvements

Another subset of accelerated HLW immobilization is evaluation of other glass property models (besides the liquidus model) and/or the implementation strategy for these models. For example, earlier frit development efforts (8) showed that frits could be developed with high melt rates that yield glasses with acceptable durability but the current durability/composition model did not adequately predict the durability response. This led to the conclusion that these frits could not be utilized since the glass durability was not adequately predicted. Efforts to evaluate and revise the strategy for demonstrating an acceptable glass, including the glass durability model itself, are being initiated.

Conclusions

SRS is initiating efforts to implement accelerated cleanup plans for completion of the high level waste project. Innovative waste removal methods are being investigated. In the DWPF, waste throughput is being maximized. Maximizing waste throughput requires a systems approach to balance waste loading in the glass product and melt rate through the melter. These factors are sometimes competing. Thus, creative approaches to modify the glass compositions are necessary. The glass composition can be varied by making changes to the frit composition for each individual batch of sludge. An integrated methodology has been developed and is being utilized to develop frits for specific sludge batches. The methodology includes assessments of (a) projected operational windows using current model predictions, (b) melt rate, and (c) ability to manufacture a frit. This integrated strategy lowers the risks of introducing a feed into DWPF that although on paper is very attractive (in terms of waste loading) results in a very difficult feed to process (in terms of melt rate). This strategy provides the basis for developing a decision matrix in which optimum waste throughput is targeted.

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