Summary of Results for PHA Glass Study: Composition and Property Measurements

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Summary of Results for PHA Glass Study: Composition and Property Measurements (U)

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SUMMARY AND CONCLUSIONS

This report provides a summary of the results obtained for a limited variability study for glasses containing Precipitate Hydrolysis Aqueous (PHA), Monosodiumtitanate (MST), and either simulated Purex or HM sludge. Twenty glasses containing Purex sludge and three glasses containing HM sludge were fabricated and tested. The fabricated glasses were tested for durability using the Product Consistency Test (PCT) and characterized by measuring the melt viscosity and an approximate, bounding liquidus temperature. The current models used by Defense Waste Processing Facility (DWPF) for predicting durability, viscosity, and liquidus temperature were applied to all 23 glasses. The goal of this work was to identify any major problems from a glass perspective that could potentially preclude the use of higher levels of PHA and MST at DWPF.

The selection of Purex sludge for the majority of the glasses fabricated in this study was based on the knowledge that this sludge type has historically been the most difficult sludge to incorporate into glass. Depleted uranium was introduced into the simulated Purex sludge to better represent the uranium content (~9 wt% oxide) of actual sludge.

One of the major concerns for this study was the effect of higher levels of PHA in the glass. The plan for this work was to vary the PHA level from 7 to 13 wt% oxide, which can be compared to the expected/design basis of 6 to 8 wt% oxide. Another concern was the potential for higher levels of titanium (from MST) into the glass. The current limit for TiO_2 in the DWPF glass is 1 wt% whereas the scope of this work varied the TiO_2 level from 1 to 2 wt%.

Frit 202 was used in this study and was the frit originally designed for coupled operations at DWPF.

As part of this study, the model predictions were made using targeted, measured, and bias-corrected measured compositions of the glasses. For glasses where there were no batching difficulties, it was demonstrated that the results were essentially insensitive to the type of composition used in these models. This provides evidence that the glasses produced were close to the targeted compositions, and that the analytical measurements were of high quality.

Glass Durability. All 23 glasses were very durable as measured by the PCT. The values ranged from 0.90 to 1.76 g/L for boron release for the Purex glasses and from 0.45 to 0.50 g/L for boron release for the HM glasses. For comparison, the reference Environmental Assessment (EA) glass has a boron rate of 16.7 g/L.

Model Prediction of Durability. The PCCS durability model predicted values for boron release that were all within the 95% prediction limits of the model.

Homogeneity. The durability model was developed for glasses that are not phase separated (amorphous) and this is controlled through application of a homogeneity discriminator. The majority of the glasses (17 of the 23 glasses) failed this discrimination model. The PCT results for the glasses of this study (all of which were rapidly quenched) revealed that measured durability is not a function of predicted phase separation based upon the current homogeneity model. Further work (including kinetic studies) is required to resolve this apparent inconsistency.

Liquidus. For this study, the liquidus temperature was bounded by performing 24-hour isothermal holds (as required) for the glass melts at 900°C, 950°C, 1000°C, and 1050°C. X-ray diffraction was used to detect crystallization, in this case Trevorite. For the 22 wt% and 26 wt% Purex glasses, no crystals were detected in the bulk at 900°C or at the top surface of the glasses. For the 30 wt% Purex glasses, crystals were evident at temperatures higher than 900°C, but at 1000°C they were below the XRD detection limit. Given the fact that liquidus temperatures were only approximated, the 30 wt% loading of Purex may be near or at the edge of acceptability for liquidus. A small amount of surface crystallization (after the isothermal hold at 900°C) was evident on the glass surface near the center of

the crucible for four of the 30 wt% Purex glasses. The crystals detected were Trevorite. For HM glasses, no crystals were detected in the bulk or on the surface after 24 hours at 900°C.

Viscosity. The melt viscosities for 11 of these 23 glasses were measured and predictions reported at 1150°C (nominal temperature of the glass within the DWPF melter). For the Purex containing glasses, the measured viscosities were within the DWPF range of 20 to 100 poise for the 22 wt% and 26 wt% Purex glasses. However, the 30 wt% Purex glasses had viscosities that were very near the lower limit of 20 poise, and consequently, may not be acceptable from an operations perspective. Although the HM sludge glass measured (10wt% PHA) had a viscosity of ~90 poise, the HM glasses at 7wt% PHA are predicted to be higher than the 100 poise limit for DWPF.

Limitations. All of the conclusions are provided based on the scope of the current work. One limitation of this scope, based upon schedule and budget, was the absence of any investigation of kinetic effects. Thus, one can not rule out that amorphous phase separation occurs with centerline cooling, for example. A second limitation was the restriction on independent variation of chemical constituents. In a major variability study, ranges are established for each element, and a statistically designed set of glasses identified which not only covers a larger region of compositional space, but also provides the potential for revealing trends in the properties over linear variations of elemental concentrations. A third limitation was that only approximate measurements of the liquidus temperatures were made. A fourth limitation was that a thorough search (beyond scanning electron microscopy) for phase separation was not conducted. This type of investigation requires considerable efforts using transmission electron microscopy (TEM) and other high resolution techniques. Finally, although the strategy was that HM and Purex containing glasses would cover the extremes, no Blend sludge glasses were fabricated to verify this.

INTRODUCTION

One of the Alternative Salt Disposition Flowsheets being considered would require that the Defense Waste Processing Facility (DWPF) vitrify a coupled feed containing High Level Waste (HLW) and Precipitate Hydrolysis Aqueous (PHA). A Technical Task Request (TTR) [1] was received by the Savannah River Technology Center (SRTC) requesting that a glass variability study be conducted to explore the processability and product quality of the glass composition region for this alternative (Small Tank TPB Precipitation) to the In-Tank Precipitation (ITP) Process. A Task Technical and Quality Assurance (TT&QA) plan [2] was issued by SRTC in response to the TTR. The objective of this task was to obtain information on the feasibility of incorporating anticipated levels of PHA into DWPF glass with and without doubling the nominal levels of monosodium titanate (MST). The study progressed through four phases of investigation, the detailed results of which have each been issued in technical reports (see references [3] through [6]). The purpose of this final report is to consolidate the results of these four phases and provide overall conclusions.

Table 1 provides a general overview of the glass compositions that were batched, fabricated, and tested as part of this study. The glasses were selected from a set of candidate glasses that involved three sludge types: Purex, HM, and Blend; covered sludge loadings (in the glass) of 22, 26, and 30 oxide weight percent (wt%); utilized PHA loadings (in the glass) of 7, 10, and 13 oxide wt%; and included MST concentrations (in the glass) at 1.25 and 2.5 wt%. For each composition, the remainder of the glass consisted of Frit 202. The goal of using Purex sludge as the major sludge type with only a few glasses containing HM was realized. However, no glasses were fabricated using the Blend sludge (a sludge representing the combination of HM and Purex). In all likelihood, DWPF macrobatches will be blends of tanks containing various waste/sludge types.

Table 1: General Compositions of the PHA Glasses

Phase	Glass ID	Sludge Type	Sludge Loading	PHA	MST	Frit 202
1	pha07	Purex	26%	7%	1.25%	65.75%
1	pha08	Purex	26%	10%	1.25%	62.75%
1	pha09	Purex	26%	13%	1.25%	59.75%
1	pha10	Purex	26%	7%	2.50%	64.50%
1	pha11	Purex	26%	10%	2.50%	61.50%
1	pha12	Purex	26%	13%	2.50%	58.50%
2	pha13	Purex	30%	7%	1.25%	61.75%
2	pha14	Purex	30%	10%	1.25%	58.75%
2	pha15	Purex	30%	13%	1.25%	55.75%
2	pha16	Purex	30%	7%	2.50%	60.50%
2	pha17	Purex	30%	10%	2.50%	57.50%
2	pha18	Purex	30%	13%	2.50%	54.50%
3	pha01	Purex	22%	7%	1.25%	69.75%
3	pha02	Purex	22%	10%	1.25%	66.75%
3	pha03	Purex	22%	13%	1.25%	63.75%
3	pha04	Purex	22%	7%	2.50%	68.50%
3	pha05	Purex	22%	10%	2.50%	65.50%
3	pha06	Purex	22%	13%	2.50%	62.50%
4	pha11c	Purex	26%	10%	2.50%	61.50%
4	pha12c	Purex	26%	13%	2.50%	58.50%
4	pha17c	Purex	30%	10%	2.50%	57.50%
4	pha18c	Purex	30%	13%	2.50%	54.50%
4	pha20	HM	22%	10%	1.25%	66.75%
4	pha26	HM	26%	10%	1.25%	62.75%
4	pha32	HM	30%	10%	1.25%	58.75%

The primary property of interest in this study was the durability (as measured by the 7-day Product Consistency Test, PCT [7]) of the test glasses. The PCT is the recognized standard for determining the durability of vitrified HLW, and the Environmental Assessment (EA) glass is the reference standard for assessing acceptable durability determined using the PCT.¹ The measurement of durability was conducted to the specifications of the PCT Method A of ASTM C1285 for each and every glass of this study. (See the analytical plan [8] and the individual phase reports, [3] through [6], for details.)

