1673198

WSRC-TR-2000-00339, Rev. 0

Crystalline Phase Separation in Phosphate Containing Waste Glasses: Relevance to INEEL HAW

by

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DOE Contract No. **DE-AC09-96SR18500**

WSRC-TR-2000-00339

Crystalline Phase Separation in Phosphate Containing Waste Glasses: Relevancy to Vitrification of Idaho National Engineering and Environmental Laboratory (INEEL) High Activity Waste (U)

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Prepared for

Tank Waste Focus Area
TTP# SR1-6-WT-31
Subtask B
Milestone B.2-4

Approved by

E.W. Holtzscheiter, Manager
Immobilization Technology Section

Publication Date: September 30, 2000

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PREPARED FOR THE U.S. DEPARTMENT OF ENERGY UNDER CONTRACT NO. DE-AC09-96SR18500

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WSRC-TR-2000-00339

Distribution Category: Unlimited
Keywords: Glass, High Level Waste, phase separation
Retention: Permanent

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EXECUTIVE SUMMARY

Phase separation is typically defined as the separation, upon cooling, of a homogeneous glass melt into two or more phases. When the two phases are both amorphous (e.g., amorphous phase separation (APS) or glass-in-glass phase separation) glass durability is governed by durability of the continuous phase. The impacts of APS on complex borosilicate waste glasses has been shown to be detrimental to glass durability although the exact type and extent of phase separation is not well know. When the two separating phases are amorphous at the melt temperature but crystallize upon cooling the phenomena is called crystalline phase separation (CPS).† Less is known about the impact of CPS on borosilicate waste glass durability than about the impact of APS on borosilicate waste glass durability. Since the presence of APS is known to limit HLW waste loadings, it is important to understand the impact of CPS.

One option for immobilization of high level waste at the Idaho National Engineering and Environmental Laboratory (INEEL) is to dissolve calcined waste and then separate the High Activity Waste (HAW) portion for vitrification. One of the proposed separation processes concentrates the radionuclides in the HAW but adds large concentrations of P_2O_5. Concentrations of P_2O_5 in excess of 2.5-3 wt% are known to cause crystalline phase separation (CPS) in borosilicate waste glasses. The phase separated phosphate rich regions are discreet liquid droplets at the melt temperature that transform into crystalline alkali phosphate phases upon cooling under even the most rapid cooling conditions.

The P_2O_5 containing borosilicate glass compositions examined in this study contained between 0.06-13.5 wt% P_2O_5. Some glasses had undergone CPS while other glasses had undergone amorphous phase separation (APS). Many of the higher P_2O_5 containing glasses that underwent CPS were more durable than the glasses containing lower P_2O_5 (<2.6 wt%) that underwent APS. Mathematical analysis of the glass durability data showed that high Al_2O_3 and ZrO_2 concentrations in the glass had the following effects:

- improved the durability of the homogeneous glasses
- stabilized high B_2O_3 containing glasses from undergoing APS
- improved the durability of the glassy matrix in glasses undergoing CPS

† As used in this study, CPS refers to glasses that are immiscible (exhibit droplet formation of a second liquid phase) at the melt temperature: the droplets transform during cooling into crystalline phases, e.g. the matrix phase remains amorphous while the droplets crystallize.

The high Al_2O_3 content did not stabilize the glass against CPS as it does in borosilicate glasses that undergo APS. Alumina and zirconia were found to stabilize the CPS glass matrix against APS and stabilize the homogeneous glasses against APS. All INEEL glasses with a combined concentration of Al_2O_3 and $ZrO_2 \geq 9$ wt% were found to have adequate durability

Due to the crystalline nature of the P_2O_5 phase separation and the minimal effect of the CPS phases on durability (based on the normalized release of B and Na), CPS should be treated as a crystallization phenomena rather than a phase separation phenomena for waste acceptance. It remains to be demonstrated that the radionuclides do not preferentially partition into the CPS phase and leach at a greater rate than the B or Na although there was no evidence in this study that uranium partitioned into the CPS phases. It also must be shown that the formation of the phosphate rich CPS phase at the melt temperature where it is still in liquid form does not adversely effect the processability of the glass, i.e. electrical resistivity and/or incompatibilities with materials of construction.

It is recommended that glass composition regions for future study include those in the table below if the high P_2O_5 separations process is chosen for INEEL.

Glass Compositional Component(s)	Ranges (wt %)
B_2O_3	≥2 wt% and ≤14wt%
$Na_2O + Li_2O + Cs_2O + K_2O$	≤22 wt%
P_2O_5	0.06 to >13.25
Al_2O_3	0-25
ZrO_2	0-15
$Al_2O_3 + ZrO_2$	≥9

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APS	Amorphous Phase Separation
ASTM:	American Standards and Testing Materials
CPS	Crystalline Phase Separation
DOE:	United States Department of Energy
DWPF:	Defense Waste Processing Facility
HAW:	High Activity Waste
INEEL:	Idaho National Engineering and Environmental Laboratory
PCT:	Product Consistency Test (ASTM C1285)
[SB]	Concentration of Strong Bases in Leachates
SEM:	Scanning Electron Microscopy
SRS:	Savannah River Site
SRTC:	Savannah River Technology Center
TEM:	Transmission Electron Microscopy
[WA]	Concentration of Weak Acids in Leachates
WSRC:	Westinghouse Savannah River Company
XRD:	X-Ray Diffraction

Crystalline Phase Separation in Phosphate Containing Waste Glasses: Relevancy to Vitrification of Idaho National Engineering and Environmental Laboratory (INEEL) High Activity Waste (U)

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1.0 INTRODUCTION

As part of the Tanks Focus Area's (TFA) effort to increase waste loading for high-level waste vitrification at various facilities in the Department of Energy (DOE) complex, the occurrence of phase separation in waste glasses spanning the Savannah River Site (SRS) and Idaho National Engineering and Environmental Laboratory (INEEL) composition ranges have been studied. The type of phase separation that occurs in the phosphate rich borosilicate waste glasses, such as those investigated for INEEL, crystallizes upon cooling. This type of phase separation mechanism is less well studied than amorphous phase separation in phosphate poor borosilicate waste glasses. Therefore, the type of phase separation, extent, and impact of phase separation on glass durability for a series of INEEL-type glasses were examined and the data statistically analyzed in this study.

