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TECHNOLOGY DEMONSTRATION OF SLUDGE MASS REDUCTION VIA ALUMINUM DISSOLUTION: GLASS FORMULATION PROCESSING WINDOW PREDICTIONS FOR SB5

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Process Science and Engineering
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Prepared for the U.S. Department of Energy Under
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EXECUTIVE SUMMARY

Composition projections for Sludge Batch 5 (SB5) were developed, based on a modeling approach at the Savannah River National Laboratory (SRNL), to evaluate possible impacts of the Al-dissolution process on the availability of viable frit compositions for vitrification at the Defense Waste Processing Facility (DWPF). The study included two projected SB5 compositions that bound potential outcomes (or degrees of effectiveness) of the Al-dissolution process, as well as a nominal SB5 composition projection based on the results of the recent Al-dissolution demonstration at SRNL. The three SB5 projections were the focus of a two-stage paper study assessment. A Nominal Stage assessment combined each of the SB5 composition projections with an array of 19,305 frit compositions over a wide range of waste loading (WL) values and evaluated them against the DWPF process control models. The Nominal Stage results allowed for the down-selection of a small number of frits that provided reasonable projected operating windows (typically 27 to 42 wt % WL). The frit/sludge systems were mostly limited by process related constraints, with only one system being limited by predictions of nepheline crystallization, a waste form affecting constraint. The criteria applied in selecting the frit compositions somewhat restricted the compositional flexibility of the candidate frits for each individual SB5 composition projection, which may limit the ability to further tailor the frit for improved melt rate.

Variation Stage assessments were then performed using the down-selected frits and the three SB5 composition projections with variation applied to each sludge component. The Variation Stage results showed that the operating windows were reduced in width, as expected when variation in the sludge composition is applied. However, several of the down-selected frits exhibited a relatively high degree of robustness to the applied sludge variation, providing WL windows of approximately 30 to 39 wt %. The maximum WLs were limited by processing constraints, liquidus temperature and low viscosity, rather than a waste form affecting constraint (e.g., nepheline crystallization) in the Variation Stage assessments.

These paper study assessments have identified candidate frits which, when combined with the SRNL projected SB5 compositions after Al-dissolution, have projected operating windows that should be reasonable for DWPF processing. As more information is obtained on the SB5 composition to be processed in DWPF, including the actual Al removed and Tank 7 mass transferred, additional paper study assessments will be performed as well as experimental frit development studies. The frits identified in this study provide insight into potential processing windows but are not the recommended frits for SB5. No information regarding melt rate can be inferred from the paper study results. Experimental studies to evaluate this critical factor in DWPF processing must be performed on the best SB5 projection before a frit recommendation could be made for any projected sludge composition.

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LIST OF ABBREVIATIONS

CPC	Chemical Process Cell
DOE	Department Of Energy
DWPF	Defense Waste Processing Facility
EVs	Extreme Vertices
hFrit	high concentration of frit components
highv	high viscosity
Homg	homogeneity
lowv	low viscosity
LWO	Liquid Waste Organization
MAR	Measurement Acceptability Region
Neph	Nepheline crystallization
NL	Normalized Leachate
PCCS	Product Composition Control System
SB4	Sludge Batch 4
SB5	Sludge Batch 5
SRS	Savannah River Site
TL	liquidus temperature
WL	Waste Loading

1.0 Introduction

Tank 51 will be blended with Purex sludge from Tank 7 to constitute Sludge Batch 5 (SB5). The Savannah River Site (SRS) Liquid Waste Organization (LWO) is performing low-temperature aluminum-dissolution in Tank 51 to reduce the total mass of sludge solids being fed to the Defense Waste Processing Facility (DWPF). Before this process was performed in the Tank Farm, a radioactive demonstration using a 3 L Tank 51 sludge slurry sample was performed at the Savannah River National Laboratory (SRNL) to determine the effectiveness of the lower temperature process.[1] The aluminum-dissolved sludge was used to determine potential downstream impacts so that technical issues could be identified before the start of SB5 processing. The potential downstream impacts assessed include the Tank Farm washing and concentration process and the DWPF Chemical Process Cell (CPC) and melter processing envelopes.

Paper study assessments of the composition projections are used to assess various frit options of interest with respect to the projected operating windows (as defined by a waste loading interval) for DWPF. More specifically, for each sludge option, the current Product Composition Control System (PCCS) models[2] are used to assess the waste loading interval over which glasses would concurrently meet all process and acceptability constraints. Candidate frits are identified that provide a reasonable projected operational window over the anticipated composition region of interest and are robust to anticipated sludge composition variations.

The two stages – Nominal Stage and Variation Stage – traditionally performed by Peeler and Edwards[3] are employed to assess the various frit/sludge combinations with respect to these key criteria. The Nominal Stage will utilize nominal compositions representing the potential scenarios outlined above (i.e., various amounts of alumina removed from the sludge). This stage identifies candidate frit compositions with respect to their ability to provide a reasonable operating window based solely on a specific nominal composition – no sludge composition variation is considered in this phase.

