Contract No:

This document was prepared in conjunction with work accomplished under Contract No. 89303321CEM000080 with the U.S. Department of Energy (DOE) Office of Environmental Management (EM).

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Evaluation of Glass Density to Support the Estimation of Fissile Mass Loadings in Sludge Batch 10 Glasses

F.C. Johnson

January 2023

SRNL-STI-2022-00054, Revision 1

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Printed in the United States of America

Prepared for
U.S. Department of Energy

Keywords: *SB10, fissile mass loading, glass density, Frit 473, Frit 625*

Retention: *Lifetime*

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Savannah River National Laboratory is operated by Battelle Savannah River Alliance for the U.S. Department of Energy under Contract No. 89303321CEM000080.

EXECUTIVE SUMMARY

Per a directive from the Department of Energy Savannah River Operations Office (DOE-SR) in 2008, the fissile mass loading concentration must remain below 897 g/m^3 in each high-level waste (HLW) glass canister produced by the Defense Waste Processing Facility (DWPF). To support Sludge Batch 5 (SB5) processing, the Savannah River National Laboratory (SRNL) developed a technical basis that facilitates the evaluation of fissile mass loading of the glass product. The calculation is based on the iron (Fe) concentration in the glass as determined by measurements from the Slurry Mix Evaporator acceptability analysis as well as the glass density. In April 2022, a subsequent DOE-SR directive increased the fissile mass loading limit to 2500 g/m^3 beginning with Sludge Batch 11. Thus, the 897 g/m^3 limit still applies to Sludge Batch 10 (SB10) processing.

For SB5 through initial Sludge Batch 9 (SB9) processing prior to coupled operation with the Salt Waste Processing Facility (SWPF), SRNL provided DWPF a bounding glass density value that was based on a statistical evaluation of density measurements. To eliminate the need for experimental work, a composition-based density model for HLW glasses was developed at SRNL in 2019. The objective of this report is to present the bounding glass density determined with the composition-based density model for SB10 sludge-only (SO) and coupled processing.

Revision 1 is a total rewrite and presents the bounding glass density based on updated SB10 projections.

SB10 is comprised of material from Tanks 11H, 13H, 15H, 26F, 40H (heel only), and Alternate Feed Stock-2 and Sodium Reactor Experiment material from H-Canyon. Frit 473 was recommended for SO and coupled processing with SWPF based on previous assessments of SB10 projections with the DWPF Product Composition Control System (PCCS) glass property models and their associated Measurement Acceptance Region constraints. Savannah River Mission Completion (SRMC) subsequently pursued Wash Cycle Y to further reduce the total sulfur in the sludge batch and increase processing flexibility at DWPF. Based on the Tank 51 confirmation sample results, the baseline Tank 40H blend projection was low in Na and a projected volume of caustic was added to increase the Na concentration comparable to previous SB10 projections. In November 2022, SRMC System Planning provided an updated SB10 Tank 40H blend projection based on the Tank 51 confirmation sample results with 7,000 gallons of added caustic.

Based on this projection and Frit 473, glass compositions were developed for SO processing and following cases for coupled operation with SWPF. Frit 625 was also evaluated for use during the SB9 to SB10 transition.

- Case 1: Single strike operation with no entrained insoluble sludge solids (ISS). This case represents the baseline.
- Case 3A: Single strike operation, with 600 mg/L of entrained ISS, which were assumed to be SB9 sludge solids.
- Sludge and strip effluent

A bounding glass density of 2.842 g/cm^3 was determined for the SB10 glass region based on both Frit 473 and Frit 625 for SO and coupled processing at 32-40% WL. Upon the start of SB10 processing, the glass density input in the PCCS online application will be updated by DWPF.