Phase separation is typically defined as the separation, upon cooling, of a homogeneous glass melt into two or more phases. When the two phases are both amorphous (e.g. amorphous phase separation (APS) or glass-in-glass phase separation) glass durability can be governed by the least durable continuous phase (Types A and B) or by the most durable continuous phase (Type C). The occurrence of Type A or Type B amorphous phase separation in a glass causes the overall glass durability to be poor. The occurrence of Type C amorphous phase separation in a glass causes little or no impact on the overall glass durability. When the two separating phases are amorphous at the melt temperature but crystallize upon cooling the phenomena is called crystalline phase separation (CPS) and little is known about the impact of CPS on borosilicate waste glass durability.

The type of phase separation found in borosilicate waste glasses in both the United States[1,2,3] and in Europe[4] has been shown to have an adverse and unpredictable effect on glass durability, the continuous phase is poorly durable and governs the durability response. For this reason, glass compositions that have a tendency to phase separate are excluded from consideration during waste processing in the SRS Defense Waste Processing Facility (DWPF).[2,5] The phase separation exhibited by these low (≤2.6 wt%) P_2O_5 containing borosilicate glasses is glass-in-glass or APS.

The type and scale of phase separation in phosphate containing borosilicate glasses, such as those proposed for the processing of high level waste at the INEEL,[6] is different than the phase separation in the low P_2O_5 containing borosilicate waste glasses. The high concentrations of P_2O_5 in the INEEL wastes arise from one of the proposed processing options: dissolution of calcined waste followed by separation of the High Activity Waste (HAW) portion for vitrification. It is the separation process that concentrates the radionuclides in the HAW and adds large amounts of P_2O_5.

In phosphate-rich glasses, phosphate-rich regions form that are molten droplet like at the melt temperature: the glass cannot be cooled quickly enough to prevent the droplets from crystallizing into alkali phosphate phases. This type of phase separation is known as crystalline phase separation (CPS). The kinetics and the effects of the phase separation and composition on glass durability are different in CPS and APS. It is the purpose of this study to (1) investigate the effects of CPS in high P_2O_5 containing waste glasses on glass durability and to (2) recommend glass composition regions for future study if the high P_2O_5 separations process is chosen for INEEL.

2.0 AMORPHOUS VS. CRYSTALLINE PHASE SEPARATION

Glasses that contain significant amounts of two or more glass-forming oxides are likely candidates for phase separation. Borosilicate glasses, although prone to APS, are widely used in the commercial glass industry. Compositions are selected to avoid the phase separated region or to take advantage of glass properties, such as thermal expansion, that are positively influenced by phase separation.

When alkali borosilicate glasses undergo APS, two liquid phases differing in composition usually develop that are not miscible in each other. Each compositional domain is usually enriched in one of the three major glass forming oxides, e.g. silica as $(SiO_4)^{-4}$ tetrahedral units, boria as $(BO_4)^{-5}$ tetrahedral units† or $(BO_3)^{-3}$ trigonal units,‡ or

† where B is surrounded by four oxygen atoms or IV coordinated

‡ where B is surrounded by three oxygen atoms or III coordinated

phosphorous oxide as $(PO_4)^{-3}$ tetrahedral units. The competition for dominant structural role causes one or more of the three types of tetrahedral units to phase separate. In borosilicate glasses, $(PO_4)^{-3}$ will separate first along with accompanying charge balancing cations.[7] In general, the strong tendency toward phase separation can be anticipated from the competitive strong field strengths of the glass-formers P^{+5}=2.1, Si^{4+}=1.57, B^{3+}_{III}=1.63 and B^{3+}_{IV} = 1.34 (where III and IV refer to the coordination of the oxygen ions surrounding each B). This causes phase separation in all the known binary systems, e.g., SiO_2-B_2O_3, SiO_2-P_2O_5, and B_2O_3-P_2O_5.[7]

In ternary borosilicate glasses with little or no phosphate (glasses predominately in the Na_2O-B_2O_3-SiO_2 system), a silica-rich phase often phase separates from an alkali-boron enriched domain. Tomozawa[8] described three types of APS in alkali borosilicate glasses based on microstructure:

Type A: Both phases are continuous and interconnected (Figure 1)
- the durability of the glass is governed by the least durable phase
- usually formed as a result of spinodal decomposition

Type B: A silica-rich phase is dispersed as droplets in a non-durable (Figure 2)
- continuous matrix which is an alkali-borate phase
- usually formed by nucleation and growth

Type C: An alkali-borate phase is dispersed as droplets in a continuous matrix of silica-rich phase (Figure 3)
- the durability of the glass is governed by the continuous phase that is highly durable
- usually formed by nucleation and growth.

Complex combinations of these types of APS can occur in the same glass, e.g., glasses are known that undergo "primary" APS of Type B while the borate-rich continuous matrix undergoes a "secondary" Type B APS. Such complex phase separation can result in glasses with 3 or more different amorphous phases[7] each with a different durability response. Indeed, Figure 2 shows "primary" phase separation of Type B (large SiO_2-rich droplets in a sodium borate rich continuous matrix). However, the borate-rich continuous matrix has undergone a "secondary" phase separation of Type B and smaller SiO_2-rich droplets can be seen within the borate-rich matrix. Likewise, in Figure 3 the "primary" phase separation is Type C and large sodium-borate rich droplets have formed in a durable SiO_2-rich continuous matrix phase. There are also smaller Type C sodium-borate