The Variation Stage assessment is performed to gain insight into the robustness of the candidate frits with respect to potential variation in the Tank Farm's projected sludge composition. This potential variation arises due to uncertainty in the planned blending strategies and tank volumes. A down-select process is used to identify the primary frit candidates from the Nominal Stage results prior to performing the Variation Stage assessment.

2.0 Objectives

This report focuses on the impacts to the development of a glass frit to be combined with the reduced Al concentration sludge for vitrification in the DWPF melter. An assessment is made of the impact of Al-dissolution on the DWPF projected operating windows as defined by the current process control models. The evaluation includes projected SB5 compositions that bound potential outcomes (or degrees of effectiveness) of the Al-dissolution process, as well as a nominal SB5 composition projection based on the outcome of the recent Al-dissolution demonstration at SRNL.[1]

The paper study assessments do not provide any estimates of melt rate performance among the various SB5 projections or frit compositions. Experimental studies will be necessary to provide melt rate information and to guide further decisions on frit compositions for processing at the DWPF.

This work is Technical Baseline Research and Development for the Department of Energy (DOE) Office of Cleanup Technologies (EM-21) and is performed under task technical and quality assurance plan WSRC-RP-2007-00512.[4]

3.0 Sludge Batch 5 Composition Projections

SRNL used a modeling approach to project the anticipated composition of SB5 in support of this study. A detailed description of the modeling methodology is provided in WSRC-STI-2008-00001.[5] The model required the following input vectors, which were constructed from available analytical data:

- Tank 51 slurry prior to dilution with Tank 40 supernate[6]
- Tank 40 supernate[a]
- Tank 7 slurry[7]
- Information on various water leaks, miscellaneous additions, missing ion chromatography data, etc.[b]

Five composition projection cases were developed for SB5 at the initiation of this study. The five cases project the potential outcomes of the low-temperature Al-dissolution process based on the partitioning of Al between Gibbsite and Boehmite in Tank 51. The amount of Gibbsite was varied between 0% and 100% in increments of 25%. The projections assumed a blend of approximately 80% material from Tank 51 and 20% material from Tank 40 to constitute the SB5 feed to DWPF (i.e., a 40 inch heel remaining in Tank 40 when the blend occurs). These composition projections for SB5 – the output of the SRNL model – are given in Table 3-1. The projections are listed as a function of Gibbsite/Boehmite partitioning, and labeled SB5 Cases A through E.

[a] Analytical Laboratories report 23Apr07 09:31 Hr
[b] Tank Farm Spreadsheet 19Jun07

Table 3-1. SB5 composition projections as a function of Gibbsite/Boehmite partitioning.

SB5 Case	A	B	C	D	E
Gibbsite (%)	0	25	50	75	100
Boehmite (%)	100	75	50	25	0
Ag	0.008	0.009	0.010	0.011	0.011
Al	17.063	15.326	13.167	11.062	10.140
Ba	0.083	0.091	0.101	0.108	0.110
Ca	1.163	1.273	1.404	1.505	1.539
Cd	0.049	0.053	0.058	0.063	0.065
Ce	0.289	0.316	0.349	0.374	0.383
Co	0.017	0.019	0.020	0.022	0.022
Cr	0.233	0.255	0.281	0.301	0.307
Cu	0.010	0.010	0.010	0.010	0.011
Fe	14.855	16.251	17.926	19.216	19.646
K	0.052	0.055	0.059	0.066	0.069
La	0.127	0.139	0.153	0.164	0.168
Mg	0.649	0.710	0.783	0.839	0.858
Mn	3.439	3.763	4.150	4.449	4.549
Na	18.877	18.413	17.938	18.164	18.545
Ni	1.960	2.144	2.365	2.535	2.592
P	0.211	0.230	0.253	0.271	0.277
Pb	0.019	0.020	0.022	0.024	0.025
Pd	0.001	0.001	0.001	0.001	0.001
Rh	0.024	0.026	0.029	0.031	0.032
Ru	0.091	0.099	0.109	0.117	0.120
S	0.235	0.243	0.254	0.272	0.281
Si	0.818	0.881	0.972	1.044	1.067
Sr	0.246	0.269	0.297	0.318	0.325
Ti	0.014	0.015	0.017	0.018	0.019
U	5.761	6.303	6.952	7.453	7.619
Zn	0.012	0.013	0.013	0.015	0.015
Zr	0.175	0.191	0.211	0.226	0.231
Total (wt %)	66.483	67.120	67.906	68.681	69.026
Tk51 Transfer (kg)	238,491	200,032	173,315	161,004	160,709
TK40 Heel (kg)	37,733	37,733	37,733	37,733	37,733
Tk51 Solids (%)	86	84	82	81	81
Tk40 Solids (%)	14	16	18	19	19

The initial results of the 3L Al-dissolution demonstration in the SRNL Shielded Cells facility showed that the Al was partitioned as approximately 39% Gibbsite.[1] This suggested that the SB5 composition would fall between Case B (25% Gibbsite) and Case C (50% Gibbsite). The model was run again with Al partitioned as 39% Gibbsite and the results are labeled as Case F. Table 3-2 lists the SB5 Case F composition projection, as well as Cases B and C, which will be the focus of the following paper study assessment. Cases B and C are included to allow for potential variation from the 39% Gibbsite value when the actual Al-dissolution process is performed in Tank 51.