Processing properties of interest for these glasses included viscosity at 1150°C and liquidus temperature. Methods used to complete these two types of measurements did not follow standard ASTM procedures. However, viscosity was measured at SRTC using a Harrop viscometer [9] that has yielded good results [10]. The standard ASTM procedure for measuring liquidus temperature, T_L , uses a gradient furnace. Since no such capability exists at SRTC, it was beyond the scope of this study to pursue this process for obtaining T_L measurements. To gain insight into this important processability property for the PHA glasses, an attempt was made to obtain an upper bound on the T_L 's using isothermal holds at 900, 950, 1000, and 1050°C and (non-quantitative) XRD evaluations (with a sensitivity of ~ 0.7 to 1 wt% for crystalline Trevorite in the glass). The results from these measurements are presented in this final report (the reports for the individual phases ([3], [4], [5], and [6]) provide greater detail).

In the report for each phase of this study, comparisons were provided between

- the measured and target compositions of the test glasses and
- the property measurements and their predictions derived from models that are used by the Defense Waste Processing Facility (DWPF) as part of its Product Composition Control System (PCCS). These models relate processability and product quality (e.g., viscosity, T_L , and durability) to glass composition.

These comparisons have been consolidated and provided in the sections that follow.

¹ For a glass to demonstrate acceptable durability its PCT leach rate must be 2-sigma better than the PCT leach rate of the EA glass.

RESULTS AND DISCUSSION

The composition and property measurements of the glasses comprising the four phases of the PHA study were conducted in parallel with glasses from the corresponding phases of the other ITP replacement alternative, Crystalline Silicotitanate. This second study was designated as the CST study (references [11], [12], [13], and [14] provide the reports for the four phases of this study). Conducting the two studies in parallel helped to ensure that the CST and PHA glasses were fabricated, characterized, and analyzed under very similar conditions. For a full description of the analytical plans or of the review of the measurement data, see the individual reports for the phases of the PHA study [3]-[6].

Target and Measured Chemical Compositions

Table A.1 in the Appendix provides the target oxide compositions for each of the PHA glasses prepared using Purex sludge. Table A.2 in the Appendix provides the target oxide compositions for the three PHA glasses prepared using HM sludge.

Measurements of the compositions of the test glasses were conducted by the SRTC-Mobile Laboratory (SRTC-ML) or by the Analytical Development Section (ADS) of SRTC. Standards were included with the test glasses and bias-corrections were conducted for many of these measurements. Details are provided in the individual phase reports. The target, measured, and bias-corrected compositions of the glasses tested as part of this study are provided in Table A.3 of the Appendix. In most instances, the property predictions were essentially insensitive to the way the compositions of these glasses were represented (i.e., whether by the targeted, measured, or bias-corrected measurements). In certain instances discrepancies between target and measured compositions were investigated to gain insight into the batching and/or targeting processes.

One such discrepancy found during Phase 2 (glasses pha13-pha18) involving the components of PHA suggested a batching error centered on the use of anhydrous sodium borate versus hydrated sodium borate. Although the problem existed in all six Phase 2 glasses, those at the lowest levels of PHA (i.e., pha13 and pha16) were least affected. The other four glasses were re-batched as part of Phase 4 (i.e., glasses pha14c, pha15c, pha17c, and pha18c) of this study. Table A.3 contains the results for all of the PHA glasses, but the original glasses: pha14, pha15, pha17, and pha18 are excluded from the other analyses presented in this summary report. For full discussion of this discrepancy see reference [4], and for a discussion of the re-batched results see reference [6]. Exhibit A.1 in the Appendix provides plots of the compositions (target, measured, and bias corrected) of these glasses by oxide.

Another discrepancy, discovered during Phases 1 and 2, was the factor used to account for the MST components in the final glass product. This problem revealed itself in compositions that failed to meet the desired TiO_2 targets and led to a re-evaluation of MST, a source of TiO_2 in the glass. A subsequent analysis of MST revealed a larger than expected moisture content. The problem was corrected for Phase 3, and additional glasses (pha11c, pha12c, pha17c, and pha18c designated in Phase 4 with a "c" suffix, see Table 1) from the first two phases were introduced into Phase 4. The TiO_2 levels were met for Phases 3 and 4.

An additional comment about this exhibit and table is worth mentioning. The sum of oxides for all of these glasses fell within the interval of 95 to 105 wt%, a measure of the quality of the analytical results.

PCT Results

All of the PHA glasses, after being batched and fabricated, were subjected to the 7-day Product Consistency Test (PCT) as an assessment of their durabilities [7]. More specifically, Method A of PCT (ASTM C1285) was used for these measurements. Durability is the critical product quality metric for vitrified nuclear waste. The PCT responses (for four elements: boron, silicon, sodium, and lithium) were normalized to the units of grams-per-liter (g/L) using the measured, measured bias-corrected, and target compositions. In addition to the PHA test glasses, the Environmental Assessment (EA) glass and the ARM glass were subjected to the PCT. The individual phase reports provide the details of the analytical plan and resulting data supporting these tests. The results from these tests are summarized in Table A.4 of the Appendix.

As seen in Table A.4, the durabilities for the PHA glasses are much better than those of EA samples. Figure 1 provides an opportunity for a closer look at these results using measured and bias-corrected compositions. Figure 1 is a plot of the DWPF model that relates the common logarithm of the normalized PCT (in this case for B) to a linear function of a free energy of hydration term (ΔG_p , kcal/100g glass) derived from the glass (measured and bias-corrected) compositions [15]. Prediction limits (at 95% confidence) for an individual (new) PCT result are also plotted around this linear fit. The PCT results for EA (shown as a diamond), ARM (shown as a "z"), and the PHA glasses (each shown as an "x") are presented on this plot. Note that the PHA results reveal acceptable and predictable PCTs. Figure 2 provides a plot of the boron results based upon target compositions, which shows a similar pattern of behavior. Exhibit A2 in the Appendix provides similar plots of the PHA durability measurements versus the DWPF durability models for B, Si, Na, and Li. The behaviors seen in the plots for Si, Na, and Li are similar to that demonstrated by the B results: acceptable and predictable durabilities.

Figure 1.
Log NL(B) (g/L) By ΔG_p
(Using PHA Measured & Bias-corrected
& EA and ARM reference compositions)

Figure 2.
Log NL(B) (g/L) By ΔG_p
(Using target compositions for the PHA glasses
& EA and ARM reference compositions)

Viscosity at 1150°C

Viscosity measurements were made on several of these PHA glasses at SRTC using a Harrop, high-temperature viscometer [9]. The viscosity (in Poise) of each of these glasses at 1150°C was estimated from a Fulcher equation fitted to a set of viscosity measurements taken over an appropriate range of temperatures. The functional form of the (three-parameter) Fulcher equation (expressed in Poise) used to fit these data is given by equation (1):

$$\ln \hat{\eta} = A + \frac{B}{(T - C)} \quad (1)$$

where A, B, and C represent the parameters of the model that were determined from the available measurements (represented by η , expressed in Poise) at various temperatures (represented by T). The fitted model was then used to predict the viscosity of the given glass at 1150°C.

Although no definitive error analysis has been completed on the use of the Harrop viscometer, SRTC has conducted several sets of viscosity measurements using this viscometer with good results [10]. Viscosity measurements were not conducted for all of these PHA glasses. Table 2 provides the results that were obtained as part of this study.

Table 2: Viscosity Results (in Poise) By Glass ID for the PHA Glasses

Glass ID	Viscosity (Poise) @ 1150 °C	Predicted (measured composition)	Predicted (bias-corrected composition)	Predicted (target composition)
pha01	43.9	57.7	57.7	70.2
pha02	Not measured	56.0	60.8	53.1
pha03	33.1	39.4	42.6	39.1
pha04	Not measured	71.8	77.3	67.4
pha05	Not measured	45.8	50.1	50.6
pha06	Not measured	39.0	42.1	36.9
pha07	52.6	65.7	66.1	58.0
pha08	39.5	53.3	54.1	42.7
pha09	31.8	42.7	44.0	30.5
pha10	50.9	67.5	68.2	55.3
pha11	37.7	59.4	60.7	40.4
pha11c	Not Measured	45.6	51.2	40.4
pha12	29.2	42.1	43.4	28.5
pha12c	27.6	32.7	36.7	28.5
pha13	Not Measured	42.4	45.4	46.7
pha14c	Not Measured	31.4	N/A	33.3
pha15c	22.8	22.0	N/A	23.0
pha16	Not Measured	39.4	42.1	44.2
pha17c	Not Measured	32.7	37.1	31.3
pha18c	Not Measured	21.7	24.8	21.3
pha20	Not Measured	96.7	105.8	102.0
pha26	Not Measured	98.4	106.3	97.0
pha32	89.7	90.4	99.5	91.9

Figure 3 provides a plot of the measured viscosities versus the viscosities predicted from the DWPF model using target, measured, and bias-corrected compositions. A 45-degree (diagonal) line is also shown to provide a point of reference.

