Reevaluation of Vitrified High-Level Waste Form Criteria for Potential Cost Savings at the Defense Waste Processing Facility - 13598

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ABSTRACT

At the Savannah River Site (SRS) the Defense Waste Processing Facility (DWPF) has been immobilizing SRS's radioactive high level waste (HLW) sludge into a durable borosilicate glass since 1996. Currently the DWPF has poured over 3,500 canisters, all of which are compliant with the U. S. Department of Energy's (DOE) Waste Acceptance Product Specifications for Vitrified High-Level Waste Forms (WAPS) and therefore ready to be shipped to a federal geologic repository for permanent disposal. Due to DOE petitioning to withdraw the Yucca Mountain License Application (LA) from the Nuclear Regulatory Commission (NRC) in 2010 and thus no clear disposal path for SRS canistered waste forms, there are opportunities for cost savings with future canister production at DWPF and other DOE producer sites by reevaluating high-level waste form requirements and compliance strategies and reducing/eliminating those that will not negatively impact the quality of the canistered waste form.

INTRODUCTION

The DWPF HLW vitrification program was defined to meet the requirements of the DOE WAPS [1] and the DOE Waste Acceptance System Requirements Document (WASRD) [2]. These specifications/requirements were developed assuming Yucca Mountain was the final repository for disposition of HLW. Currently, the DOE has petitioned to withdraw the application from the NRC for HLW disposition and is pursuing alternatives. A "Blue Ribbon" panel of experts was convened to evaluate alternative approaches for disposition. The panel has provided recommendations to DOE based on their review but no definitive plans have been announced by DOE.

In light of these potential changes, the existing WAPS and WASRD requirements should be reviewed to determine which of the requirements are repository driven. For those that are repository driven, a technical review of applicability to the future disposal site should be performed once the alternate repository is selected. Testing may also be required to support the change in repository location. An example of this might be a change in the environmental conditions for disposal and the associated performance testing. For those criteria that are not repository driven, a technical review should be performed to determine whether the criteria are still applicable given the roughly 17 years of radioactive operation at SRS and experience at the West Valley Demonstration Project (WVDP). The technical basis could then be provided to support elimination of the non-relevant criteria. An example might be evaluating whether a different canister material could be used given the leach resistance of the glass and the fact that credit is not taken for the canister in the repository safety analyses.

The regulations and repository waste form requirements drive the cost of compliance up for HLW vitrification, which in turn has an associated impact on treatment schedule and costs. Reductions in programmatic costs can be obtained by changing the DOE requirement documents (WAPS/WASRD) and/or changing the compliance strategy documents at DWPF by taking advantage of the 17 years of production data. These Producer-generated documents include the DWPF Waste Form Compliance Plan (WCP) [3], which describes the compliance strategies and the methods/programs to demonstrate compliance, and the DWPF Waste Form Qualification Report (WQR) [4], which documents the technical bases for these compliance strategies.

DISCUSSION

Conservatism with Glass Models

A primary constraint or acceptance limit for meeting current durability requirements for HLW glass is related to the benchmark Environmental Assessment (EA) glass. More specifically, the durability response of the HLW glass in question as defined by the Product Consistency Test (PCT) must be better than the PCT response of the EA glass with the equivalent of two standard deviations confidence. The accepted boron release is 16.70 g/L for the EA glass. Historically, HLW glasses produced at the DWPF have been an order of magnitude better than the EA glass release with normalized boron releases in the range of 1 g/L. Therefore, a gap exists between where glasses are currently being formulated and their durability response relative to the EA benchmark. The data suggest that investigations should be made to determine the potential positive impacts on mission life reduction if DWPF could target glass compositions that are less durable but still meet the current durability requirements or potentially requirements that may change as a result of repository changes. Some examples might be changes to the Tank Farm operations (e.g., sludge batch washing) or changes in waste loading or processing rates. Realization of any of these options would still require production of an acceptable glass (i.e., melter processing constraints including liquidus temperature and viscosity would also have to be met) but would take advantage of the significant conservatism in durability that currently exists.

Another approach that could be taken without reducing the conservatism that currently exists is to take advantage of the large amount of glass data that has been generated since the start-up of DWPF. This data could be folded into the current durability model to update the model coefficients. It may also lead to the development of an alternative model that more effectively predicts the linear response seen thus far for DWPF. Moreover, the data to be added to the model should more adequately cover the composition region anticipated for future DWPF batches due to the numerous process changes that have occurred since DWPF start-up and the original development of the durability model. Once this data is fitted, the data gaps for out-year processing should more easily be identified. Ultimately, it is the application of the process control models and their associated constraints that will limit projected (and actual) waste loadings and restrict HLW system planning with respect to glass formulations that could be processed through DWPF.