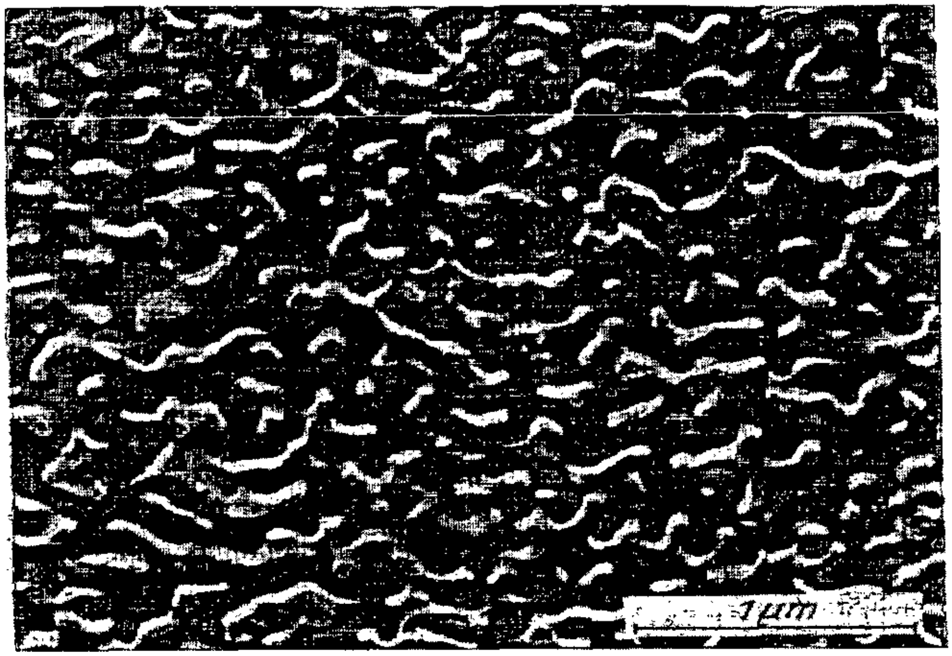

Figure 1. Example of Type A phase separation (described above) in sodium borosilicate glass.[7]

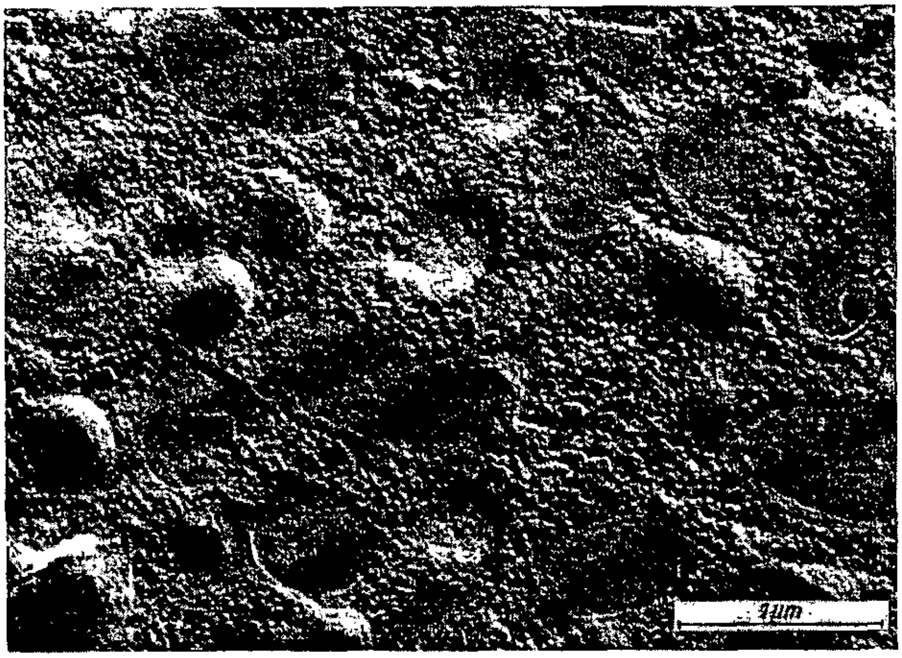

Figure 2. Example of Type B phase separation (described above) in sodium borosilicate glass.[7]

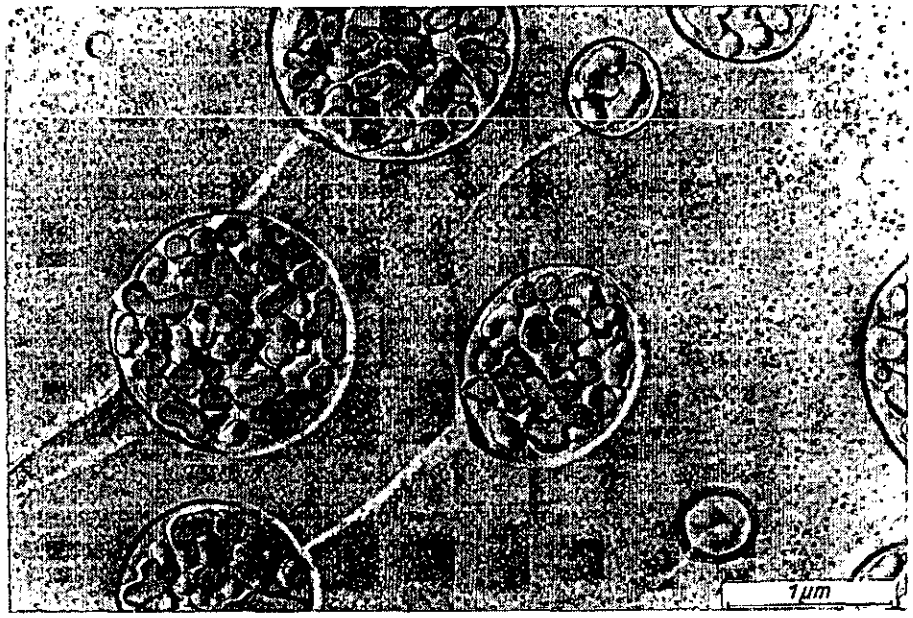

Figure 3. Example of Type C phase separation (described above) in sodium borosilicate glass.[7]

rich precipitates in the continuous SiO_2-rich matrix formed by "secondary" phase separation. In addition, the large sodium-rich droplets have undergone a Type B "secondary" phase separation. The resulting glass has 4 different phases – three Type C separated phases and one Type B separated phase.[7]

In phosphate containing borosilicate glasses all three major glass formers are present in structural tetrahedral or trigonal groups, e.g. $(SiO_4)^{-4}$, $(BO_4)^{-5}$, $(BO_3)^{-3}$, and $(PO_4)^{-3}$. The competition for dominant structural role causes the cation with the highest field strength, P^{+5}, to dominate the de-mixing process. If the difference in the field strengths (ΔF) between the major glass forming (structural) elements exceeds 0.3, then the phase separation will be of a crystalline and not amorphous nature according to Dietzel,[7] e.g. the differences between B and P and Si and P exceed the limit of 0.3 while the ΔF difference between B and Si is only 0.06. Therefore, the phase separation in phosphate systems is of a crystalline nature, CPS. Glasses in these phosphate systems phase separate to crystalline or partially crystalline solids, depending on quench rate (Figures 4 and 5).