Figure 3: Viscosity Measurements versus Property Predictions

For the Purex glasses, the measured viscosities were within the DWPF range of 20 to 100 poise for the 22 wt% and 26 wt% Purex loadings. For these glasses, the measured viscosities ranged from 28 to 53 poise, values that are much lower than current operations. The measured viscosity of glass pha12c is only 28 poise and is approaching the lower limit for viscosity. The results show, as expected, that the viscosity values decrease as the PHA concentration in the glass is increased.

For the 30 wt% Purex glasses at high PHA loadings, the measured viscosities were very near the lower limit of 20 poise and, consequently, may not be acceptable from an operations perspective. Although the HM sludge glass (pha32) had a measured viscosity of ~90 poise at 10 wt% PHA, the HM glasses at 7 wt% PHA are predicted to be ~120 poise, a value that is significantly higher than the 100 poise upper limit for DWPF.

There are several interesting trends observed in these data. Whether one uses the target, measured, or bias-corrected measured compositions to predict viscosities using the current model, the predicted viscosities are almost always higher than the viscosities determined from measurements for the glasses batched using Purex sludge. This possible overprediction also occurs for the glasses made using HM sludge.

Liquidus Temperature (T_L)

The standard ASTM procedure for measuring liquidus temperature uses a gradient furnace. The equipment for determining liquidus temperature by this method is being installed and tested within SRTC in a clean laboratory. Due to the presence of depleted uranium in the glass samples (as well as the early stage of equipment setup), this method for liquidus determination was not available at SRTC for these analyses. A decision was therefore made to perform isothermal holds using reasonable quantities of the glass to bound the liquidus temperature.

XRD was selected as the method of detection for crystal formation in the glasses after an isothermal hold. It is estimated that the sensitivity of XRD (non-quantitative) is ~ 0.7 to 1 wt% for a crystalline phase (in this case, Trevorite [16]). Therefore, for this type of measurement, absence of detection of a crystalline phase was evidence that the liquidus temperature is less than the temperature of that isothermal hold. On the other hand, detection of Trevorite (or any other primary crystalline phase) indicates that the liquidus temperature is higher than the temperature of the isothermal hold. The bounds on the liquidus temperatures for these PHA glasses, estimated to the detection capabilities of XRD, are given in Table 3. The model predictions for T_L based upon target, measured, and bias-corrected compositions are also provided in this table.

Table 3: Liquidus Temperature ($^{\circ}\text{C}$) By Glass ID for the CST Glasses

Glass ID	Estimated T_L By Bounding Procedure	Predicted (measured composition)	Predicted (bias-corrected composition)	Predicted (target composition)
pha01	<900 $^{\circ}\text{C}$	954.6	950.8	949.3
pha02	<900 $^{\circ}\text{C}$	957.1	949.4	956.1
pha03	<900 $^{\circ}\text{C}$	966.5	958.4	963.6
pha04	<900 $^{\circ}\text{C}$	923.1	917.3	952.0
pha05	<900 $^{\circ}\text{C}$	967.5	959.0	959.1
pha06	<900 $^{\circ}\text{C}$	968.5	960.3	966.9
pha07	<900 $^{\circ}\text{C}$	966.8	973.2	949.3
pha08	<900 $^{\circ}\text{C}$	973.2	980.0	956.1
pha09	<900 $^{\circ}\text{C}$	969.2	975.2	963.6
pha10	<900 $^{\circ}\text{C}$	953.7	959.3	952.0
pha11	<900 $^{\circ}\text{C}$	957.0	962.7	959.1
pha11c	<900 $^{\circ}\text{C}$	987.3	977.5	1001.5
pha12	<900 $^{\circ}\text{C}$	973.1	979.2	966.9
pha12c	<900 $^{\circ}\text{C}$	997.5	987.1	1012.2
pha13	<900 $^{\circ}\text{C}$	1040.7	1033.4	1032.9
pha14c	<1000 $^{\circ}\text{C}$	1059.8	N/A	1045.4
pha15c	<1000 $^{\circ}\text{C}$	1073.9	N/A	1059.3
pha16	<900 $^{\circ}\text{C}$	1039.7	1032.7	1037.9
pha17c	<1000 $^{\circ}\text{C}$	1050.9	1037.6	1051.0
pha18c	<950 $^{\circ}\text{C}$	1073.8	1059.2	1065.7
pha20	<900 $^{\circ}\text{C}$	916.2	910.2	906.3
pha26	<900 $^{\circ}\text{C}$	928.3	922.3	935.6
pha32	<900 $^{\circ}\text{C}$	959.2	950.8	970.7

Figure 4 provides a comparison between the T_L predictions of Table 12 and the corresponding bounding measurements. A 45-degree (diagonal) line is also shown to provide a point of reference.

Figure 4: T_L Measurements versus Property Predictions

For the 22 wt% and 26 wt% Purex glasses, no crystals were detected in the bulk of the glasses at 900°C. For the 30 wt% Purex glasses, crystals were evident at temperatures higher than 900°C, but at 1000°C they were below the XRD detection limit. Given the fact that liquidus temperatures were only approximated, the 30 wt% loading of Purex may be near or at the edge of acceptability for liquidus. For HM glasses, no crystals were detected in the bulk after 24 hours at 900°C. Thus, these approximate bounding measurements of liquidus temperature suggest that the liquidus constraint (~1025°C [17]) can be met for these Purex and HM glasses.

Surface Crystallization

For liquidus measurements, crystal formation only in the interior or bulk glass region is considered. Therefore, samples submitted for XRD analysis were bulk samples. However, crystals can form at the interface of the glass and the crucible and/or the glass and air. For completeness, the detection of these surface crystals on the top of the glass is provided in Table 4 as a function of temperature.

Table 4. Surface Crystals for the PHA Glasses as a Function of Temperature

----after the 24 hour heat treatment----

	1150°C	1000°C	950°C	900°C
pha01	No test	No test	No test	None
pha02	No test	No test	No test	None
pha03	No test	No test	No test	None
pha04	No test	No test	No test	None
pha05	No test	No test	No test	None
pha06	No test	No test	No test	None
pha07	No test	No test	No test	None
pha08	No test	No test	No test	None
pha09	No test	No test	No test	None
pha10	No test	No test	No test	None
pha11	No test	No test	No test	None
pha11c	No test	No test	No test	None
pha12	No test	No test	No test	None
pha12c	No test	No test	No test	None
pha13	none	none	none	none
pha14c	No test	none	none	crystals
pha15c	No test	none	No test	crystals
pha16	none	none	none	none
pha17c	No test	none	none	crystals
pha18c	No test	No test	No test	crystals
pha20	No test	No test	No test	None
pha26	No test	No test	No test	None
pha32	No test	No test	No test	None

The crystals observed were located on the top glass surface at the center of the crucible. The chemical composition of the surface crystals was determined by EDS and is consistent with the Trevorite (NiFe_2O_4) crystal structure. Trevorite has also been observed during previous studies on these types of systems. The chemical composition indicates that other cations are substituted into the crystal structure in small amounts and these include Ti, Cr, and Mn.

Phase Separation

The formation of separate amorphous phases in glass is referred to as amorphous phase separation or inhomogeneity. Crystal formation, as determined by liquidus temperature measurements, on the other hand may indicate a "separation of phases," but reflects crystalline particles within the glass matrix. Amorphous phase separation is to be avoided since the models currently used to predict durability do not apply for glasses predicted to be phase separated. The property acceptance region, or PAR, limit for the homogeneity constraint in the Product Composition Control System (PCCS) is nominally a value of 211 [17]. The limit for the measurement acceptance region, or MAR, for this property will be even higher. In order to pass this constraint, the predicted homogeneity based upon the measured composition must be greater than the MAR value. The homogeneity values calculated using the targeted and measured chemical compositions are all well below the PAR value. These values are provided in Table 5.