Re-examination of the Sludge Batch Qualification Process

When the initial HLW qualification programs were written for DWPF and WVDP, U.S. operating experience with vitrification of radioactive glass at large scales did not exist, nor was it extensive internationally. Therefore, conservatism was written into the requirements as well as in the associated compliance plans from the operating sites. The DWPF Glass Product Control Program (GPCP) was developed early in the DWPF waste acceptance program and was based on the operating philosophy that controlling the composition of feed to the melter will ensure that an acceptable glass product will be made. Figure 1 displays the key elements of the GPCP.

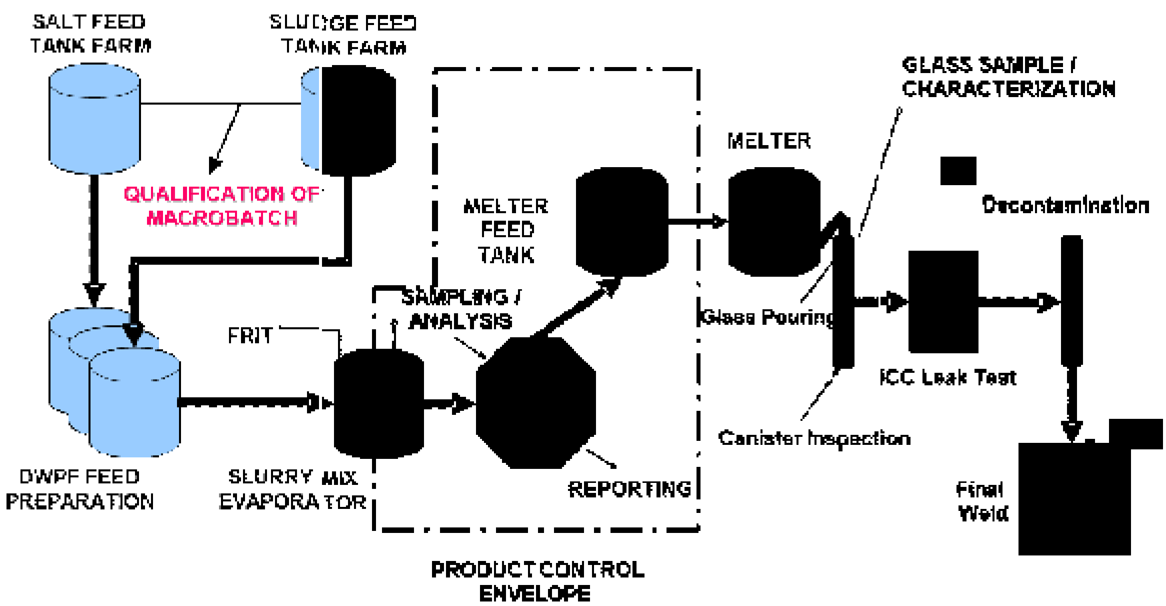

Fig. 1. Schematic providing overview of the DWPF GPCP.

For each sludge batch to be processed, qualification work has been or will be performed as part of the GPCP to demonstrate that the sludge batch (or macrobatch) can be processed at the DWPF and make a compliant glass product. The current compliance strategy for the sludge batch qualification task includes:

- characterization of the chemical and radionuclide constituents of the sludge prepared in the Tank Farm,
- demonstration of the DWPF Sludge Receipt and Adjustment Tank (SRAT) and Slurry Mix Evaporator (SME) process with the qualification sample,
- fabrication and testing of the glass made from the qualification sample SME product, and
- validation of the DWPF durability model over the anticipated glass composition range for the batch (i.e., variability study).

The other elements of the DWPF GPCP shown in Figure 1 include sampling and analysis of the SME during DWPF processing, prediction and targeting of the SME glass composition via the process control algorithm Process Composition Control System (PCCS), verification and reporting of glass composition before transferring to the Melter Feed Tank (MFT), characterization and reporting of the actual radioactive constituents in the as-processed sludge from the DWPF Feed Tank (i.e., WAPS sample), and characterization and durability testing of DWPF glass pour stream samples.

The characterization portion of the sludge batch qualification process has some areas for potential improvement. At this point, it seems unlikely that reduction of the chemical constituent analyses would be possible because of the potential impact on the glass formulation or solubility limits. However, the required analyses, as well as the reporting requirements for out-year projections, for the radionuclides should be reviewed. This should be undertaken in light of the 17 years of production, the potential changes in the repository, and the known radionuclide constituents in HLW. Reduction in the number of radionuclides that are measured versus estimated from other components or known history could save production costs from analyses and from reporting. This would apply to both the qualification sample and the WAPS sample, which provides data used for reporting radionuclide inventory in the DWPF Production Records for the final canistered waste forms. For past sludge batches processed at DWPF, over 30 radionuclides have been required to be reported per macrobatch to meet the WAPS waste form reporting specifications.