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LIST OF ABBREVIATIONS

DOE-SR	Department of Energy Savannah River Operations Office
DWPF	Defense Waste Processing Facility
EVs	extreme vertices
HLW	high-level waste
ISS	insoluble sludge solids
MAR	Measurement Acceptance Region
MST/SS	monosodium titanate/sludge solids
NGS	Next Generation Solvent
PCCS	Product Composition Control System
SB5	Sludge Batch 5
SB9	Sludge Batch 9
SB10	Sludge Batch 10
SE	strip effluent
SME	Slurry Mix Evaporator
SO	sludge-only
SRAT	Sludge Receipt and Adjustment Tank
SRNL	Savannah River National Laboratory
SRMC	Savannah River Mission Completion
SSRT	Sludge Solids Receipt Tank
SWPF	Salt Waste Processing Facility
wt.%	weight percent

1.0 Introduction

Per a directive from the Department of Energy Savannah River Operations Office (DOE-SR) in 2008, the fissile mass loading concentration must remain below 897 g/m^3 in each high-level waste (HLW) glass canister produced by the Defense Waste Processing Facility (DWPF).[1,2] To support Sludge Batch 5 (SB5) processing, the Savannah River National Laboratory (SRNL) developed a technical basis that facilitates the evaluation of fissile mass loading of the glass product.[3] The calculation is based on the iron (Fe) concentration in the glass as determined by measurements from the Slurry Mix Evaporator (SME) acceptability analysis as well as the glass density. In April 2022, a subsequent DOE-SR directive increased the fissile mass loading limit to 2500 g/m^3 beginning with Sludge Batch 11.[4] Thus, the 897 g/m^3 limit still applies to Sludge Batch 10 (SB10) processing.

For SB5 through initial Sludge Batch 9 (SB9) processing prior to coupled operation with the Salt Waste Processing Facility (SWPF), SRNL provided DWPF a bounding glass density value that was based on a statistical evaluation of density measurements.[3,5-9] To eliminate the need for experimental work, a composition-based density model for HLW glasses was developed at SRNL in 2019.[10,a] The objective of this report is to present the bounding glass density determined with the composition-based density model for SB10 sludge-only (SO) and coupled processing.

Revision 1 is a total rewrite and presents the bounding glass density based on updated SB10 projections.

2.0 Quality Assurance

This work was requested via a Technical Task Request and directed by a Task Technical and Quality Assurance Plan.[11,12] The Functional Classification of this task is Safety Class. Glass density is waste form affecting and needs to follow the quality assurance requirements of RW-0333P.[13] Microsoft Excel and JMP Version 16.0.0 were used to support this work.[14,15] Requirements for performing reviews of technical reports and the extent of review are established in Manual E7, Procedure 2.60.[16] This document, including calculations, was reviewed by a Design Verification. SRNL documents the Design Verification using the SRNL Technical Report Design Checklist contained in WSRC-IM-2002-00011, Rev. 2.[17] The Design Checklists and summarized glass density values for this report are stored in electronic laboratory notebook experiment C7592-00311-50.

3.0 Inputs and Assumptions

SB10 is comprised of material from Tanks 11H, 13H, 15H, 26F, 40H (heel only), and Alternate Feed Stock-2 and Sodium Reactor Experiment material from H-Canyon. Frit 473[b] was recommended for SO and coupled processing with SWPF based on previous assessments of SB10 projections with the DWPF Product Composition Control System (PCCS) glass property models and their associated Measurement Acceptance Region (MAR) constraints.[18,19] Savannah River Mission Completion (SRMC) subsequently pursued Wash Cycle Y to further reduce the total sulfur in the sludge batch and increase processing flexibility at DWPF. In October 2022, SRMC System Planning provided an updated SB10 Tank 40H blend baseline projection based on the Tank 51 confirmation sample results.[20,21] Due to the reduced Na concentration relative to previous SB10 projections, two additional projections based on the addition of 7,000 gallons of caustic were subsequently provided.[22] MAR assessments confirmed that Frit 473 remains viable for SB10 processing with the caustic addition of 7,000 gallons.[23] The viability of Frit 625[c] was also evaluated for use during the SB9 to SB10 transition.

[a] Revision 1 of SRNL-STI-2018-00599 was issued in 2022 to change the Functional Classification to Safety Class.

[b] $8B_2O_3$-$8Li_2O$-$5Na_2O$-$79SiO_2$ in weight percent.