Table 5: Homogeneity Property Predictions

Glass ID	Homogeneity Property Prediction based on (Acceptability Requires a Value > 211)		
	Target Composition	Measured Composition	Bias-Corrected Composition
pha01	200.1	204.2	199.9
pha02	199.8	199.1	196.9
pha03	199.4	198.6	196.6
pha04	198.4	195.7	193.9
pha05	198.0	193.3	191.3
pha06	197.7	198.7	196.6
pha07	207.9	197.1	201.3
pha08	207.5	202.9	207.2
pha09	207.2	198.3	202.4
pha10	206.1	196.3	200.4
pha11	205.8	200.5	204.7
pha11c	205.8	203.7	200.5
pha12	205.4	202.0	206.1
pha12c	205.4	205.2	202.0
pha13	215.6	215.7	217.5
pha14c	215.3	214.5	N/A
pha15c	214.9	212.6	N/A
pha16	213.9	221.7	223.0
pha17c	213.5	211.1	207.1
pha18c	213.2	218.9	214.8
pha20	201.1	199.2	197.4
pha26	209.1	206.0	203.5
pha32	217.1	208.8	206.6

Figure 5 provides a plot of the (common logarithm of the) normalized PCT response versus the homogeneity prediction based upon target, measured, and bias-corrected compositions. A vertical line on the x-axis at 211 represents the PAR value for the homogeneity property constraint. The horizontal line at ~ 1.23 represents the common logarithm of the normalized PCT response of EA glass. The reference response for EA is 16.7 g/L.

Figure 5: Homogeneity Predictions versus $\text{Log}_{10}(\text{Normalized PCT Response})$

The PCT responses of the PHA glasses made using Purex cover the a wide range of homogeneity predictions and their PCT response all fall within the interval of 0.90 g/L to 1.76g/L. The PCT responses for the three PHA glasses made using HM sludge fall within the interval of 0.45 g/L to 0.50 g/L.

The homogeneity constraint was developed for glasses that do contain PHA. Therefore, the predictability of phase separation by this model should be applicable. It turns out that 17 of the 23 PHA glasses failed to meet this homogeneity constraint. A significant search for phase separation in these glasses was beyond the scope of work for this task. Therefore, it is unclear whether phase separation has occurred, or would occur upon center-line cooling, in these glasses. On the other hand, all of the PCT results are acceptable and predictable and the results shown in Figure 5 suggest no dependence of durability on the homogeneity value for these glasses.

Further work (including kinetic studies) would be required to resolve this apparent inconsistency.

CONCLUSIONS

This report provides a summary of the results obtained for a limited variability study for glasses containing PHA, MST, and either simulated Purex or HM sludge. Twenty glasses containing Purex sludge and three glasses containing HM sludge were fabricated and tested. The fabricated glasses were tested for durability using the PCT and characterized by measuring the melt viscosity and an approximate, bounding liquidus temperature. The current models used by DWPF for predicting durability, viscosity, and liquidus temperature were applied to all 23 glasses. The goal of this work was to identify any major problems from a glass perspective that could potentially preclude the use of higher levels of PHA and MST at DWPF.

The selection of Purex sludge for the majority of the glasses fabricated in this study was based on the knowledge that this sludge type has historically been the most difficult sludge to incorporate into glass. Depleted uranium was introduced into the simulated Purex sludge to better represent the uranium content (~ 9 wt% oxide) of actual sludge.

One of the major concerns for this study was the effect of higher levels of PHA in the glass. The plan for this work was to vary the PHA level from 7 to 13 wt% oxide, which can be compared to the expected/design basis of 6 to 8 wt% oxide. Another concern was the potential for higher levels of titanium (from MST) into the glass. The current limit for TiO_2 in the DWPF glass is 1 wt% whereas the scope of this work varied the TiO_2 level from 1 to 2 wt%.

Frit 202 was used in this study and was the frit originally designed for coupled operations at DWPF.

As part of this study, the model predictions were made using targeted, measured, and bias-corrected measured compositions of the glasses. For glasses where there were no batching difficulties, it was demonstrated that the results were essentially insensitive to the type of composition used in these models. This provides evidence that the glasses produced were close to the targeted compositions, and that the analytical measurements were of high quality.

Glass Durability. All 23 glasses were very durable as measured by the PCT. The values ranged from 0.90 to 1.76 g/L for boron release for the Purex glasses and from 0.45 to 0.50 g/L for boron release for the HM glasses. For comparison, the reference Environmental Assessment (EA) glass has a boron rate of 16.7 g/L.

Model Prediction of Durability. The PCCS durability model predicted values for boron release that were all within the 95% prediction limits of the model.

Homogeneity. The durability model was developed for glasses that are not phase separated (amorphous) and this is controlled through application of a homogeneity discriminator. The majority of the glasses (17 of the 23 glasses) failed this discrimination model. The PCT results for the glasses of this study (all of which were rapidly quenched) revealed that measured durability is not a function of predicted phase separation based upon the current homogeneity model. Further work (including kinetic studies) is required to resolve this apparent inconsistency.

Liquidus. For this study, the liquidus temperature was bounded by performing 24-hour isothermal holds (as required) for the glass melts at 900°C, 950°C, 1000°C, and 1050°C. X-ray diffraction was used to detect crystallization, in this case Trevorite. For the 22 wt% and 26 wt% Purex glasses, no crystals were detected in the bulk at 900°C or at the top surface of the glasses. For the 30 wt% Purex glasses, crystals were evident at temperatures higher than 900°C, but at 1000°C they were below the XRD detection limit. Given the fact that liquidus temperatures were only approximated, the 30 wt% loading of Purex may be near or at the edge of acceptability for liquidus. A small amount of surface crystallization (after the isothermal hold at 900°C) was evident on the glass surface near the center of the crucible for four of the 30 wt% Purex glasses. The crystals detected were Trevorite. For HM glasses, no crystals were detected in the bulk or on the surface after 24 hours at 900°C.

Viscosity. The melt viscosities for 11 of these 23 glasses were measured and predictions reported at 1150°C (nominal temperature of the glass within the DWPF melter). For the Purex containing glasses, the measured viscosities were within the DWPF range of 20 to 100 poise for the 22 wt% and 26 wt% Purex glasses. However, the 30 wt% Purex glasses had viscosities that were very near the lower limit of 20 poise, and consequently, may not be acceptable from an operations perspective. Although the HM sludge glass measured (10wt% PHA) had a viscosity of ~90 poise, the HM glasses at 7wt% PHA are predicted to be higher than the 100 poise limit for DWPF.

Limitations. All of the conclusions are provided based on the scope of the current work. One limitation of this scope, based upon schedule and budget, was the absence of any investigation of kinetic effects. Thus, one can not rule out that amorphous phase separation occurs with centerline cooling, for example. A second limitation was the restriction on independent variation of chemical constituents. In a major variability study, ranges are established for each element, and a statistically designed set of glasses identified which not only covers a larger region of compositional space, but also provides the potential for revealing trends in the properties over linear variations of elemental concentrations. A third limitation was that only approximate measurements of the liquidus temperatures were made. A fourth limitation was that a thorough search (beyond scanning electron microscopy) for phase separation was not conducted. This type of investigation requires considerable efforts using transmission electron microscopy (TEM) and other high-resolution techniques. Finally, although the strategy was that HM and Purex containing glasses would cover the extremes, no Blend sludge glasses were fabricated to verify this.

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Appendix:

Supplemental Tables and Exhibits

Table A.1: Target Oxide Compositions (in weight percents, wt%'s) of the CST Glasses

Purex Sludge	Glass																
	MST	CST	Frit	ID	Al ₂ O ₃	B ₂ O ₃	BaO	CaO	Cr ₂ O ₃	CuO	Fe ₂ O ₃	K ₂ O	Li ₂ O	MgO	MnO	Na ₂ O	
22	1.250	3	73.750	cst01	2.556	8.223	0.084	0.841	0.106	0.047	9.843	0.092	4.750	0.067	1.727	8.848	
22	1.250	6	70.750	cst02	2.539	7.889	0.084	0.841	0.106	0.047	9.843	0.092	4.556	0.067	1.727	8.760	
22	1.250	9	67.750	cst03	2.521	7.554	0.084	0.841	0.106	0.047	9.843	0.092	4.363	0.067	1.727	8.672	
22	2.500	3	72.500	cst04	2.549	8.084	0.084	0.841	0.106	0.047	9.843	0.092	4.669	0.067	1.727	8.900	
22	2.500	6	69.500	cst05	2.531	7.749	0.084	0.841	0.106	0.047	9.843	0.092	4.476	0.067	1.727	8.812	
22	2.500	9	66.500	cst06	2.514	7.415	0.084	0.841	0.106	0.047	9.843	0.092	4.283	0.067	1.727	8.725	
26	1.250	3	69.750	cst07	2.918	7.777	0.099	0.994	0.125	0.056	11.633	0.109	4.492	0.079	2.041	9.022	
26	1.250	6	66.750	cst08	2.901	7.443	0.099	0.994	0.125	0.056	11.633	0.109	4.299	0.079	2.041	8.934	
26	1.250	9	63.750	cst09	2.883	7.108	0.099	0.994	0.125	0.056	11.633	0.109	4.106	0.079	2.041	8.846	
26	2.500	3	68.500	cst10	2.911	7.638	0.099	0.994	0.125	0.056	11.633	0.109	4.411	0.079	2.041	9.074	
26	2.500	6	65.500	cst11	2.893	7.303	0.099	0.994	0.125	0.056	11.633	0.109	4.218	0.079	2.041	8.986	
26	2.500	9	62.500	cst12	2.876	6.969	0.099	0.994	0.125	0.056	11.633	0.109	4.025	0.079	2.041	8.898	
30	1.250	3	65.750	cst13	3.281	7.331	0.114	1.146	0.144	0.064	13.423	0.126	4.234	0.091	2.355	9.195	
30	1.250	6	62.750	cst14	3.263	6.997	0.114	1.146	0.144	0.064	13.423	0.126	4.041	0.091	2.355	9.107	
30	1.250	9	59.750	cst15	3.245	6.662	0.114	1.146	0.144	0.064	13.423	0.126	3.848	0.091	2.355	9.019	
30	2.500	3	64.500	cst16	3.273	7.192	0.114	1.146	0.144	0.064	13.423	0.126	4.154	0.091	2.355	9.247	
30	2.500	6	61.500	cst17	3.255	6.857	0.114	1.146	0.144	0.064	13.423	0.126	3.961	0.091	2.355	9.159	
30	2.500	9	58.500	cst18	3.238	6.523	0.114	1.146	0.144	0.064	13.423	0.126	3.767	0.091	2.355	9.071	