Table 3-2. SB5 composition projection (SB5 Case F) based on 39% dissolution of aluminum. Cases B and C are included for comparison.

SB5 Case	F	B	C
Gibbsite (%)	39	25	50
Boehmite (%)	61	75	50
Ag	0.009	0.009	0.010
Al	14.38	15.326	13.167
Ba	0.096	0.091	0.101
Ca	1.345	1.273	1.404
Cd	0.056	0.053	0.058
Ce	0.331	0.316	0.349
Co	0.019	0.019	0.020
Cr	0.266	0.255	0.281
Cu	0.010	0.010	0.010
Fe	17.120	16.251	17.926
K	0.057	0.055	0.059
La	0.145	0.139	0.153
Mg	0.747	0.710	0.783
Mn	3.967	3.763	4.150
Na	17.967	18.413	17.938
Ni	2.249	2.144	2.365
P	0.242	0.230	0.253
Pb	0.021	0.020	0.022
Pd	0.001	0.001	0.001
Rh	0.028	0.026	0.029
Ru	0.104	0.099	0.109
S	0.249	0.243	0.254
Si	0.921	0.881	0.972
Sr	0.286	0.269	0.297
Ti	0.016	0.015	0.017
U	6.612	6.303	6.952
Zn	0.013	0.013	0.013
Zr	0.202	0.191	0.211
Total (wt %)	67.459	67.120	67.906
Tk51 Transfer (kg)	268,317	200,032	173,315
TK40 Heel (kg)	54,322	37,733	37,733
Tk51 Solids (%)	83	84	82
Tk40 Solids (%)	17	16	18

4.0 Candidate Frit Compositions

An array of frit compositions was developed to combine with SB5 Cases B, C and F in the Nominal Stage assessment. The frit components and their concentration ranges were chosen based on SRNL experience in previous frit development efforts,[8-15] DWPF operational constraints and practicality issues related to frit production. Frit components and their concentrations defining the frit array are shown in Table 4-1. For each frit composition, the concentration of SiO_2 was allowed to float as necessary to accommodate the concentrations of the other oxide components. A total of 19,305 frits were defined using this array.

Table 4-1. Frit components and concentration ranges used to define the frit composition array for paper study assessments.

Component	Min. Concentration (wt %)	Max. Concentration (wt %)	Increment (wt %)
B_2O_3	8.0	20.0	1.0
CaO	0.0	8.0	2.0
Li_2O	4.0	12.0	1.0
MgO	0.0	4.0	2.0
Na_2O	2.0	12.0	1.0
SiO_2	44.0	86.0	1.0

5.0 Nominal Stage Assessments

Sludge Cases B, C and F were each combined with the array of frits over a waste loading (WL) interval of 25 to 60 wt % and evaluated against the models currently implemented in the DWPF to constitute the Nominal Stage assessment. Property predictions assessed include those for liquidus temperature (TL), viscosity (η), durability (normalized leachate for boron, NL[B]), homogeneity (Homg), high viscosity (highv), low viscosity (lowv), high chromia concentration (Cr2O3), high sulfate concentration (SO4), high concentration of frit components (hFrit) and nepheline formation (Neph).[a]

The constraints associated with minimum Al_2O_3 concentrations in glass were also used in these assessments. Current PCCS criteria dictate that the Al_2O_3 content in the glass must be at least 3 wt % (not including uncertainties). For glasses containing more than 3 wt % Al_2O_3 but less than 4 wt %, there is an additional constraint limiting the sum of alkaline oxides in the glass to 19.3 wt % or less. For glasses containing at least 4 wt % Al_2O_3 (not including uncertainties), there is not an implied upper alkali constraint over the glass compositional regions previously tested. These constraints were implemented in PCCS based on the recommendations by Edwards et al.[17]

It should also be noted that a SO_4^{2-} solubility limit of 0.4 wt % was used in these assessments. It is anticipated that the sulfate limit for the SB5 system will be the same as that for the Sludge Batch 4 (SB4) system: 0.60 wt % SO_4^{2-} or 0.88 wt % Na_2SO_4 in glass. This potential increase in the SO_4^{2-} limit should be taken into consideration if any frit/sludge systems are found to be restricted by the 0.4 wt % SO_4^{2-} constraint in the Nominal Stage assessment.

The Nominal Stage results for the three SB5 projections combined with the 19,305 frits were evaluated and a smaller number of frits were down-selected for additional study. Two additional criteria were used in order to reduce the number of candidate frit compositions to a reasonable amount. First, only frits that provided operating windows of at least 15 percentage points (in terms of wt % WL) were considered. Second, the frits could not fail the nepheline constraint at WLs below 45 wt %. Applying these criteria left 44 potential frit compositions for SB5 Case B, 60 potential frit compositions for SB5 Case C, and 98 potential frit compositions for SB5 Case F.