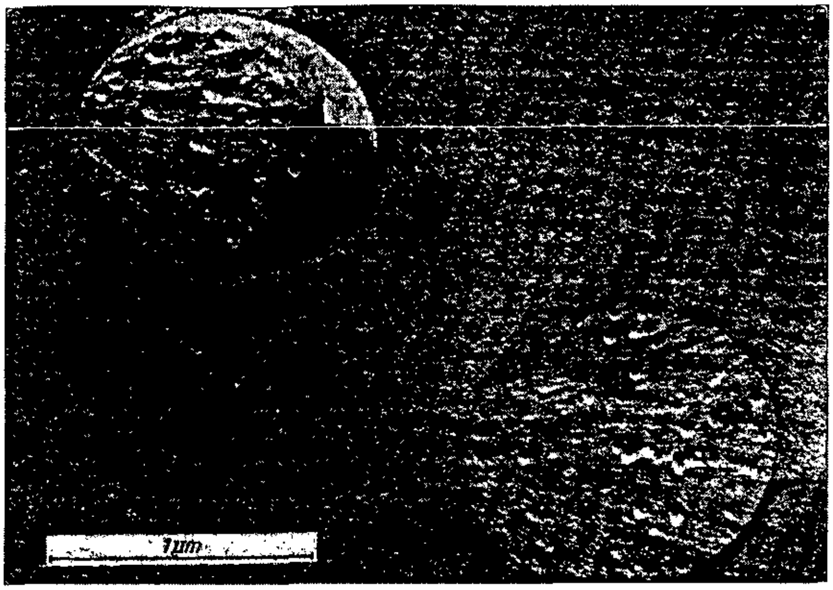

Figure 4. $CaO-P_2O_5$ rich droplets which have crystallized in a phosphate rich borosilicate glass (from Vogel, ref.[7]).

Figure 5. Li_3PO_4 crystalline droplets in an SRS phosphate rich borosilicate glass (from Jantzen[9]).

3.0 HISTORICAL BACKGROUND

The earliest work reported on CPS in nuclear waste glasses was that of Jantzen[9] in 1986 (Table I; Figure 5): crystalline Li_3PO_4 was found to preferentially form as droplets in Li_2O containing borosilicate waste glasses when the solubility limit of P_2O_5 in the glass was exceeded (≥2.6 wt% P_2O_5). Other studies (Table I) showed that $Na_3(PO_4)_2$ crystallized when Na_2O was the only alkali oxide present while $Ca_3(PO_4)_2$ or the mixed phosphate phase, $Na_2Ca_4(PO_3)_2SiO_4$ crystallized when both Na_2O and CaO oxides were present. The P_2O_5 solubility limits in each study were in the 2.5-3.0 wt% range depending on the particular alkali and alkaline earth species present.

Table I. Phase Separation in Borosilicate Glasses Containing P_2O_5

P_2O_5 (wt%)	B_2O_3 (wt%)	ΣM_2O (wt%)‡	SiO_2/ B_2O_3	Major Alkali	CPS Phase	Glass/Melt Temp. (°C)	Ref
Borosilicate Glasses Containing Li_2O							
≥2.6	7.2	~16	7.64	Na_2O + Li_2O	Li_3PO_4	DWPF 165 @ 1150°C	Jantzen (1986)[9]
3.33-7.33	13.06 - 13.63	10.92 - 11.15	3.83	Li_2O + Na_2O	Li_3PO_4 + Cr_2O_3 + $AlPO_4$	PFP‡‡ Waste @ 1150°C	Langowski (1996)[10]
Borosilicate Glasses Without Li_2O							
2.68-5.05	17.6-19.5	16-17.66	3.06-3.47	Na_2O	SiO_2 (Cristobalite + Tridymite)	Na_2O-B_2O_3-SiO_2 @1150°C	Cozzi, (1998)[11]
~3	5.00	20.33	11.36	Na_2O + CaO + K_2O	Na_3PO_4 and $Na_2Ca_4(PO_3)_2SiO_4$	LLW @ 1300°C	Crichton et. al. (1995)[12]
~2.5	17	6.5	4.16	Na_2O + CaO	$Ca_3(PO_4)_2$	Comm. Pyrex	Vogel, (1985)[7]

‡ where $M_2O = \Sigma Na_2O+Li_2O+K_2O+Cs_2O$ ‡‡Pu Finishing Plant

4.0 EXPERIMENTAL

Glasses from the INEEL waste glass composition variability study[Error! Bookmark not defined.] were examined by x-ray diffraction (XRD) analyses in order to identify the CPS phases present. Analyses performed in this study are shaded in Table II. The INEEL database containing 44 "as batched" glass compositions was combined with a mixed waste treatability study database containing 51 analyzed actual mixed waste glasses from vitrification of SRS M-Area wastes (8 from pilot scale runs and 43 crucible studies). The M-Area data has here-to-fore not been previously published. All the previously unpublished data is shaded in Tables II, III, and IV. The INEEL glasses had all been melted in platinum crucibles at 1150°C except glass IG1-33 which was melted at 1450°C. The INEEL glasses had been rapidly quenched by pouring onto a steel block. The M-Area waste glasses had been melted in 99.9% Al_2O_3 crucibles since this was high Al_2O_3 containing waste there was no interaction with the ceramic crucibles. The glasses had been cooled in the crucibles undergoing a somewhat slower quench than the INEEL glasses. The remainder of the M-Area glasses were poured from a pilot scale melter at 1150°C. The glass was poured into stainless steel cans about 8" in diameter and 12" deep. This glass underwent the slowest quench rate of all the glasses studied. Details of the fabrication of the INEEL, M-Area, and other glasses is given in Table II. The CPS phases in the M-Area waste glasses had been analyzed by XRD.

The INEEL waste glasses contained up to 13.5 wt% P_2O_5 while the M-Area glasses contained a maximum of ~ 4 wt% P_2O_5. The INEEL glasses contained no U_3O_8 while the M-Area glasses contained up to 5.66 wt% U_3O_8. The INEEL waste glasses contained up to 14 wt% ZrO_2 because some of the INEEL waste calcines are high in ZrO_2. The M-Area wastes contained no ZrO_2 but this component was added to enhance glass durability (maximum level of 2 wt%). The Al_2O_3 ranged from 4 to 15 wt% in the INEEL study and from 12.75 to 29 wt% in the M-Area glasses. Concentrations of Na_2O in the INEEL waste glasses spanned between 5 and 21.3 wt% while the M-Area glasses spanned between 6.8 and 27.09 wt%. Lastly, 4.37 to 15 wt% B_2O_3 was used as a glass forming flux in the high ZrO_2 containing INEEL waste glasses while 5.19 to 32.62 wt% B_2O_3 was used as a flux in the high Al_2O_3 containing M-Area waste. A comparison of the compositional ranges spanned by the borosilicate M-Area and INEEL studies is given in Table V. Ten of the M-Area glasses are soda-lime-silicate glasses. Individual measured and "as batched" glass compositions are given in Table III.