Purex Sludge	Glass																
	MST	CST	Frit	ID	Nb ₂ O ₅	NiO	P ₂ O ₅	PbO	SiO ₂	TiO ₂	U ₃ O ₈	ZnO	ZrO ₂	F	Cl	(SO ₄) ⁻	
22	1.250	3	73.750	cst01	0.660	0.930	0.030	0.096	55.760	2.149	2.003	0.086	0.619	0.032	0.240	0.173	
22	1.250	6	70.750	cst02	1.320	0.930	0.030	0.096	54.144	3.199	2.003	0.086	1.129	0.032	0.240	0.173	
22	1.250	9	67.750	cst03	1.980	0.930	0.030	0.096	52.528	4.249	2.003	0.086	1.639	0.032	0.240	0.173	
22	2.500	3	72.500	cst04	0.660	0.930	0.030	0.096	54.837	3.247	2.003	0.086	0.619	0.032	0.240	0.173	
22	2.500	6	69.500	cst05	1.320	0.930	0.030	0.096	53.221	4.297	2.003	0.086	1.129	0.032	0.240	0.173	
22	2.500	9	66.500	cst06	1.980	0.930	0.030	0.096	51.604	5.347	2.003	0.086	1.639	0.032	0.240	0.173	
26	1.250	3	69.750	cst07	0.660	1.099	0.036	0.114	52.928	2.149	2.367	0.102	0.639	0.038	0.283	0.205	
26	1.250	6	66.750	cst08	1.320	1.099	0.036	0.114	51.311	3.199	2.367	0.102	1.149	0.038	0.283	0.205	
26	1.250	9	63.750	cst09	1.980	1.099	0.036	0.114	49.695	4.249	2.367	0.102	1.659	0.038	0.283	0.205	
26	2.500	3	68.500	cst10	0.660	1.099	0.036	0.114	52.004	3.247	2.367	0.102	0.639	0.038	0.283	0.205	
26	2.500	6	65.500	cst11	1.320	1.099	0.036	0.114	50.388	4.297	2.367	0.102	1.149	0.038	0.283	0.205	
26	2.500	9	62.500	cst12	1.980	1.099	0.036	0.114	48.771	5.347	2.367	0.102	1.659	0.038	0.283	0.205	
30	1.250	3	65.750	cst13	0.660	1.268	0.041	0.132	50.095	2.149	2.731	0.118	0.659	0.043	0.327	0.236	
30	1.250	6	62.750	cst14	1.320	1.268	0.041	0.132	48.479	3.199	2.731	0.118	1.169	0.043	0.327	0.236	
30	1.250	9	59.750	cst15	1.980	1.268	0.041	0.132	46.862	4.249	2.731	0.118	1.679	0.043	0.327	0.236	
30	2.500	3	64.500	cst16	0.660	1.268	0.041	0.132	49.172	3.247	2.731	0.118	0.659	0.043	0.327	0.236	
30	2.500	6	61.500	cst17	1.320	1.268	0.041	0.132	47.555	4.297	2.731	0.118	1.169	0.043	0.327	0.236	
30	2.500	9	58.500	cst18	1.980	1.268	0.041	0.132	45.939	5.347	2.731	0.118	1.679	0.043	0.327	0.236	

Table A.2: Target Compositions of PHA Phase 4 HM Glasses

Glass ID	pha20	pha26	pha32	Glass ID	pha20	pha26	pha32
Sludge	22	26	30	Sludge	22	26	30
MST	1.25	1.25	1.25	MST	1.25	1.25	1.25
PHA	10	10	10	PHA	10	10	10
Frit 202	66.75	62.75	58.75	Frit 202	66.75	62.75	58.75
Al ₂ O ₃	6.046	7.048	8.051	NiO	0.336	0.397	0.458
B ₂ O ₃	8.803	8.488	8.174	P ₂ O ₅	0.032	0.038	0.044
BaO	0.045	0.053	0.061	PbO	0.053	0.063	0.073
CaO	0.496	0.562	0.628	SiO ₂	54.086	51.656	49.226
Cr ₂ O ₃	0.073	0.086	0.099	TiO ₂	1.127	1.125	1.123
CuO	0.761	0.764	0.767	U ₃ O ₈	0.677	0.800	0.923
Fe ₂ O ₃	6.363	7.507	8.652	ZnO	0.014	0.016	0.019
K ₂ O	4.723	4.736	4.749	ZrO ₂	0.119	0.141	0.163
Li ₂ O	4.579	4.305	4.030	F-	0.037	0.044	0.051
MgO	1.463	1.409	1.355	Cl-	0.124	0.147	0.169
MnO	1.955	2.311	2.667	(SO ₄)-	0.127	0.150	0.173
Na ₂ O	7.776	7.980	8.183				

Table A.3: Target, Measured and Bias-Corrected Compositions (in wt%) for the CST Glasses