[a] Note that SRNL has previously recommended that the homogeneity and high frit constraints be removed for sludge only processing in the DWPF.[16] However, these changes have not yet been implemented in PCCS.

A final set of criteria was applied in order to identify a small number of candidate frits for each SB5 composition projection. The candidate frits were chosen based on:

- Relatively high B_2O_3 and Na_2O concentrations, which are expected to improve melt rate,
- Minimal Li_2O concentrations to reduce frit cost, and
- Minimal CaO concentrations to avoid potential crystallization of calcium-rich phases in the melter.

These criteria aided in the selection of three candidate frits for each SB5 projection. However, it should be noted that there were more than three frits available for each of the SB5 projections that met all of the above criteria. Therefore, the number of frits chosen as candidates represents a number that was considered reasonable for performing the paper study assessments rather than a complete set of the available options.

The compositions of the three candidate frits selected for each SB5 composition projection are given in Table 5-1. Note that the criteria applied in selecting the frit compositions somewhat restrict the compositional flexibility of the candidate frits for each individual SB5 composition projection, which may limit the ability to further tailor the frit for improved melt rate. The candidate frits cover a relatively narrow range of B_2O_3 concentrations, which may hinder the ability to use B_2O_3 to improve melt rate and/or suppress nepheline crystallization. In general, the concentration of Li_2O in the candidate frits is relatively high – even though candidates with minimal Li_2O concentrations were chosen – as compared to the frits used in recent DWPF processing (e.g., Frits 418 and 510 each contain 8 wt % Li_2O). The concentration of Na_2O in the candidate frits is relatively low (e.g., Frit 418 contains 8 wt % Na_2O, and Frit 510 contains 9 wt % Na_2O).

Table 5-1. Candidate frits for SB5 down-selected from the Nominal Stage results.

SB5 Composition Projection	Frit ID	B_2O_3 (wt %)	CaO (wt %)	Li_2O (wt %)	MgO (wt %)	Na_2O (wt %)	SiO_2 (wt %)
Case B	B-1	10	2	12	0	2	74
	B-2	8	0	10	0	4	78
	B-3	9	0	12	0	3	76
Case C	C-1	8	2	10	0	5	75
	C-2	8	0	10	0	5	76
	C-3	8	0	9	0	7	76
Case F	F-1	10	0	12	0	3	75
	F-2	11	2	11	0	3	73
	F-3	8	0	11	0	5	76

A summary of the Nominal Stage assessment for the three candidate frits identified for SB5 Case B is given in Table 5-2. The available operating windows range from 15 to 18 percentage points in terms of available WLs. The minimum WL was limited for Frits B-2 and B-3 by the homogeneity constraint (at the Property Acceptability Region). The maximum WL was limited by liquidus temperature predictions. The liquidus temperature constraint relates to the DWPF process and does not necessarily affect waste form performance.

Table 5-2. Summary of Nominal Stage results for SB5 Case B with the candidate frits.

	B-1	B-2	B-3
Operating Window (% WL)	25-43	27-43	27-42
Lower Limiting Constraint(s)	none	Homg	Homg
Upper Limiting Constraint(s)	TL	TL	TL

A summary of the Nominal Stage assessment for the three candidate frits identified for SB5 Case C is given in Table 5-3. The available operating windows range from 15 to 17 percentage points in terms of available WLs. The minimum WL was limited by the homogeneity constraint for Frits C-2 and C-3. The maximum WL was limited by liquidus temperature predictions. The avoidance of nepheline as a limiting constraint for SB5 Case C is due to the reduced concentrations of Al_2O_3 and Na_2O in this composition projection, as well as the increased SiO_2 concentration. This could be considered beneficial for DWPF processing since the constraint limiting the upper WL for these frits combined with SB5 Case C is process related, rather than waste form affecting.

Table 5-3. Summary of Nominal Stage results for SB5 Case C with the candidate frits.

	C-1	C-2	C-3
Operating Window (% WL)	25-42	28-43	28-43
Lower Limiting Constraint(s)	none	Homg	Homg
Upper Limiting Constraint(s)	TL	TL	TL

A summary of the Nominal Stage assessment for the three candidate frits identified for SB5 Case F is given in Table 5-4. The available operating windows range from 14 to 16 percentage points in terms of available WLs. The minimum WL was limited by the homogeneity constraint for Frits F-1 and F-3. The maximum WL was limited by liquidus temperature predictions, as well as predictions of nepheline crystallization for Frit F-3. Again, the liquidus temperature constraint relates to the DWPF process and does not necessarily affect waste form performance. However, nepheline formation can reduce the durability of the glass product and is of greater concern.

Table 5-4. Summary of Nominal Stage results for SB5 Case F with the candidate frits.

	F-1	F-2	F-3
Operating Window (% WL)	28-42	25-41	28-44
Lower Limiting Constraint(s)	Homg	none	Homg
Upper Limiting Constraint(s)	TL	TL	TL, Neph

Overall, the Nominal Stage results for the three SB5 compositions with their respective candidate frits are quite similar. The operating windows are relatively wide, and are mostly limited by process related constraints, with only one frit/sludge combination being limited by a waste form affecting constraint. The complete results of the Nominal Stage assessment for the candidate frits combined with SB5 Cases B, C and F are given in Tables A1, A2 and A3, respectively, in Appendix A.