The durability of the M-Area and INEEL waste glasses were tested using ASTM C1285-97 (Method A for the INEEL glasses and Method B for the M-Area glasses at the reference 7 day, 90°C, and 1:10 ratio of mass solids: mass solution). The crushed glass tested was sieved to –100+200 mesh and the fines washed first in water and then in ethanol. While it is recognized in the ASTM C1285-97 procedure that removal of the fines adhering to the −100+200 mesh fraction in water can preferentially remove soluble components from the glass, comparison testing by washing in ethanol only is in progress. A mathematical approach is used in Section 6.0 to determine if any constituents of interest were preferentially removed due to the water wash. Measured normalized release rates are given in Table IV.

The 85 borosilicate glasses in the INEEL and M-Area studies spanned a wide composition range in terms of the alkalis, Al_2O_3, ZrO_2, P_2O_5 and U_3O_8. Another 8 data points from Cozzi[11] on simple three and four component sodium borosilicate glasses with and without P_2O_5 were added to the database (see Tables II, III, and IV). Only 6 of these 8 glasses could be used to assess glass durability since PCT measurements were missing for 2 of the glasses. The 4 waste glasses from the work of Langowski, Li, et. al[10] could not be used because durability data was not available. Only "as-batched" glass compositions were available from the Cozzi and Langowski studies. A total of 91 borosilicate glasses (41 from M-Area, 44 from INEEL and 6 from the Cozzi study) of the 107 glasses given in Tables II, III, and IV compose the database used in this study. The 10 soda-lime silica glasses, 2 Cozzi glasses, and 4 Langowski glasses are omitted from further consideration.

5.0 TYPES OF CRYSTALLINE PHASE SEPARATION (CPS) OBSERVED

The CPS phases identified in the glasses are summarized in Table V. Eight of 41 M-Area glasses crystallized Li_3PO_4 as the primary crystalline phase and 3 crystallized Li_3PO_4 along with an additional silicate or SiO_2. Only one glass, MN-5 exceeded its solubility limit and crystallized α-Al_2O_3. Glass MN-26 appeared to have incompletely reacted and sodium carbonate (a glass forming additive) was identified. Twenty four of the 44 INEEL glasses crystallized Li_3PO_4 and/or Li_2NaPO_4. One glass crystallized $Li_2MgP_2O_7$ and another crystallized Na_2ZrSiO_5. Only one glass, IG1-30 appeared to have exceeded its solubility limit and crystallized ZrO_2.

Many of the glasses appeared visually to be amorphous and were found to be amorphous during XRD analyses (Table II). However, it was suspected that several of the glasses had undergone amorphous phase separation (APS) that would not have been observed by XRD. Of the glasses examined by TEM, those classified as inhomogeneous were found to have crystalline phases.

Table II. Database of Borosilicate Glasses Containing P_2O_5: Fabrication Methods and Homogeneity

Sample ID	Melt Temperature (°C)	Melt /Quench Technique	Type of Glass	Analytic Tool Used	Crystalline Phase Separated
IG1-01	1150	Crucible/Patty	Borosilicate	TEM/XRD	Li_3PO_4
IG1-02	1150	Crucible/Patty	Borosilicate	XRD	Li_3PO_4
IG1-03	1150	Crucible/Patty	Borosilicate	XRD	Na_3PO_4
IG1-04	1150	Crucible/Patty	Borosilicate	TEM/XRD	Li_3PO_4
IG1-05	1150	Crucible/Patty	Borosilicate	TEM/XRD	$Li_2MgP_2O_7$
IG1-06	1150	Crucible/Patty	Borosilicate	XRD	Amorphous
IG1-07	1150	Crucible/Patty	Borosilicate	XRD	Amorphous
IG1-08	1150	Crucible/Patty	Borosilicate	TEM	Li_3PO_4
IG1-09	1150	Crucible/Patty	Borosilicate	TEM/XRD	Li_2NaPO_4
IG1-10	1150	Crucible/Patty	Borosilicate	XRD	Amorphous
IG1-11	1150	Crucible/Patty	Borosilicate	XRD	Amorphous
IG1-12	1150	Crucible/Patty	Borosilicate	TEM	Amorphous
IG1-13	1150	Crucible/Patty	Borosilicate	TEM	Amorphous
IG1-14	1150	Crucible/Patty	Borosilicate	TEM/XRD	Amorphous
IG1-15	1150	Crucible/Patty	Borosilicate	XRD	Li_3PO_4
IG1-16	1150	Crucible/Patty	Borosilicate	TEM	Amorphous
IG1-17	1150	Crucible/Patty	Borosilicate	TEM/XRD	Li_2NaPO_4
IG1-18	1150	Crucible/Patty	Borosilicate	XRD	Li_3PO_4
IG1-19	1150	Crucible/Patty	Borosilicate	XRD	Li_3PO_4
IG1-20	1150	Crucible/Patty	Borosilicate	TEM	Li_3PO_4
IG1-21	1150	Crucible/Patty	Borosilicate	TEM/XRD	Li_3PO_4
IG1-22	1150	Crucible/Patty	Borosilicate	XRD	Li_3PO_4
IG1-23	1150	Crucible/Patty	Borosilicate	XRD	Li_3PO_4
IG1-24	1150	Crucible/Patty	Borosilicate	TEM/XRD	Li_3PO_4 Na_2ZrSiO_5
IG1-25	1150	Crucible/Patty	Borosilicate	XRD	Li_3PO_4
IG1-26	1150	Crucible/Patty	Borosilicate	TEM/XRD	Li_3PO_4
IG1-27	1150	Crucible/Patty	Borosilicate	XRD	Li_3PO_4
IG1-28	1150	Crucible/Patty	Borosilicate	XRD	Li_3PO_4
IG1-29	1150	Crucible/Patty	Borosilicate	XRD	Amorphous
IG1-30	1150	Crucible/Patty	Borosilicate	XRD	ZrO_2 (tr)
IG1-31	1150	Crucible/Patty	Borosilicate	XRD	Amorphous
IG1-32	1150	Crucible/Patty	Borosilicate	XRD	Li_3PO_4
IG1-33	1450	Crucible/Patty	Borosilicate	XRD	Amorphous
IG1-34	1150	Crucible/Patty	Borosilicate	TEM/XRD	Li_3PO_4
IG1-35	1150	Crucible/Patty	Borosilicate	TEM/XRD	Amorphous
IG1-36	1150	Crucible/Patty	Borosilicate	XRD	Amorphous
IG1-37	1150	Crucible/Patty	Borosilicate	TEM/XRD	Amorphous
IG1-38	1150	Crucible/Patty	Borosilicate	TEM/XRD	Amorphous
IG1-39	1150	Crucible/Patty	Borosilicate	TEM/XRD	Amorphous
IG1-40	1150	Crucible/Patty	Borosilicate	XRD	Li_3PO_4