[c] $1Al_2O_3$-$8B_2O_3$-$7Li_2O$-$6Na_2O$-$78SiO_2$ in weight percent.

3.1 SB10 SO Projections

In November 2022, SRMC System Planning provided two updated SB10 Tank 40H blend projections with 7,000 gallons of added caustic[d] based on Tank 51 heels of 10 inches and 30 inches. The elemental concentrations were converted to oxides and normalized to 100 weight percent (wt.%) as shown in Appendix Table A-1.

3.2 SRNL Developed Inputs for Coupled Operation

SRNL performed subsequent calculations with the SO projection in Appendix Table A-1 to estimate compositions in the Sludge Receipt and Adjustment Tank (SRAT) during coupled operation with SWPF. These calculations were based on projected compositions for strip effluent (SE) as well as the following three cases for compositions of monosodium titanate/sludge solids (MST/SS) in the Sludge Solids Receipt Tank (SSRT) effluent that were originally developed for SB9:[24-27]

- Case 1: Single strike operation with no entrained insoluble sludge solids (ISS). This case represents the baseline.
- Case 3A: Single strike operation, with 600 mg/L of entrained ISS, which were assumed to be SB9 sludge solids.[28]
- Sludge and SE only

Case 2 (single strike with no aluminum) and Case 5 (double strike with no aluminum) were eliminated as these cases were not applicable to SB10.[e,25] Case 3B (single strike with 1200 mg/L of entrained SB9 ISS) and Case 4 (double strike) are unlikely conditions and were not evaluated.

Other pertinent inputs include:

- DWPF receives 5700 gallons of sludge slurry from Tank 40H per SRAT batch[29]
- Nominal single strike operation results in 2800 gallons of the SSRT effluent stream (MST/SS)[25]
  - Incremental variation in these transfer volumes were initially requested for SB9,[25] which SRNL assumed to be ±400 gallons for these assessments; however, DWPF also requested that 4500 gallons be evaluated for single strike operation.[30] This evaluation only included a volume of 4500 gallons as the previous SB10 glass density evaluation (Revision 0 of this report) demonstrated that this volume resulted in the highest glass density as shown in Appendix A Table A-2.
- DWPF receives 15,000 gallons of SE per SRAT batch (BOBCalixC6 solvent[f] or Next Generation Solvent (NGS)[g])[25,29]
- Cs-137 concentration in SE is 66 Ci/gallon[31]

4.0 Method for Determining a Bounding Glass Density

As proposed in Reference 10, SRNL added the capability of predicting densities of glass compositions evaluated during PCCS MAR assessments.[10] Note that Gd_2O_3 is present in the updated Tank 40H blend projections at ~0.1 wt.%; however, it is not an included component in PCCS.[18] Per the DWPF Glass Product Control Program, Gd_2O_3 is currently considered a trace component (elemental concentration <0.5 wt.%) and can be ignored for SB10 process control.[32] Thus, glass densities were initially completed without Gd_2O_3 during PCCS MAR assessments and a second set of calculations were performed to account for Gd_2O_3. A summary of this method is provided below.

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[d] 50 weight percent NaOH.

[e] Per SRR-WSE-2018-00025, more conservative cases were developed for SB9 where the aluminum concentration in the SSRT effluent stream was set to 0 mg/L. SB10 has a higher Al_2O_3 concentration than SB9, thus there is no concern with the Al_2O_3 concentration being too low in glass for SB10 and Cases 2 and 5 were eliminated.

[f] BOBCalixC6 is calix[4]arene-bis(tert-octylbenzo-crown-6) , which uses a nitric acid strip solution.

[g] NGS contains the extractant MaxCalix (1,3-alt-25,27-bis(3,7- dimethyloctyl-1-oxy) calix[4]arene-benzocrown-6), which uses a boric acid strip solution.