6.0 Variation Stage Assessments

The focus of the Variation Stage assessments is to evaluate the performance of a small number of candidate frits when the anticipated compositional variation is applied to the sludge systems of interest. Variation was applied to the components of each projection based on their concentrations.[a] For the major components – Al_2O_3, Fe_2O_3, Na_2O and U_3O_8 – a variation of 7.5 % of each component's concentration was applied. Other important components with lower concentrations were treated individually. A variation of 0.25 wt % was applied to CaO, MgO, MnO and NiO. A variation of 0.1 wt % was applied to SO_4^{2-} and a variation of 0.5 wt % was applied to SiO_2. The remaining sludge components were grouped into a category called 'Others'. A variation of 0.25 wt % was applied to the total concentration of the 'Others' components. The compositions of SB5 Cases B, C and F with the variation applied are given in Table 6-1.

Table 6-1. Compositions of SB5 Cases B, C and F with variation applied.

Component	Variation	SB5 Case B		SB5 Case C		SB5 Case F	
		Min. (wt %)	Max. (wt %)	Min. (wt %)	Max. (wt %)	Min. (wt %)	Max. (wt %)
Al_2O_3	7.5 %	26.787	31.131	23.014	26.746	25.133	29.209
CaO	0.25 wt %	1.531	2.031	1.714	02.214	1.632	2.132
Fe_2O_3	7.5 %	21.492	24.977	23.706	27.550	22.641	26.313
MgO	0.25 wt %	0.927	1.427	1.048	1.548	0.989	1.489
MnO	0.25 wt %	4.608	5.108	5.109	5.609	4.872	5.372
Na_2O	7.5 %	22.959	26.682	22.367	25.994	22.403	26.036
NiO	0.25 wt %	2.479	2.979	2.759	3.259	2.612	3.112
SO_4^{2-}	0.1 wt %	0.629	0.829	0.661	0.861	0.646	0.846
SiO_2	0.5 wt %	1.385	2.385	1.580	2.580	1.469	2.469
U_3O_8	7.5 %	6.875	7.990	7.583	8.813	7.212	8.382
Others	0.25 wt %	1.159	1.659	1.299	1.799	2.266	2.766

Statistical mixture experimental design methods were used to obtain an initial set of feasible sludge compositions based on the variation applied to SB5 Cases B, C and F. These methods included algorithms that were used to determine the extreme vertices (EVs) of the sludge region (the bounding compositions) for each case. After the EVs were determined for each sludge region, the Variation Stage assessments were made over the same waste loading interval (25 to 60 wt %) using the DWPF PCCS models. Acceptable predicted properties for this assessment were based on satisfying the more restrictive Measurement Acceptability Region (MAR) limits of PCCS – consistent with the Nominal Stage assessment. All MAR constraints were based on the current PCCS limits.[2]

[a] The amount of compositional variation applied to each individual component of a projected sludge batch composition has been refined by SRNL through frit development efforts for Sludge Batches 3 and 4. Based on the success of these prior Variation Stage assessments in guiding optimal frit selection, the same amount of variation was applied to the SB5 projections in this study.

A summary of the Variation Stage results for SB5 Case B with its three candidate frits is given in Table 6-2. The operating windows indicate regions where all of the EVs satisfied the MAR criteria when combined with the given frit at the indicated WL. As is typically the case, the projected operating windows are reduced as compared to the Nominal Stage assessment. The operating windows for SB5 Case B range from 6 to 14 percentage points. The minimum WLs are limited by the homogeneity and high viscosity constraints. The maximum WLs are limited by the liquidus temperature and low viscosity constraints.

Table 6-2. Summary of Variation Stage results for SB5 Case B with the candidate frits.

	Frit B-1	Frit B-2	Frit B-3
Operating Window (% WL)	26-40	29-41	33-39
Lower Limiting Constraint(s)	Homg	Homg	highv
Upper Limiting Constraint(s)	TL, lowv	TL	TL

A summary of the Variation Stage results for SB5 Case C with its three candidate frits is given in Table 6-3. The widths of the operating windows range from 8 to 12 percentage points. The minimum WLs are limited by the homogeneity constraint. The maximum WLs are limited by the liquidus temperature constraint for Frits C-1 and C-3, and by the low viscosity constraint for Frit C-2.

Table 6-3. Summary of Variation Stage results for SB5 Case C with the candidate frits.

	Frit C-1	Frit C-2	Frit C-3
Operating Window (% WL)	27-39	31-39	31-41
Lower Limiting Constraint(s)	Homg	Homg	Homg
Upper Limiting Constraint(s)	TL	lowv	TL

A summary of the Variation Stage results for SB5 Case F with its three candidate frits is given in Table 6-4. The widths of the operating windows range from 8 to 13 percentage points. The minimum WLs are limited by the homogeneity constraint. The maximum WLs are limited by the low viscosity constraint, as well as the liquidus temperature constraint for Frits F-2 and F-3.