Sample ID	Melt Temperature (°C)	Melt /Quench Technique	Type of Glass	Analytic Tool Used	Crystalline Phase Separated
IG1-41	1150	Crucible/Patty	Borosilicate	XRD	Li_3PO_4
IG1-42	1150	Crucible/Patty	Borosilicate	TEM/XRD	Li_3PO_4
IG1-43	1150	Crucible/Patty	Borosilicate	TEM/XRD	Amorphous
IG1-44	1150	Crucible/Patty	Borosilicate	TEM/XRD	Li_3PO_4
MN-1	1400	Crucible/In Crucible	Borosilicate	XRD	Amorphous
MN-2	1260	Crucible/In Crucible	Borosilicate	XRD	Amorphous
MN-3	1150	Crucible/In Crucible	Borosilicate	XRD	Amorphous
MN-4	1400	Crucible/In Crucible	Borosilicate	XRD	Amorphous
MN-5	1300	Crucible/In Crucible	Borosilicate	XRD	α-Al_2O_3
MN-6	1150	Crucible/In Crucible	Borosilicate	XRD	Amorphous
MN-7	1400	Crucible/In Crucible	Borosilicate	XRD	Amorphous
MN-8	1150	Crucible/In Crucible	Borosilicate	XRD	Amorphous
MN-9A	1200	Crucible/In Crucible	Borosilicate	XRD	Amorphous
MN-9B	1200	Crucible/In Crucible	Borosilicate	XRD	Amorphous
MN-10	1200	Crucible/In Crucible	Borosilicate	XRD	Li_3PO_4
MN-11	1150	Crucible/In Crucible	Borosilicate	XRD	Li_3PO_4
MN-12	1150	Crucible/In Crucible	Borosilicate	XRD	Amorphous
MN-13	1150	Crucible/In Crucible	Borosilicate	XRD	Amorphous
MN-14	1150	Crucible/In Crucible	Borosilicate	XRD	$Li_{3.2}Mg_{0.4}SiO_4$
MN-15	1200	Crucible/In Crucible	Borosilicate	XRD	Amorphous
MN-16	1150	Crucible/In Crucible	Borosilicate	XRD	Amorphous
MN-17	1350	Crucible/In Crucible	Soda-Lime	XRD	Li_3PO_4 $Ca_3Mg(SiO_4)_2$
MN-18	1150	Crucible/In Crucible	Soda-Lime	XRD	Amorphous
MN-19	1150	Crucible/In Crucible	Soda-Lime	XRD	Amorphous
MN-20	1200	Crucible/In Crucible	Soda-Lime	XRD	Li_3PO_4
MN-21	1150	Crucible/In Crucible	Soda-Lime	XRD	Amorphous
MN-22	1150	Crucible/In Crucible	Soda-Lime	XRD	$Li_{2.8}Mg_{0.6}SiO_4$
MN-23	1400	Crucible/In Crucible	Soda-Lime	XRD	$Na_2Ca_3Al_2(PO_4)_2(SiO_4)_2$
MN-24	1400	Crucible/In Crucible	Soda-Lime	XRD	Li_3PO_4
MN-25	1150	Crucible/In Crucible	Soda-Lime	XRD	Amorphous
MN-26	1150	Crucible/In Crucible	Soda-Lime	XRD	$Na(HCO_3)_2$
MHSi-1	1400	Crucible/In Crucible	Borosilicate	XRD	Amorphous
MHSi-2	1300	Crucible/In Crucible	Borosilicate	XRD	Amorphous
MHSi-3	1150	Crucible/In Crucible	Borosilicate	XRD	Amorphous
MHSi-4	1400	Crucible/In Crucible	Borosilicate	XRD	Amorphous
MHSi-5	1150	Crucible/In Crucible	Borosilicate	XRD	Amorphous
MHSi-6	1200	Crucible/In Crucible	Borosilicate	XRD	Amorphous
MHSi-11	1350	Crucible/In Crucible		XRD	$LiAlO_2$
MLSi-1	1300	Crucible/In Crucible		XRD	Amorphous
MLSi-2	1150	Crucible/In Crucible	Borosilicate	XRD	Amorphous
MLSi-3	1400	Crucible/In Crucible	Borosilicate	XRD	Amorphous
MLSi-4	1150	Crucible/In Crucible	Borosilicate	XRD	Amorphous