Compositional variation (±) was applied to the SO and coupled processing SRAT compositions (described in Section 3.0) to account for likely, but not necessarily bounding, differences that may be present in the material that is transferred from Tank 40H and SWPF into the SRAT during the processing of SB10. The compositional variation for the individually tracked oxides was represented by the larger of 0.5 wt.% or 7.5% of the nominal concentration.[33] Those oxides not tracked individually were grouped into an "Others" component (without Gd_2O_3).[h] Extreme vertices (EVs) were generated using the oxide intervals for each SB10 SRAT scenario and represent the corner points of a multidimensional SB10 composition space. These EVs were combined with either Frit 473 or Frit 625 over the interval of 32-40% waste loading (WL). This WL interval represents the SB10 target WL of 36% ± 4 percentage points. Approximately 2.4 million glass compositions were evaluated against the PCCS MAR criteria to determine whether each composition would pass the SME acceptability process.[18] Glass compositions that failed any MAR constraints were excluded from further evaluation. Densities of the remaining acceptable glass compositions were predicted with the density model and the maximum glass densities of each SB10 scenario were determined (SO, sludge and SE, Case 1 and Case 3A).[10] The specific SRAT EVs resulting in the maximum glass densities for each scenario were determined and Gd_2O_3 was added to the "Others" component. Glass compositions were recalculated and glass densities including Gd_2O_3 were determined.

5.0 Results

Based on the assumptions provided in Section 3.0, a bounding density of 2.842 g/cm^3 was determined for the SB10 glass region, which was calculated by applying the density model uncertainty of 0.123 g/cm^3 to the maximum density value of 2.719 g/cm^3. The uncertainty reflects 99% coverage at 99% confidence.[10] As shown in Appendix A Table A-3 and Table A-4, this maximum density value corresponds to Frit 625 with Case 3A (4500 gallons of the MST/SS stream and BOBCalixC6 solvent) at 40% WL for the projection with the 30" heel. Variation of the maximum glass density for Case 3A between Frit 625 and Frit 473 is minimal (~0.004 g/cm^3). The inclusion of the projected SB10 Gd_2O_3 concentration had no impact on the bounding glass density.

6.0 Conclusions

A bounding glass density of 2.842 g/cm^3 was determined for the SB10 glass region based on both Frit 473 and Frit 625 for SO and coupled processing at 32-40% WL.

7.0 Path Forward

Upon the start of SB10 processing, the glass density input in the PCCS online application will be updated by DWPF with the bounding glass density of 2.842 g/cm^3 or if needed, a case-specific maximum glass density as shown in Appendix Table A-3 or Table A-4.[34]

8.0 Recommendation

For Sludge Batch 11, it is recommended that the glass density script used in JMP Version 16.0.0 be updated to include other elements of interest (e.g. Gd) so that a second set of calculations is not required. This change would also prompt a revision to the various software quality assurance documents.

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Appendix A. Tank 40H Blend Projections and Glass Density Results

Table A-1. Normalized SB10 Confirmation Sample Tank 40H Blend Projections – November 2022 (wt.%)

Oxide	**7000 gal Caustic 10" Heel**	**7000 gal Caustic 30" Heel**
Al_2O_3	28.10	27.42
B_2O_3	0.05	0.05
BaO	0.09	0.09
CaO	1.33	1.35
Ce_2O_3	0.24	0.24
Cr_2O_3	0.23	0.23
CuO	0.07	0.07
Fe_2O_3	22.03	22.27
Gd_2O_3	0.09	0.09
K_2O	0.17	0.17
La_2O_3	0.06	0.06
Li_2O	0.09	0.09
MgO	0.49	0.48
MnO	6.20	6.29
Na_2O	30.43	30.74
NiO	0.90	0.93
PbO	0.05	0.05
SO_4^{2-}	0.88	0.88
SiO_2	2.00	2.06
ThO_2	2.26	2.21
TiO_2	0.04	0.04
U_3O_8	4.08	4.08
ZnO	0.04	0.04
ZrO_2	0.09	0.09

Table A-2. Summary of Glass Density Results (August 2022 Projection - Revision 0)