Next, the demonstrations of the DWPF process with the radioactive qualification samples have been performed for 17 years covering nine different sludge batches. At this point, the chemical reactions that occur during feed preparation in the DWPF are fairly well replicated by simulant testing across a range of conditions whereas testing with actual radioactive sludge samples is still performed from a glass processing or compliance testing. Therefore, the need for this demonstration should be re-evaluated based on the available data. Savannah River National Laboratory (SRNL) has already provided the technical basis for the elimination of the fabrication of a glass sample during the qualification process. DWPF is reviewing deletion of this requirement with DOE. Further reductions should be pursued and the program should be modified to determine the analytes of importance and other characteristics of the actual waste that cannot be replicated with simulants (e.g., rheology). This has the potential to shorten the duration of the qualification process, while reducing some associated costs with the qualification.

Finally, the glass durability requirements and potential modifications to reduce costs and schedule were discussed earlier. When DWPF was going through startup testing, there were concerns about model applicability over the projected composition region to be processed in DWPF so the compromise was to experimentally verify the models for each batch to ensure applicability via a variability study. This has shown to be an effective process but the process could be improved by implementing the generated data from previous sludge batch variability studies into the prediction/verification process.

Restrictions with the 897 g/m^3 Yucca Mountain Fissile Limit

Section 1.14.2.3.2.4 of the Yucca Mountain Repository License Application (LA) Safety Analysis Report (SAR) currently states the estimated fissile isotope concentration in SRS HLW canisters to be 897 g/m^3 [5]. The discussion in the Yucca Mountain SAR acknowledges that the fissile concentration is ~1 order of magnitude lower than the ANSI/ANS-8.1-1998 minimum subcritical limit and that the HLW glass has a significant margin of subcriticality. Because the HLW canisters are safely subcritical, the SAR documents that no further analysis is required to demonstrate the subcriticality of the individual HLW glass canisters.

In August 2007, SRS submitted a report in response to a Yucca Mountain Project request for the SRS glass composition and noted that the report's projected composition was not to be used for environmental modeling or accident analysis. The projected curie content of DWPF canisters did not include Pu "drops" from the SRS H Canyon or the Plutonium Disposition Project. In August 2008, DOE mandated to the SRS contractor that the total fissile concentration in DWPF glass to be at or below 897 g/m^3 to stay below the Yucca Mountain SAR value discussed above [6]. Sludge Batch 5 had to be limited to a maximum waste loading of 37 wt% to protect the 897 g/m^3 fissile limit and the contractor had to target an even lower 33% waste loading in DWPF SME batches to account for uncertainties.

The concentration of plutonium is typically low (on the order of 0.01 wt % PuO_2) in HLW glasses made from the reprocessing of DOE spent fuel. The behavior of plutonium in the glass at these levels has been analyzed and determined not to impact processing, criticality safety or glass performance. Recently, additional excess nuclear material plutonium has been identified for disposition with HLW vitrification considered as a primary disposition path. However, the 897 g/m^3 fissile limit has restricted the amount of plutonium that could be disposed into any one sludge batch. As discussed above, this limit does not have a sound technical basis and was put into effect because of the SRS HLW glass compositions that were incorrectly used in the Yucca Mountain SAR.

The DOE Office of Environmental Management (DOE-EM) tasked SRNL to assess the glass plutonium limit for typical HLW glasses to provide a technical basis to solve this problem [7]. The testing considered the solubility of PuO_2, glass durability, irradiation damage due to alpha-emitting plutonium ions and effects on glass processing.

SRNL determined that a plutonium loading of 1 wt% in glass was possible after completing initial studies with hafnium (Hf) as a surrogate for plutonium [8]. The 1 wt% in glass plutonium concentration translated to ~18 kg plutonium per DWPF canister. This would be ~ 10X the current allowed limit per the WAPS / International Atomic Energy Agency (IAEA) specification (2500 g/m^3 of glass) and about 30X the current 897 g/m^3 limit. The studies showed that the plutonium was homogeneously distributed and did not result in any formation of plutonium-containing crystalline phases as long as the glass was prepared under "well-mixed" conditions. The Hf surrogate results indicated that this higher concentration did not adversely impact glass viscosity or glass durability. Irradiation effects due to incorporation of plutonium must be considered specifically as related to long-term performance of the glass. Since plutonium is an alpha-emitter and alpha decay events will result in atom displacements, the recoil damage must be considered and the effects of these displacements on the glass must be understood. Based on the data available, the 1 wt% target appears to have minimal impact. Finally, evaluation of DWPF glass pour stream samples that had plutonium concentrations below the 897 g/m^3 limit showed that Pu concentrations in the glass pour stream were close to targeted compositions in the melter feed indicating that Pu neither volatilized from the melt nor stratified in the melter when processed in the DWPF melter. Therefore, incorporation of up to 1 wt% plutonium in glass appears to be a viable option through the DWPF vitrification processes.