Sample ID	Melt Temperature (°C)	Melt /Quench Technique	Type of Glass	Analytic Tool Used	Crystalline Phase Separated
MLSi-5	1150	Crucible/In Crucible	Borosilicate	XRD	Amorphous
MLSi-7	1400	Crucible/In Crucible	Borosilicate	XRD	Amorphous
MLSi-8	1150	Crucible/In Crucible	Borosilicate	XRD	Amorphous
MLSi-9	1150	Crucible/In Crucible	Borosilicate	XRD	Amorphous
MLSi-11	1150	Crucible/In Crucible	Borosilicate	XRD	Amorphous
MIC4-144	1150	Melter/In Can	Borosilicate	XRD	Amorphous
MIC4-149A	1150	Melter/In Can	Borosilicate	XRD	Li_3PO_4
MIC4-149C	1150	Melter/In Can	Borosilicate	XRD	Li_3PO_4
MIC5-9A	1150	Melter/In Can	Borosilicate	XRD	Li_3PO_4
MIC5-13A	1150	Melter/In Can	Borosilicate	XRD	Li_3PO_4
MIC5-21A	1150	Melter/In Can	Borosilicate	XRD	Li_3PO_4 + SiO_2
MIC5-42A	1150	Melter/In Can	Borosilicate	XRD	Li_3PO_4
MIC5-42C	1150	Melter/In Can	Borosilicate	XRD	Li_3PO_4
COZZI 12X	1350	Crucible/Patty	Na_2O-P_2O_5-B_2O_3-SiO_2	TEM	SiO_2
COZZI 12X-HT	1350	Crucible/Patty	Na_2O-P_2O_5-B_2O_3-SiO_2	TEM	SiO_2
COZZI 12XP3	1350	Crucible/Patty	Na_2O-P_2O_5-B_2O_3-SiO_2	TEM	Amorphous
COZZI 12XP3-HT	1350	Crucible/Patty	Na_2O-P_2O_5-B_2O_3-SiO_2	TEM	Amorphous
COZZI 12XP5	1300	Crucible/Patty	Na_2O-P_2O_5-B_2O_3-SiO_2	TEM	Amorphous
COZZI 12XP5-HT	1300	Crucible/Patty	Na_2O-P_2O_5-B_2O_3-SiO_2	TEM	Amorphous
PFP-1	1150	Crucible/Patty	Borosilicate	XRD/SEM	Amorphous
PFP-2	1150	Crucible/Patty	Borosilicate	XRD/SEM	Li_3PO_4 + Cr2O3
PFP-3	1150	Crucible/Patty	Borosilicate	XRD/SEM	Li_3PO_4 + Cr2O3
PFP-4	1150	Crucible/Patty	Borosilicate	XRD/SEM	Li_3PO_4 + Cr_2O_3

Table III. Database of Borosilicate Glasses Containing P_2O_5: Compositions

Sample ID	Al_2O_3	B_2O_3	CaO	Cs_2O	Fe_2O_3	K_2O	Li_2O	MgO	Na_2O	NiO	P_2O_5	SiO_2	U_3O_8	ZrO_2	Others	Oxides
IG1-01	5.52	9.47	0.08	0.09	0.05	4.25	5.46	0.00	14.32	0.00	7.03	47.06	0.00	6.49	0.13	99.95
IG1-02	6.56	13.12	0.08	0.08	0.04	8.74	7.14	0.00	8.19	0.00	13.12	42.73	0.00	0.00	0.13	99.93
IG1-03	0.00	4.38	0.08	0.08	0.04	8.75	0.10	0.00	21.30	0.00	13.12	39.71	0.00	12.24	0.13	99.93
IG1-04	0.00	4.37	0.08	0.08	0.04	0.00	7.87	0.00	9.53	0.00	13.10	52.48	0.00	12.25	0.13	99.93
IG1-05	6.57	13.13	0.07	0.08	0.04	0.00	0.14	0.00	21.31	0.00	13.10	45.37	0.00	0.00	0.13	99.94
IG1-06	0.00	15.00	0.08	0.09	0.05	10.00	4.82	0.00	5.00	0.00	0.00	57.21	0.00	7.51	0.14	99.91
IG1-07	15.01	15.01	0.09	0.09	0.05	0.00	8.45	0.00	5.00	0.00	0.00	53.08	0.00	3.01	0.14	99.93
IG1-08	13.12	4.37	0.08	0.08	0.04	8.75	7.21	0.00	8.19	0.00	13.12	42.21	0.00	2.62	0.13	99.92
IG1-09	3.60	13.12	0.08	0.08	0.04	0.00	3.33	0.00	20.19	0.00	13.12	34.11	0.00	12.15	0.13	99.95
IG1-10	0.00	6.54	0.09	0.09	0.05	0.00	9.00	0.00	15.60	0.00	0.00	54.41	0.00	14.01	0.14	99.93
IG1-11	14.64	5.00	0.09	0.09	0.05	0.00	5.11	0.00	20.00	0.00	0.00	54.79	0.00	0.00	0.14	99.91
IG1-12	3.17	15.01	0.08	0.09	0.05	10.01	0.00	0.00	15.30	0.00	0.00	42.06	0.00	14.00	0.14	99.92
IG1-13	7.59	5.01	0.09	0.09	0.05	10.01	7.09	0.00	17.20	0.00	0.00	52.66	0.00	0.00	0.14	99.93
IG1-14	10.87	7.24	0.08	0.09	0.05	2.42	5.07	0.00	16.74	0.00	3.62	50.23	0.00	3.38	0.14	99.93
IG1-15	6.44	6.77	0.08	0.09	0.05	6.77	6.00	0.00	10.86	0.00	10.16	49.43	0.00	3.16	0.13	99.93
IG1-16	7.25	7.25	0.08	0.09	0.05	2.42	4.64	0.00	16.74	0.00	3.62	47.52	0.00	10.14	0.14	99.94
IG1-17	6.77	6.78	0.08	0.08	0.04	2.26	3.86	0.00	17.63	0.00	10.16	42.66	0.00	9.48	0.13	99.93
IG1-18	6.84	11.29	0.08	0.08	0.04	2.26	5.63	0.00	10.86	0.00	10.16	49.41	0.00	3.16	0.13	99.94
IG1-19	6.83	11.26	0.08	0.08	0.04	2.26	5.64	0.00	10.86	0.00	10.16	49.43	0.00	3.16	0.13	99.93
IG1-20	4.49	11.29	0.08	0.08	0.04	2.26	5.28	0.00	10.86	0.00	10.16	45.78	0.00	9.48	0.13	99.93
IG1-21	8.90	12.07	0.08	0.09	0.05	7.24	6.52	0.00	9.50	0.00	3.62	43.23	0.00	8.48	0.14	99.92
IG1-22	10.13	11.18	0.08	0.09	0.05	2.26	6.10	0.00	13.16	0.00	10.17	40.44	0.00	6.15	0.13	99.93
IG1-23	9.79	6.77	0.08	0.09	0.05	6.63	6.10	0.00	10.86	0.00	10.16	42.82	0.00	6.47	0.13	99.94
IG1-24	8.63	12.07	0.08	0.09	0.05	7.24	6.52	0.00	9.70	0.00	3.62	43.02	0.00	8.76	0.14	99.92
IG1-25	7.24	7.24	0.08	0.09	0.05	7.24	4.92	0.00	13.34	0.00	3.62	45.80	0.00	10.14	0.14	99.90
IG1-26	5.52	9.47	0.08	0.09	0.05	4.25	5.46	0.00	14.32	0.00	7.03	47.06	0.00	6.49	0.13	99.94
IG1-27	13.12	4.37	0.08	0.08	0.04	8.75	7.21	0.00	8.19	0.00	13.12	42.21	0.00	2.63	0.13	99.93