Table 6-4. Summary of Variation Stage results for SB5 Case F with the candidate frits.

	Frit F-1	Frit F-2	Frit F-3
Operating Window (% WL)	30-38	26-39	30-41
Lower Limiting Constraint(s)	Homg	Homg	Homg
Upper Limiting Constraint(s)	lowv	TL, lowv	TL, lowv

The complete Variation Stage results for SB5 Cases B, C and F are included in Tables A4, A5 and A6, respectively, in Appendix A. The results indicate that there is a reduction in the operating window width for each frit/sludge combination. This response is typical when variation is applied to a sludge composition projection and the magnitude of the reduction is consistent with previous studies.[14, 15, 18, 19] For each SB5 projection, there are candidate frits available that appear to be sufficiently robust to variation in sludge composition. The frits continue to provide adequate operating windows (assuming DWPF will process SB5 at a target WL of 34-38 wt %) that are limited by process related – rather than waste form affecting – constraints.

By relaxing some of the criteria used earlier when down-selecting frit compositions from the Nominal Stage assessment results, it may be possible to identify other frit compositions with which the Variation Stage assessment would predict wider operating windows. However, adjusting these criteria may lead to frit/sludge systems that are limited by waste form affecting constraints.

It is important to note that these paper study results do not include any predictions of melt rate performance. Experimental studies are used to provide melt rate data to aid in any frit recommendation decisions.

7.0 Summary

Composition projections for SB5 were developed, based on a modeling approach at SRNL, to evaluate possible impacts of the Al-dissolution process on the availability of viable frit compositions for vitrification at the DWPF. The study included two projected SB5 compositions that bound potential outcomes (or degrees of effectiveness) of the Al-dissolution process, as well as a nominal SB5 composition projection based on the results of the recent Al-dissolution demonstration at SRNL. The three SB5 projections were the focus of a two-stage paper study assessment.

A Nominal Stage assessment combined each of the SB5 composition projections with an array of 19,305 frit compositions over a wide range of WL values and evaluated them against the DWPF process control models. The Nominal Stage results allowed for the down-selection of a small number of frits that provided reasonable projected operating windows (typically 27 to 42 wt % WL). The frit/sludge systems were mostly limited by process related constraints, with only one system being limited by predictions of nepheline crystallization, a waste form affecting constraint. The criteria applied in selecting the frit compositions somewhat restricted the compositional flexibility of the candidate frits for each individual SB5 composition projection, which may limit the ability to further tailor the frit for improved melt rate.

Variation Stage assessments were then performed using the down-selected frits and the three SB5 composition projections with variation applied to each sludge component. The Variation Stage results showed that the operating windows were reduced in width, as expected when variation in the sludge composition is applied. However, several of the down-selected frits exhibited a relatively high degree of robustness to the applied sludge variation, providing WL windows of approximately 30 to 39 wt %. The maximum WLs were limited by processing constraints, liquidus temperature and low viscosity, rather than a waste form affecting constraint (e.g., nepheline crystallization) in the Variation Stage assessments.

These paper study assessments have identified candidate frits which, when combined with the current, projected SB5 compositions after Al-dissolution, have projected operating windows that should be reasonable for DWPF processing. Changes in the SB5 composition are anticipated as the data on the actual Al-dissolution effectiveness and Tank 7 transfer mass become available and, will require additional paper study assessments as well as experimental frit development studies. The frits identified in this study provide insight into potential processing windows but are not the recommended frits for SB5 vitrification in DWPF. No information regarding melt rate can be inferred from the paper study results. Experimental studies to evaluate this critical factor in DWPF processing will need to be performed before a frit recommendation could be made for any projected sludge composition.

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Appendix A

Complete Results for the Nominal and Variation Stage Assessments

Table A1. Complete Nominal Stage results for Sludge Case B with Frits B-1, B-2 and B-3.

WL	Frit B-1	Frit B-2	Frit B-3
25		Homg hFrit	highv Homg hFrit
26		Homg	highv Homg
27			
28			
29			
30			
31			
32			
33			
34			
35			
36			
37			
38			
39			
40			
41			
42			
43			TL
44	TL	TL	TL
45	TL Neph	TL Neph	TL Neph
46	TL lowv Neph	TL Neph	TL Neph
47	TL lowv Neph	TL Neph	TL Neph
48	TL lowv Neph	TL Neph	TL Neph
49	TL lowv Neph	TL Neph	TL Neph
50	TL lowv Neph	TL Neph	TL Neph
51	TL lowv Neph	TL Neph	TL Neph
52	TL lowv Neph	TL Neph	TL Neph
53	TL lowv Neph	TL Neph	TL Neph
54	TL lowv Neph	TL Neph	TL Neph
55	TL lowv SO4 Neph	TL lowv SO4 Neph	TL SO4 Neph
56	TL lowv SO4 Neph	TL lowv SO4 Neph	TL SO4 Neph
57	TL lowv SO4 Neph	TL lowv SO4 Neph	TL lowv SO4 Neph
58	TL lowv SO4 Neph	TL lowv SO4 Neph	TL lowv SO4 Neph
59	TL lowv SO4 Neph	TL lowv SO4 Neph	TL lowv SO4 Neph
60	TL lowv SO4 Neph	TL lowv SO4 Neph	TL lowv SO4 Neph

Table A2. Complete Nominal Stage results for Sludge Case C with Frits C-1, C-2 and C-3.