	Phase 1 - pha07			Phase 1 - pha08			Phase 1 - pha09		
	Target	Measured	Bias-cor.	Target	Measured	Bias-cor.	Target	Measured	Bias-cor.
Al ₂ O ₃	2.901	2.650	2.758	2.883	2.711	2.822	2.865	2.707	2.817
B ₂ O ₃	7.660	7.688	7.403	8.488	8.766	8.435	9.317	9.410	9.068
CaO	1.092	1.119	1.051	1.088	1.129	1.060	1.083	1.099	1.032
Cr ₂ O ₃	0.125	0.121	0.129	0.125	0.113	0.120	0.125	0.113	0.121
CuO	0.576	0.492	0.514	0.800	0.693	0.725	1.023	0.761	0.795
Fe ₂ O ₃	11.685	10.308	10.855	11.683	10.626	11.201	11.681	9.951	10.475
K ₂ O	3.365	3.084	3.071	4.745	4.352	4.335	6.125	5.294	5.273
Li ₂ O	4.510	4.236	4.468	4.305	4.080	4.304	4.099	4.037	4.259
MgO	1.381	1.382	1.417	1.322	1.331	1.365	1.262	1.272	1.304
MnO	2.041	1.863	1.938	2.041	1.875	1.950	2.041	1.905	1.981
Na ₂ O	8.116	8.071	7.910	8.264	8.391	8.222	8.412	8.374	8.206
NiO	1.099	0.866	0.902	1.099	0.861	0.897	1.099	0.857	0.893
SiO ₂	50.766	50.274	51.040	48.486	50.006	50.778	46.206	48.081	48.940
TiO ₂	1.126	0.691	0.689	1.125	0.698	0.697	1.124	0.707	0.705
U ₃ O ₈	2.367	2.633	2.633	2.367	2.161	2.161	2.367	2.565	2.565
ZrO ₂	0.129	0.128	0.137	0.129	0.126	0.134	0.129	0.132	0.140
Sum of Oxides	98.939	95.647	96.958	98.950	97.963	99.250	98.958	97.306	98.618
	Phase 1 - pha10			Phase 1 - pha11			Phase 1 - pha12		
	Target	Measured	Bias-cor.	Target	Measured	Bias-cor.	Target	Measured	Bias-cor.
Al ₂ O ₃	2.894	2.636	2.743	2.876	2.664	2.773	2.858	2.626	2.733
B ₂ O ₃	7.561	7.985	7.693	8.390	8.967	8.646	9.219	9.724	9.375
CaO	1.090	1.109	1.043	1.086	1.102	1.036	1.081	1.114	1.047
Cr ₂ O ₃	0.125	0.113	0.121	0.125	0.117	0.124	0.125	0.111	0.119
CuO	0.576	0.501	0.523	0.800	0.676	0.707	1.023	0.818	0.855
Fe ₂ O ₃	11.684	9.701	10.205	11.682	9.879	10.403	11.680	10.344	10.880
K ₂ O	3.365	3.123	3.111	4.745	4.255	4.239	6.125	5.514	5.491
Li ₂ O	4.425	4.295	4.533	4.219	4.091	4.315	4.013	3.929	4.145
MgO	1.356	1.337	1.370	1.297	1.285	1.317	1.237	1.230	1.261
MnO	2.041	1.853	1.928	2.041	1.840	1.914	2.041	1.837	1.910
Na ₂ O	8.191	8.341	8.178	8.339	8.432	8.266	8.487	8.543	8.373
NiO	1.099	0.838	0.873	1.099	0.861	0.898	1.099	0.849	0.885
SiO ₂	49.816	51.343	52.180	47.536	51.236	52.113	45.256	48.669	49.502
TiO ₂	2.224	1.369	1.365	2.223	1.337	1.334	2.222	1.350	1.346
U ₃ O ₈	2.367	2.409	2.409	2.367	2.462	2.462	2.367	2.456	2.456
ZrO ₂	0.129	0.126	0.134	0.129	0.122	0.130	0.129	0.122	0.129
Sum of Oxides	98.943	97.171	98.504	98.954	99.415	100.764	98.962	99.327	100.600
	Phase 2 - pha13			Phase 2 - pha14			Phase 2 - pha15		
	Target	Measured	Bias-cor.	Target	Measured	Bias-cor.	Target	Measured	Bias-cor.
Al ₂ O ₃	3.263	3.246	3.725	3.245	3.120	3.580	3.227	3.116	3.576
B ₂ O ₃	7.345	7.492	7.498	8.174	10.060	10.069	9.003	11.647	11.657
CaO	1.239	1.357	1.196	1.234	1.272	1.132	1.230	1.371	1.204
Cr ₂ O ₃	0.144	0.152	0.149	0.144	0.146	0.143	0.144	0.153	0.150
CuO	0.585	0.573	0.575	0.808	0.739	0.741	1.031	0.959	0.962
Fe ₂ O ₃	13.472	13.615	13.236	13.470	13.851	13.466	13.467	14.684	14.275
K ₂ O	3.380	3.470	3.694	4.760	4.150	4.417	6.140	4.607	4.905
Li ₂ O	4.236	4.214	4.171	4.030	4.006	3.964	3.824	3.766	3.727
MgO	1.314	1.284	1.570	1.255	1.228	1.502	1.195	1.145	1.401
MnO	2.355	2.272	2.359	2.355	2.176	2.259	2.355	2.156	2.239
Na ₂ O	8.363	8.342	8.673	8.511	9.175	9.539	8.659	9.959	10.354
NiO	1.268	1.142	1.089	1.268	1.109	1.057	1.268	1.134	1.081
SiO ₂	47.849	46.844	47.619	45.569	45.191	45.916	43.289	41.318	42.011
TiO ₂	1.125	0.738	0.736	1.123	0.742	0.740	1.122	0.721	0.719
U ₃ O ₈	2.731	2.792	2.792	2.731	2.921	2.921	2.731	1.988	1.988
ZrO ₂	0.149	0.182	0.194	0.149	0.171	0.182	0.149	0.183	0.195
Sum of Oxides	98.818	97.792	99.353	98.826	100.131	101.702	98.834	98.990	100.526

Table A.3: Target, Measured, and Bias-Corrected Compositions (in wt%) for the CST Glasses
(continued)

	Phase 2 - pha16			Phase 2 - pha17			Phase 2 - pha18		
	Target	Measured	Measured Bias-cor.	Target	Meas.	Bias-cor.	Target	Measured	Measured Bias-cor.
Al ₂ O ₃	3.256	3.266	3.748	3.238	3.146	3.610	3.220	3.057	3.508
B ₂ O ₃	7.246	8.555	8.562	8.075	9.512	9.520	8.904	11.880	11.890
CaO	1.237	1.603	1.370	1.233	1.374	1.203	1.228	1.442	1.247
Cr ₂ O ₃	0.144	0.152	0.149	0.144	0.149	0.146	0.144	0.149	0.145
CuO	0.585	0.549	0.551	0.808	0.741	0.744	1.031	0.960	0.963
Fe ₂ O ₃	13.471	13.805	13.421	13.469	13.834	13.449	13.466	14.339	13.940
K ₂ O	3.379	3.231	3.440	4.759	4.547	4.841	6.139	4.895	5.209
Li ₂ O	4.150	4.237	4.193	3.945	3.981	3.940	3.739	3.825	3.786
MgO	1.289	1.300	1.590	1.230	1.176	1.438	1.170	1.155	1.412
MnO	2.355	2.292	2.380	2.355	2.344	2.434	2.355	2.125	2.206
Na ₂ O	8.438	8.855	9.206	8.586	9.104	9.465	8.734	10.049	10.447
NiO	1.268	1.139	1.086	1.268	1.154	1.101	1.268	1.101	1.050
SiO ₂	46.899	47.632	48.374	44.619	44.669	45.383	42.339	42.640	43.350
TiO ₂	2.223	1.460	1.456	2.221	1.449	1.445	2.220	1.460	1.456
U ₃ O ₈	2.731	2.919	2.919	2.731	2.487	2.487	2.731	3.091	3.091
ZrO ₂	0.149	0.179	0.190	0.149	0.169	0.180	0.149	0.206	0.221
Sum of Oxides	98.820	101.263	102.724	98.830	99.910	101.457	98.837	102.451	103.997
	Phase 3 - pha01*			Phase 3 - pha02*			Phase 3 - pha03		
	Target	Measured	Measured Bias-cor.	Target	Measured	Bias-cor.	Target	Measured	Measured Bias-cor.
Al ₂ O ₃	2.540	2.868	2.856	2.522	2.718	2.707	2.504	2.542	2.531
B ₂ O ₃	7.974	9.171	9.147	8.803	8.523	8.497	9.632	9.368	9.341
CaO	0.945	1.041	1.003	0.941	1.009	0.987	0.936	0.941	0.921
Cr ₂ O ₃	0.106	0.178	0.170	0.106	0.169	0.161	0.106	0.172	0.164
CuO	0.568	0.580	0.578	0.791	0.680	0.677	1.014	1.009	1.004
Fe ₂ O ₃	9.899	9.970	9.550	9.897	9.859	9.445	9.894	9.995	9.575
K ₂ O	3.350	2.973	3.095	4.730	3.663	3.813	6.110	5.606	5.836
Li ₂ O	4.785	4.751	4.661	4.579	4.679	4.591	4.373	4.458	4.374
MgO	1.448	1.464	1.453	1.389	1.387	1.376	1.329	1.358	1.347
MnO	1.727	2.065	2.056	1.727	1.999	1.990	1.727	1.699	1.691
Na ₂ O	7.869	8.873	8.692	8.017	8.249	8.081	8.165	8.181	8.014
NiO	0.930	0.949	0.902	0.930	0.924	0.878	0.930	0.851	0.809
SiO ₂	53.684	52.702	51.831	51.404	51.174	51.557	49.124	48.797	49.167
TiO ₂	1.128	1.160	1.115	1.127	1.170	1.124	1.125	1.168	1.123
U ₃ O ₈	2.003	1.542	1.542	2.003	1.659	1.659	2.003	2.760	2.760
ZrO ₂	0.109	0.160	0.101	0.109	0.153	0.097	0.109	0.223	0.135
Sum of Oxides	99.065	99.093	98.689	99.075	98.088	97.713	99.081	99.201	98.865
	Phase 3 - pha04			Phase 3 - pha05			Phase 3 - pha06		
	Target	Measured	Measured Bias-cor.	Target	Measured	Bias-cor.	Target	Measured	Measured Bias-cor.
Al ₂ O ₃	2.532	2.517	2.507	2.514	2.508	2.497	2.496	2.602	2.591
B ₂ O ₃	7.876	8.901	8.875	8.705	7.974	7.953	9.534	9.196	9.173
CaO	0.943	0.953	0.932	0.939	0.920	0.900	0.934	0.972	0.951
Cr ₂ O ₃	0.106	0.137	0.131	0.106	0.150	0.143	0.106	0.153	0.146
CuO	0.568	0.565	0.563	0.791	0.722	0.719	1.014	0.953	0.949
Fe ₂ O ₃	9.898	8.351	8.000	9.896	9.975	9.555	9.893	10.019	9.598
K ₂ O	3.350	3.099	3.226	4.730	4.309	4.485	6.110	5.757	5.994
Li ₂ O	4.699	4.936	4.843	4.493	4.596	4.509	4.288	4.353	4.271
MgO	1.423	1.475	1.464	1.364	1.386	1.375	1.304	1.316	1.305
MnO	1.727	1.794	1.786	1.727	1.629	1.621	1.727	1.789	1.780
Na ₂ O	7.944	8.401	8.230	8.092	8.054	7.889	8.240	8.270	8.101
NiO	0.930	0.828	0.787	0.930	0.846	0.804	0.930	0.874	0.831
SiO ₂	52.734	55.094	55.471	50.454	48.402	48.834	48.174	48.432	48.793
TiO ₂	2.226	2.373	2.280	2.225	2.296	2.206	2.224	2.309	2.219
U ₃ O ₈	2.003	1.081	1.081	2.003	2.869	2.869	2.003	2.441	2.441
ZrO ₂	0.109	0.135	0.086	0.109	0.153	0.096	0.109	0.160	0.101
Sum of Oxides	99.068	100.714	100.334	99.078	96.861	96.532	99.086	99.670	99.318