Future of RW-0333P QA Program

The HLW vitrification program at DWPF currently operates under the quality assurance requirements of Revision 20 of RW-0333P [9]. Maintenance of this program is a cost burden to the site contractors, since SRS already maintains a NQA-1 quality assurance program. The need for maintenance of the RW-0333P program in light of the change in repository location should be reviewed given its potential cost savings. Since DOE-EM has been designated as the organization that will perform the previous defined functions of the Office of Civilian

Radioactive Waste Management (RW), it would seem prudent to evaluate whether the EM Quality Assurance Program (EM-QA-001) coupled with ASME NQA-1-2008 and the NQA-1a-2009 Addenda would be acceptable for the DWPF QA program.

Use of Higher Capacity Canisters (HCCs)

Under the existing WAPS, a specification is given for the size of the canisters containing HLW glass. Both SRS and Hanford contractors have expressed interest in reducing the wall thickness on the main body of the canister (not the top head or bottom). The current nominal wall thickness of the baseline DWPF and the Hanford Waste Treatment Plant (WTP) canisters is 0.95 cm (0.375 inches). The DWPF HCC has a nominal canister wall thickness of 0.342 cm (0.1345 inches - 10 gauge). This wall thickness reduction would reduce the amount of material used in fabrication but also allow for an increased volume of glass (e.g., 104%) to be poured in the same sized canister.

An HCC drop test literature search and engineering evaluation was completed in 2011 and it was concluded that proposed DWPF HCC design at a maximum glass fill height and a maximum glass specific gravity would not breach, rupture or leak material during or after a 7-meter bottom-end drop [10]. There was also conclusive evidence that the HCC design would pass through a 64 cm cylindrical cavity after the bottom-end drop [10].

The national laboratories could be used to resolve any remaining technical issues with the HCC including the effect of the reduced wall thickness on corrosion and material compatibility. The HCC design has the potential for significant benefit because of the total number of canisters to be produced at WTP (15,000+ canisters) and the remaining canisters to be poured at DWPF (3,000+ canisters).

CONCLUSIONS

As a result of DOE looking at alternative approaches for disposition of HLW following the termination of the Yucca Mountain Project, it is an opportune time to review the existing DOE HLW requirements documents as well as the Producer's compliance strategy documents to recommend changes that can result in significant production cost savings over the lifetime of DWPF and other HLW Producer sites. Compliance areas to be targeted for changes include reevaluating the uncertainties associated with glass models to allow higher waste loadings, reexamining radionuclide reporting requirements and associated strategies, reassessing the overall approach to "real waste" qualification and the testing to be performed, removing the arbitrary 897 g/m^3 fissile limit for DWPF canisters, evaluating whether the costly RW-0333P QA program continues to be necessary given the current repository situation, and continuing to study the HCC design that will allow 4% more glass volume per canister.

REFERENCES

1. DOE Office of Environmental Management, **Waste Acceptance Product Specifications for Vitrified High Level Waste Forms**, DOE/EM-0093, Revision 2 (December 1996).
2. DOE Office of Civilian Radioactive Waste Management, **Waste Acceptance System Requirements Document**, DOE/RW-0351, Revision 5 (May 2007).
3. WSRC-IM-91-116-0, **DWPF Waste Form Compliance Plan (U)**, Revision 9, Savannah River Site (June 2012).
4. WSRC-IM-91-116-X (X = 1 through 13), **DWPF Waste Form Qualification Report**, Savannah River Site (multiple revision dates).
5. DOE Office of Civilian Radioactive Waste Management, **Yucca Mountain Repository License Application Safety Analysis Report**, DOE/RW-0573, Revision 1 (November 2008).
6. T. J. SPEARS, "Fissile Isotope Concentration in Defense Waste Processing Facility (DWPF Canisters," WDPD-08-075 (August 2008).
7. J. C. MARRA and C. L. CRAWFORD, "Task Technical and Quality Assurance Plan: Pu Solubility Studies," SRNL-RP-2009-01450, Savannah River National Laboratory (December 2009).
8. J. C. MARRA, C. L. CRAWFORD, K. M. FOX and N. E. BIBLER, "Plutonium Solubility in High-Level Waste Alkali Borosilicate Glass," SRNL-STI-2010-00766, Savannah River National Laboratory (January 2011).
9. DOE Office of Civilian Radioactive Waste Management, **Quality Assurance Requirements and Description for the Civilian Radioactive Waste Management Program**, DOE/RW-0333P, Revision 20 (October 2008).
10. S. SHIRAGA, "Higher Capacity Canister (HCC) Drop Test Literature Search and Engineering Evaluation," EnergySolutions (September 2011).

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