WL	Frit C-1	Frit C-2	Frit C-3
25		Homg hFrit	Homg hFrit
26		Homg	Homg
27		Homg	Homg
28			
29			
30			
31			
32			
33			
34			
35			
36			
37			
38			
39			
40			
41			
42			
43	TL		
44	TL	TL	TL
45	TL Neph	TL lowv	TL Neph
46	TL Neph	TL lowv Neph	TL Neph
47	TL lowv Neph	TL lowv Neph	TL Neph
48	TL lowv Neph	TL lowv Neph	TL lowv Neph
49	TL lowv Neph	TL lowv Neph	TL lowv Neph
50	TL lowv Neph	TL lowv Neph	TL lowv Neph
51	TL lowv Neph	TL lowv Neph	TL lowv Neph
52	TL lowv Neph	TL lowv Neph	TL lowv Neph
53	TL lowv SO4 Neph	TL lowv SO4 Neph	TL lowv SO4 Neph
54	TL lowv SO4 Neph	TL lowv SO4 Neph	TL lowv SO4 Neph
55	TL lowv SO4 Neph	TL lowv SO4 Neph	TL lowv SO4 Neph
56	TL lowv SO4 Neph	TL lowv SO4 Neph	TL lowv SO4 Neph
57	TL lowv SO4 Cr2O3 Neph	TL lowv SO4 Cr2O3 Neph	TL lowv SO4 Cr2O3 Neph
58	TL lowv SO4 Cr2O3 Neph	TL lowv SO4 Cr2O3 Neph	TL lowv SO4 Cr2O3 Neph
59	TL lowv SO4 Cr2O3 Neph	TL lowv SO4 Cr2O3 Neph	TL lowv SO4 Cr2O3 Neph
60	TL lowv SO4 Cr2O3 Neph	TL lowv SO4 Cr2O3 Neph	TL lowv SO4 Cr2O3 Neph

Table A3. Complete Nominal Stage results for Sludge Case F with Frits F-1, F-2 and F-3.

WL	Frit F-1	Frit F-2	Frit F-3
25	Homg hFrit		Homg hFrit
26	Homg		Homg
27	Homg		Homg
28			
29			
30			
31			
32			
33			
34			
35			
36			
37			
38			
39			
40			
41			
42		TL	
43	TL	TL	
44	TL lowv	TL	
45	TL lowv Neph	TL lowv Neph	TL Neph
46	TL lowv Neph	TL lowv Neph	TL Neph
47	TL lowv Neph	TL lowv Neph	TL lowv Neph
48	TL lowv Neph	TL lowv Neph	TL lowv Neph
49	TL lowv Neph	TL lowv Neph	TL lowv Neph
50	TL lowv Neph	TL lowv Neph	TL lowv Neph
51	TL lowv Neph	TL lowv Neph	TL lowv Neph
52	TL lowv Neph	TL lowv Neph	TL lowv Neph
53	TL lowv Neph	TL lowv Neph	TL lowv Neph
54	TL lowv SO4 Neph	TL lowv SO4 Neph	TL lowv SO4 Neph
55	TL lowv SO4 Neph	TL lowv SO4 Neph	TL lowv SO4 Neph
56	TL lowv SO4 Neph	TL lowv SO4 Neph	TL lowv SO4 Neph
57	TL lowv SO4 Neph	TL lowv SO4 Neph	TL lowv SO4 Neph
58	TL lowv SO4 Neph	TL lowv SO4 Neph	TL lowv SO4 Neph
59	TL lowv SO4 Neph	TL lowv SO4 Neph	TL lowv SO4 Neph
60	TL lowv SO4 Cr2O3 Neph	TL lowv SO4 Cr2O3 Neph	TL lowv SO4 Cr2O3 Neph

Table A4. Results of the Variation Stage assessment for SB5 Case B with the candidate frits.