Table A.3: Target, Measured and Bias-Corrected Compositions (in wt%) for the CST Glasses
(continued)

	Phase 4 - pha11c			Phase 4 - pha12c			Phase 4 - pha17c		
	Target	Measured	Bias-cor.	Target	Meas.	Bias-cor.	Target	Measured	Bias-cor.
Al ₂ O ₃	2.876	2.783	2.784	2.858	2.705	2.707	3.238	3.065	3.067
B ₂ O ₃	8.390	8.420	8.429	9.219	9.283	9.297	8.075	7.763	7.770
CaO	1.086	1.118	0.966	1.081	1.159	0.999	1.233	1.270	1.093
Cr ₂ O ₃	0.125	0.156	0.156	0.125	0.146	0.146	0.144	0.182	0.183
CuO	0.800	0.754	0.749	1.023	0.991	0.985	0.808	0.773	0.768
Fe ₂ O ₃	11.682	11.171	10.672	11.680	11.348	10.842	13.469	13.470	12.869
K ₂ O	4.745	4.109	4.307	6.125	5.397	5.661	4.759	4.510	4.733
Li ₂ O	4.219	4.403	4.245	4.013	4.280	4.126	3.945	4.024	3.879
MgO	1.297	1.295	1.270	1.237	1.248	1.224	1.230	1.091	1.070
MnO	2.041	2.067	2.052	2.041	1.984	1.969	2.355	2.659	2.639
Na ₂ O	8.339	8.310	8.094	8.487	8.510	8.289	8.586	8.208	7.994
NiO	1.099	0.971	0.932	1.099	0.980	0.940	1.268	1.200	1.152
SiO ₂	47.536	48.730	49.149	45.256	46.927	47.333	44.619	44.409	44.795
TiO ₂	2.223	2.219	2.140	2.222	2.222	2.143	2.221	2.291	2.210
U ₃ O ₈	2.367	2.101	2.101	2.367	2.328	2.328	2.731	2.111	2.111
ZrO ₂	0.129	0.173	0.139	0.129	0.155	0.125	0.149	0.181	0.145
Sum of Oxides	98.954	98.854	98.262	98.962	99.737	99.187	98.830	97.280	96.551
	Phase 4 - pha18c			Phase 4 - pha20			Phase 4 - pha26		
	Target	Measured	Bias-cor.	Target	Measured	Bias-cor.	Target	Measured	Bias-cor.
Al ₂ O ₃	3.220	3.458	3.460	6.046	5.457	5.461	7.048	6.453	6.457
B ₂ O ₃	8.904	8.998	9.007	8.803	8.684	8.693	8.488	8.365	8.373
CaO	1.228	1.361	1.179	0.496	0.553	0.472	0.562	0.660	0.531
Cr ₂ O ₃	0.144	0.180	0.180	0.073	0.113	0.113	0.086	0.120	0.120
CuO	1.031	1.081	1.074	0.761	0.706	0.702	0.764	0.691	0.687
Fe ₂ O ₃	13.466	13.985	13.361	6.363	6.983	6.671	7.507	7.348	7.020
K ₂ O	6.139	5.399	5.662	4.723	4.388	4.601	4.736	4.445	4.662
Li ₂ O	3.739	3.907	3.767	4.579	4.585	4.420	4.305	4.391	4.233
MgO	1.170	1.202	1.180	1.463	1.202	1.179	1.409	1.286	1.262
MnO	2.355	2.617	2.598	1.955	2.024	2.009	2.311	2.320	2.303
Na ₂ O	8.734	9.046	8.810	7.776	7.509	7.314	7.980	7.952	7.745
NiO	1.268	1.207	1.158	0.336	0.341	0.327	0.397	0.377	0.362
SiO ₂	42.339	42.936	43.322	54.086	53.279	53.711	51.656	52.378	52.548
TiO ₂	2.220	2.319	2.237	1.127	1.146	1.105	1.125	1.173	1.131
U ₃ O ₈	2.731	2.274	2.274	0.677	0.763	0.763	0.800	0.954	0.954
ZrO ₂	0.149	0.181	0.146	0.119	0.155	0.124	0.141	0.171	0.138
Sum of Oxides	98.837	100.226	99.490	99.383	97.959	97.738	99.315	99.102	98.814
	Phase 4 - pha32			Phase 4 - pha14c		Phase 4 - pha15c			
	Target	Measured	Bias-cor.	Target	Measured	Target	Measured		
Al ₂ O ₃	8.051	7.112	7.117	3.245	3.293	3.227	3.322		
B ₂ O ₃	8.174	7.996	8.004	8.174	7.729	9.003	8.515		
CaO	0.628	0.697	0.597	1.234	1.393	1.230	1.351		
Cr ₂ O ₃	0.099	0.126	0.127	0.144	0.154	0.144	0.157		
CuO	0.767	0.790	0.786	0.808	0.813	1.031	1.022		
Fe ₂ O ₃	8.652	8.286	7.916	13.470	13.760	13.467	13.621		
K ₂ O	4.749	4.356	4.567	4.760	4.487	6.140	5.389		
Li ₂ O	4.030	4.080	3.933	4.030	3.979	3.824	3.797		
MgO	1.355	1.104	1.083	1.255	1.271	1.195	1.215		
MnO	2.667	2.738	2.717	2.355	2.483	2.355	2.500		
Na ₂ O	8.183	8.030	7.821	8.511	8.457	8.659	8.694		
NiO	0.458	0.439	0.421	1.268	1.195	1.268	1.186		
SiO ₂	49.226	49.049	49.453	45.569	44.149	43.289	41.745		
TiO ₂	1.121	1.157	1.116	1.123	1.139	1.122	1.135		
U ₃ O ₈	0.923	0.923	0.923	2.731	2.460	2.731	2.611		
ZrO ₂	0.163	0.205	0.165	0.149	0.215	0.149	0.196		
Sum of Oxides	99.246	97.165	96.822	98.826	97.052	98.834	96.530		