Frit	Case	MST/SS Transfer Volume (gal)	Solvent	Max Density (g/cm^3)	Max Density + Uncertainty (g/cm^3)
473	Sludge-only	0	None	2.707	2.830
	Case 1	2,400	BOBCalixC6	2.712	2.835
	Case 1	2,800	BOBCalixC6	2.711	2.834
	Case 1	4,500	BOBCalixC6	2.711	2.834
	Case 3A	2,400	BOBCalixC6	2.721	2.844
	Case 3A	2,800	BOBCalixC6	2.722	2.845
	Case 3A	4,500	BOBCalixC6	2.726	2.849
	Sludge and SE	0	BOBCalixC6	2.713	2.836
	Case 1	2,400	NGS	2.710	2.833
	Case 1	2,800	NGS	2.710	2.833
	Case 1	4,500	NGS	2.710	2.833
	Case 3A	2,400	NGS	2.720	2.843
	Case 3A	2,800	NGS	2.721	2.844
	Case 3A	4,500	NGS	2.725	2.848
	Sludge and SE	0	NGS	2.711	2.834
625	Sludge-only	0	None	2.711	2.834
	Case 1	2,400	BOBCalixC6	2.715	2.838
	Case 1	2,800	BOBCalixC6	2.715	2.838
	Case 1	4,500	BOBCalixC6	2.715	2.838
	Case 3A	2,400	BOBCalixC6	2.725	2.848
	Case 3A	2,800	BOBCalixC6	2.726	2.849
	Case 3A	4,500	BOBCalixC6	2.730	2.853
	Sludge and SE	0	BOBCalixC6	2.717	2.840
	Case 1	2,400	NGS	2.714	2.837
	Case 1	2,800	NGS	2.714	2.837
	Case 1	4,500	NGS	2.714	2.837
	Case 3A	2,400	NGS	2.724	2.847
	Case 3A	2,800	NGS	2.725	2.848
	Case 3A	4,500	NGS	2.729	2.852
	Sludge and SE	0	NGS	2.715	2.838

Table A-3. Summary of Glass Density Results (November 2022 Projection – 10” Heel)

Frit	Case	MST/SS Transfer Volume (gal)	Solvent	Max Density (g/cm^3)	Max Density + Uncertainty (g/cm^3)
473	Sludge-only	0	None	2.690	2.813
	Case 1	4,500	NGS	2.697	2.820
	Case 3A	4,500	NGS	2.713	2.836
	Sludge and SE	0	NGS	2.695	2.818
	Case 1	4,500	BOBCalixC6	2.698	2.821
	Case 3A	4,500	BOBCalixC6	2.714	2.837
	Sludge and SE	0	BOBCalixC6	2.696	2.819
625	Sludge-only	0	None	2.694	2.817
	Case 1	4,500	NGS	2.701	2.824
	Case 3A	4,500	NGS	2.717	2.840
	Sludge and SE	0	NGS	2.699	2.822
	Case 1	4,500	BOBCalixC6	2.702	2.825
	Case 3A	4,500	BOBCalixC6	2.718	2.841
	Sludge and SE	0	BOBCalixC6	2.700	2.823

Table A-4. Summary of Glass Density Results (November 2022 Projection – 30” Heel)

Frit	Case	MST/SS Transfer Volume (gal)	Solvent	Max Density (g/cm^3)	Max Density + Uncertainty (g/cm^3)
473	Sludge-only	0	None	2.692	2.815
	Case 1	4,500	NGS	2.699	2.822
	Case 3A	4,500	NGS	2.714	2.837
	Sludge and SE	0	NGS	2.697	2.820
	Case 1	4,500	BOBCalixC6	2.700	2.823
	Case 3A	4,500	BOBCalixC6	2.715	2.838
	Sludge and SE	0	BOBCalixC6	2.698	2.821
625	Sludge-only	0	None	2.696	2.819
	Case 1	4,500	NGS	2.703	2.826
	Case 3A	4,500	NGS	2.718	2.841
	Sludge and SE	0	NGS	2.701	2.824
	Case 1	4,500	BOBCalixC6	2.704	2.827
	Case 3A	4,500	BOBCalixC6	2.719	2.842
	Sludge and SE	0	BOBCalixC6	2.702	2.825