WL	Frit B-1		Frit B-2		Frit B-3	
	Percent of EVs	Limiting Constraint(s)	Percent of EVs	Limiting Constraint(s)	Percent of EVs	Limiting Constraint(s)
25	99.6	Homg	19.4	Homg hFrit highv	0.0	Homg hFrit highv
26	100.0		36.1	Homg hFrit	0.8	Homg hFrit highv
27	100.0		83.3	Homg	38.2	Homg highv
28	100.0		97.7	Homg	57.0	Homg highv
29	100.0		100.0		98.1	highv
30	100.0		100.0		83.8	highv
31	100.0		100.0		92.8	highv
32	100.0		100.0		97.9	highv
33	100.0		100.0		100.0	
34	100.0		100.0		100.0	
35	100.0		100.0		100.0	
36	100.0		100.0		100.0	
37	100.0		100.0		100.0	
38	100.0		100.0		100.0	
39	100.0		100.0		100.0	
40	100.0		100.0		99.9	TL
41	77.0	TL lowv	100.0		95.9	TL
42	61.9	TL lowv Neph	96.7	TL	88.4	TL
43	27.4	TL lowv Neph	74.1	TL Neph	62.1	TL Neph
44	5.0	TL lowv Neph	45.0	TL Neph	36.3	TL Neph
45	0.0	TL lowv Neph	3.0	TL Neph	1.7	TL Neph
46	0.0	TL lowv Neph	0.0	TL Neph	0.0	TL Neph
47	0.0	TL lowv Neph	0.0	TL Neph	0.0	TL Neph
48	0.0	TL lowv Neph	0.0	TL Neph	0.0	TL Neph
49	0.0	TL lowv Neph SO4	0.0	TL lowv SO4 Neph	0.0	TL SO4 Neph
50	0.0	TL lowv Neph SO4	0.0	TL lowv SO4 Neph	0.0	TL SO4 Neph
51	0.0	TL lowv Neph SO4	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
52	0.0	TL lowv Neph SO4	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
53	0.0	TL lowv Neph SO4	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
54	0.0	TL lowv Neph SO4	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
55	0.0	TL lowv Neph SO4	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
56	0.0	TL lowv Neph SO4	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
57	0.0	TL lowv Neph SO4	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
58	0.0	TL lowv Neph SO4	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
59	0.0	TL lowv Neph SO4	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
60	0.0	TL lowv Neph SO4	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph

Table A5. Results of the Variation Stage assessment for SB5 Case C with the candidate frits.

WL	Frit C-1		Frit C-2		Frit C-3	
	Percent of EVs	Limiting Constraint(s)	Percent of EVs	Limiting Constraint(s)	Percent of EVs	Limiting Constraint(s)
25	88.5	Homg	0.0	Homg hFrit	0.0	Homg hFrit
26	98.6	Homg	18.4	Homg	18.4	Homg
27	100.0		31.5	Homg	31.5	Homg
28	100.0		76.1	Homg	76.1	Homg
29	100.0		94.9	Homg	94.9	Homg
30	100.0		77.8	Homg	99.8	Homg
31	100.0		100.0		100.0	
32	100.0		100.0		100.0	
33	100.0		100.0		100.0	
34	100.0		100.0		100.0	
35	100.0		100.0		100.0	
36	100.0		100.0		100.0	
37	100.0		100.0		100.0	
38	100.0		100.0		100.0	
39	100.0		100.0		100.0	
40	99.4	TL	84.7	lowv	100.0	
41	94.4	TL	72.1	TL lowv	100.0	
42	71.5	TL lowv	64.2	TL lowv	95.9	TL
43	39.2	TL lowv Neph	50.1	TL lowv	47.1	TL Neph
44	6.5	TL lowv Neph	13.0	TL lowv Neph	14.5	TL lowv Neph
45	0.0	TL lowv Neph	1.1	TL lowv Neph	2.0	TL lowv Neph
46	0.0	TL lowv Neph	0.0	TL lowv Neph	0.0	TL lowv Neph
47	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
48	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
49	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
50	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
51	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
52	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
53	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
54	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
55	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
56	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
57	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
58	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
59	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
60	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph

Table A6. Results of the Variation Stage assessment for SB5 Case F with the candidate frits.

WL	Frit F-1		Frit F-2		Frit F-3	
	Percent of EVs	Limiting Constraint(s)	Percent of EVs	Limiting Constraint(s)	Percent of EVs	Limiting Constraint(s)
25	13.5	Homg hFrit	97.7	Homg	13.5	Homg hFrit
26	28.5	Homg	100.0		28.5	Homg
27	64.6	Homg	100.0		64.6	Homg
28	93.7	Homg	100.0		93.7	Homg
29	99.6	Homg	100.0		99.6	Homg
30	100.0		100.0		100.0	
31	100.0		100.0		100.0	
32	100.0		100.0		100.0	
33	100.0		100.0		100.0	
34	100.0		100.0		100.0	
35	100.0		100.0		100.0	
36	100.0		100.0		100.0	
37	100.0		100.0		100.0	
38	100.0		100.0		100.0	
39	85.7	lowv	100.0		100.0	
40	71.5	lowv	75.9	TL lowv	100.0	
41	65.7	TL lowv	57.3	TL lowv	100.0	
42	56.2	TL lowv	44.4	TL lowv	79.6	TL lowv
43	38.1	TL lowv	11.2	TL lowv Neph	40.3	TL lowv
44	8.1	TL lowv Neph	0.2	TL lowv Neph	11.6	TL lowv Neph
45	0.0	TL lowv Neph	0.0	TL lowv Neph	0.5	TL lowv Neph
46	0.0	TL lowv Neph	0.0	TL lowv Neph	0.0	TL lowv Neph
47	0.0	TL lowv Neph	0.0	TL lowv Neph	0.0	TL lowv Neph
48	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
49	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
50	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
51	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
52	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
53	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
54	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
55	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
56	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
57	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
58	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
59	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph
60	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph	0.0	TL lowv SO4 Neph

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