Table A.4: Normalized PCTs for the CST Glasses

Glass ID	Composition	log NL [B (g/L)]	log NL [Si (g/L)]	log NL [Na (g/L)]	log NL [Li (g/L)]	B (g/L)	Si (g/L)	Na (g/L)	Li (g/L)
EA	Phase 1	1.24851	0.61951	1.15692	0.99969	17.72	4.16	14.35	9.99
EA	Phase 2	1.27261	0.61353	1.17131	1.02046	18.73	4.11	14.84	10.48
EA	Phases 3-4	1.28207	0.63353	1.18838	1.04022	19.15	4.30	15.43	10.97
ARM	Phase 1	-0.22197	-0.50764	-0.22580	-0.16767	0.60	0.31	0.59	0.68
ARM	Phase 2	-0.27166	-0.55545	-0.26983	-0.20863	0.53	0.28	0.54	0.62
ARM	Phases 3-4	-0.29085	-0.54854	-0.27396	-0.21652	0.51	0.28	0.53	0.61
pha01	measured	0.16802	-0.17747	0.11093	0.13608	1.47	0.66	1.29	1.37
pha01	measured bc	0.16916	-0.17023	0.11988	0.14439	1.48	0.68	1.32	1.39
pha01	target	0.22876	-0.18548	0.16308	0.13299	1.69	0.65	1.46	1.36
pha02	measured	0.10400	-0.22546	0.06252	0.08270	1.27	0.60	1.15	1.21
pha02	measured bc	0.10532	-0.22870	0.07145	0.09095	1.27	0.59	1.18	1.23
pha02	target	0.08996	-0.22741	0.07491	0.09208	1.23	0.59	1.19	1.24
pha03	measured	0.24524	-0.15256	0.19801	0.20929	1.76	0.70	1.58	1.62
pha03	measured bc	0.24650	-0.15584	0.20697	0.21755	1.76	0.70	1.61	1.65
pha03	target	0.23317	-0.15546	0.19886	0.21765	1.71	0.70	1.58	1.65
pha04	measured	0.18716	-0.13764	0.15247	0.17857	1.54	0.73	1.42	1.51
pha04	measured bc	0.18843	-0.14060	0.16140	0.18683	1.54	0.72	1.45	1.54
pha04	target	0.24029	-0.11862	0.17676	0.19994	1.74	0.76	1.50	1.58
pha05	measured	0.11243	-0.20595	0.04395	0.06108	1.30	0.62	1.11	1.15
pha05	measured bc	0.11357	-0.20981	0.05294	0.06938	1.30	0.62	1.13	1.17
pha05	target	0.07434	-0.22399	0.04190	0.07092	1.19	0.60	1.10	1.18
pha06	measured	0.19514	-0.17575	0.15701	0.17256	1.57	0.67	1.44	1.49
pha06	measured bc	0.19623	-0.17897	0.16598	0.18082	1.57	0.66	1.47	1.52
pha06	target	0.17947	-0.17343	0.15859	0.17909	1.51	0.67	1.44	1.51
pha07	measured	-0.02381	-0.26919	-0.06227	0.00931	0.95	0.54	0.87	1.02
pha07	measured bc	-0.00742	-0.27576	-0.05351	-0.01391	0.98	0.53	0.88	0.97
pha07	target	-0.02225	-0.27343	-0.06468	-0.01793	0.95	0.53	0.86	0.96
pha08	measured	0.00957	-0.25541	0.00584	0.05032	1.02	0.56	1.01	1.12
pha08	measured bc	0.02632	-0.26206	0.01467	0.02706	1.06	0.55	1.03	1.06
pha08	target	0.02358	-0.24200	0.01248	0.02697	1.06	0.57	1.03	1.06
pha09	measured	0.08636	-0.23318	0.07789	0.11318	1.22	0.58	1.20	1.30
pha09	measured bc	0.10245	-0.24088	0.08672	0.08993	1.27	0.57	1.22	1.23
pha09	target	0.09068	-0.21591	0.07595	0.10653	1.23	0.61	1.19	1.28
pha10	measured	-0.04560	-0.27228	-0.05734	0.00946	0.90	0.53	0.88	1.02
pha10	measured bc	-0.02942	-0.27930	-0.04879	-0.01400	0.93	0.53	0.89	0.97
pha10	target	-0.02188	-0.25917	-0.04947	-0.00349	0.95	0.55	0.89	0.99
pha11	measured	0.03705	-0.26094	0.03230	0.08296	1.09	0.55	1.08	1.21
pha11	measured bc	0.05290	-0.26830	0.04094	0.05974	1.13	0.54	1.10	1.15
pha11	target	0.06596	-0.22838	0.03710	0.06953	1.16	0.59	1.09	1.17
pha11c	measured	0.00658	-0.27729	-0.00643	0.00240	1.02	0.53	0.99	1.01
pha11c	measured bc	0.00612	-0.28101	0.00504	0.01831	1.01	0.52	1.01	1.04
pha11c	target	0.00815	-0.26651	-0.00793	0.02095	1.02	0.54	0.98	1.05
pha12	measured	0.17482	-0.20543	0.14331	0.17769	1.50	0.62	1.39	1.51
pha12	measured bc	0.19069	-0.21280	0.15203	0.15448	1.55	0.61	1.42	1.43
pha12	target	0.19798	-0.17386	0.14617	0.16850	1.58	0.67	1.40	1.47
pha12c	measured	0.17737	-0.19919	0.15490	0.15261	1.50	0.63	1.43	1.42
pha12c	measured bc	0.17672	-0.20293	0.16637	0.16851	1.50	0.63	1.47	1.47
pha12c	target	0.18039	-0.18344	0.15610	0.18061	1.51	0.66	1.43	1.52
pha13	measured	0.00224	-0.29231	-0.00987	-0.01053	1.01	0.51	0.98	0.98
pha13	measured bc	0.00188	-0.29945	-0.02678	-0.00603	1.00	0.50	0.94	0.99
pha13	target	0.01087	-0.30154	-0.01099	-0.01280	1.03	0.50	0.98	0.97
pha14c	measured	0.06317	-0.25322	0.03246	0.03802	1.16	0.56	1.08	1.09
pha14c	target	0.03885	-0.26697	0.02967	0.03252	1.09	0.54	1.07	1.08
pha15c	measured	0.24023	-0.19416	0.17513	0.18018	1.74	0.64	1.50	1.51
pha15c	target	0.21601	-0.20994	0.17687	0.17711	1.64	0.62	1.50	1.50

Table A.4: Normalized PCTs for the CST Glasses

(continued)

Glass ID	Composition	log NL [B (g/L)]	log NL [Si (g/L)]	log NL [Na (g/L)]	log NL [Li (g/L)]	B (g/L)	Si (g/L)	Na (g/L)	Li (g/L)
pha16	measured	0.02870	-0.28244	0.00482	0.01993	1.07	0.52	1.01	1.05
pha16	measured bc	0.02834	-0.28916	-0.01207	0.02446	1.07	0.51	0.97	1.06
pha16	target	0.10080	-0.27571	0.02577	0.02890	1.26	0.53	1.06	1.07
pha17c	measured	0.08084	-0.26107	0.07914	0.06536	1.20	0.55	1.20	1.16
pha17c	measured bc	0.08047	-0.26483	0.09060	0.08127	1.20	0.54	1.23	1.21
pha17c	target	0.06374	-0.26312	0.05958	0.07393	1.16	0.55	1.15	1.19
pha18c	measured	0.24209	-0.19310	0.19460	0.20194	1.75	0.64	1.57	1.59
pha18c	measured bc	0.24166	-0.19699	0.20606	0.21784	1.74	0.64	1.61	1.65
pha18c	target	0.24667	-0.18702	0.20982	0.22103	1.76	0.65	1.62	1.66
pha20	measured	-0.30200	-0.52025	-0.27890	-0.24396	0.50	0.30	0.53	0.57
pha20	measured bc	-0.30245	-0.52376	-0.26743	-0.22806	0.50	0.30	0.54	0.59
pha20	target	-0.30793	-0.52678	-0.29405	-0.24342	0.49	0.30	0.51	0.57
pha26	measured	-0.33060	-0.54519	-0.31561	-0.26829	0.47	0.28	0.48	0.54
pha26	measured bc	-0.33098	-0.54660	-0.30414	-0.25238	0.47	0.28	0.50	0.56
pha26	target	-0.33691	-0.53916	-0.31714	-0.25972	0.46	0.29	0.48	0.55
pha32	measured	-0.33341	-0.53492	-0.30177	-0.25942	0.46	0.29	0.50	0.55
pha32	measured bc	-0.33383	-0.53848	-0.29031	-0.24352	0.46	0.29	0.51	0.57
pha32	target	-0.34298	-0.53649	-0.30997	-0.25411	0.45	0.29	0.49	0.56

Exhibit A.1: Comparisons of Measured Versus Target Compositions

Exhibit A.1: Comparisons of Measured Versus Target Compositions

(Continued)

Exhibit A.1: Comparisons of Measured Versus Target Compositions

(Continued)

Exhibit A.1: Comparisons of Measured Versus Target Compositions

(Continued)

Exhibit A.1: Comparisons of Measured Versus Target Compositions

(Continued)

Exhibit A.1: Comparisons of Measured Versus Target Compositions

(Continued)

Exhibit A.1: Comparisons of Measured Versus Target Compositions

(Continued)

Exhibit A.1: Comparisons of Measured Versus Target Compositions

(Continued)

Exhibit A.1: Comparisons of Measured Versus Target Compositions

(Continued)

Exhibit A.1: Comparisons of Measured Versus Target Compositions

(Continued)

Exhibit A.1: Comparisons of Measured Versus Target Compositions

(Continued)

Exhibit A.1: Comparisons of Measured Versus Target Compositions

(Continued)

Exhibit A.1: Comparisons of Measured Versus Target Compositions

(Continued)

Exhibit A.1: Comparisons of Measured Versus Target Compositions

(Continued)

Exhibit A.1: Comparisons of Measured Versus Target Compositions

(Continued)

Exhibit A.1: Comparisons of Measured Versus Target Compositions

(Continued)

Exhibit A.1: Comparisons of Measured Versus Target Compositions

(Continued)

Exhibit A.2: Durability Predictions Versus Measurements

PCTs normalized and del G_p determined from measured and measured bias-corrected compositions.

Exhibit A.2: Durability Predictions Versus Measurements
(continued)

PCTs normalized and ΔG_p determined from measured and measured bias-corrected compositions.

Exhibit A.2: Durability Predictions Versus Measurements
(continued)

PCTs normalized and del G_p determined from measured and measured bias-corrected compositions.

Exhibit A.2: Durability Predictions Versus Measurements
(continued)

PCTs normalized and $\text{del } G_p$ determined from measured and measured bias-corrected compositions.

Exhibit A.2: Durability Predictions Versus Measurements
(continued)

Log NL(B) (g/L) By del G_p

PCTs normalized and del G_p determined from target compositions.

**Exhibit A.2: Durability Predictions Versus Measurements
(continued)**

PCTs normalized and del G_p determined from target compositions.

**Exhibit A.2: Durability Predictions Versus Measurements
(continued)**

PCTs normalized and del G_p determined from target compositions.

**Exhibit A.2: Durability Predictions Versus Measurements
(continued)**

PCTs normalized and del G_p determined from target compositions.

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