Sample ID	Al_2O_3	B_2O_3	CaO	Cs_2O	Fe_2O_3	K_2O	Li_2O	MgO	Na_2O	NiO	P_2O_5	SiO_2	U_3O_8	ZrO_2	Others	Oxides
IG1-28	13.12	4.37	0.08	0.08	0.04	8.75	7.22	0.00	8.19	0.00	13.12	42.21	0.00	2.63	0.13	99.94
IG1-29	14.00	8.39	0.00	0.03	2.00	0.00	7.00	0.00	10.63	0.24	0.06	56.99	0.00	0.00	0.46	99.82
IG1-30	2.03	6.07	0.46	0.22	0.46	0.00	7.07	0.46	18.32	1.97	0.49	45.97	0.00	11.12	3.71	98.42
IG1-31	15.00	15.00	0.09	0.10	0.05	0.00	9.00	0.00	11.87	0.00	0.00	45.65	0.00	3.00	0.15	99.92
IG1-32	15.00	5.00	0.09	0.10	0.05	10.00	8.36	0.00	5.00	0.00	5.00	51.16	0.00	0.00	0.15	99.92
IG1-33	0.00	5.00	0.09	0.10	0.05	10.00	1.16	0.00	20.00	0.00	0.00	49.36	0.00	14.00	0.15	99.92
IG1-34	0.00	7.01	0.09	0.10	0.05	0.00	9.00	0.00	5.00	0.00	5.00	59.50	0.00	14.00	0.15	99.92
IG1-35	7.50	15.00	0.09	0.10	0.05	0.00	0.16	0.00	20.00	0.00	5.00	51.86	0.00	0.00	0.15	99.92
IG1-36	7.50	5.00	0.09	0.10	0.05	10.00	9.00	0.00	11.74	0.00	0.00	56.27	0.00	0.00	0.15	99.92
IG1-37	7.62	10.13	0.09	0.10	0.05	4.97	5.68	0.00	11.91	0.00	2.60	49.02	0.00	7.59	0.15	99.92
IG1-38	3.75	12.50	0.09	0.10	0.05	2.50	6.31	0.00	8.75	0.00	1.25	53.96	0.00	10.50	0.15	99.92
IG1-39	7.51	12.50	0.09	0.10	0.05	7.50	6.75	0.00	8.83	0.00	1.25	44.69	0.00	10.49	0.15	99.92
IG1-40	5.46	10.22	0.09	0.10	0.05	7.50	5.59	0.00	8.75	0.00	3.75	54.75	0.00	3.50	0.15	99.92
IG1-41	5.13	7.50	0.09	0.10	0.05	7.50	4.71	0.00	12.68	0.00	3.75	54.75	0.00	3.50	0.15	99.92
IG1-42	10.84	7.50	0.09	0.10	0.05	7.34	6.75	0.00	8.75	0.00	3.75	47.43	0.00	7.16	0.15	99.92
IG1-43	7.04	12.50	0.09	0.10	0.05	2.50	2.49	0.00	16.25	0.00	3.75	44.49	0.00	10.50	0.15	99.92
IG1-44	0.00	7.01	0.09	0.10	0.05	0.00	9.00	0.00	5.00	0.00	5.00	59.50	0.00	14.00	0.15	99.92
MN-1	21.67	10.16	0.58	0.00	1.76	1.50	0.02	0.27	10.21	0.63	1.92	47.07	5.04	0.02	0.30	101.15
MN-2	16.85	21.83	0.37	0.00	1.63	1.33	0.01	0.23	8.75	0.55	1.54	38.76	4.44	0.02	0.38	96.68
MN-3	14.61	28.26	0.31	0.00	1.55	1.28	0.02	0.21	7.96	0.50	1.14	35.55	4.20	0.01	0.35	95.94
MN-4	21.24	7.52	0.42	0.00	1.76	1.51	0.00	0.26	12.50	0.62	2.01	44.79	4.99	0.01	0.38	98.01
MN-5	29.02	11.69	0.29	0.00	1.74	1.09	0.00	0.19	11.89	0.48	1.75	34.15	3.88	0.01	0.35	96.52
MN-6	16.76	18.89	0.29	0.00	1.43	1.20	0.00	0.21	15.28	0.49	1.10	35.42	3.81	0.01	0.29	95.17
MN-7	21.39	5.19	0.39	0.00	1.71	1.48	0.00	0.26	14.86	0.62	2.46	45.39	5.00	0.01	0.34	99.08
MN-8	18.38	19.22	0.33	0.00	1.64	1.40	0.03	0.24	8.91	0.56	1.29	38.91	4.59	0.01	0.30	95.82
MN-9A	20.26	14.09	0.35	0.00	1.53	0.92	0.00	0.20	16.80	0.49	2.01	35.98	3.52	0.06	0.27	96.49
MN-9B	19.70	13.70	0.34	0.00	1.45	0.91	0.00	0.19	18.50	0.47	1.89	34.97	3.53	0.08	0.27	96.00
MN-10	18.97	5.68	0.43	0.00	1.86	1.54	4.80	0.27	9.71	0.63	2.19	45.95	4.94	0.02	0.37	97.36
MN-11	17.77	10.65	0.33	0.00	1.59	1.30	10.04	0.22	8.58	0.52	1.94	37.97	4.20	0.02	0.28	95.39
MN-12	18.33	14.26	0.30	0.00	1.56	1.10	14.67	0.20	6.80	0.47	1.78	32.77	3.72	0.02	0.29	96.28

Sample ID	Al_2O_3	B_2O_3	CaO	Cs_2O	Fe_2O_3	K_2O	Li_2O	MgO	Na_2O	NiO	P_2O_5	SiO_2	U_3O_8	ZrO_2	Others	Oxides
MN-13	17.53	5.09	0.35	0.00	1.81	1.40	11.15	0.25	7.87	0.59	2.16	43.00	4.42	0.02	0.32	95.94
MN-14	15.91	7.45	0.43	0.00	1.59	1.24	16.86	0.22	7.64	0.52	2.22	37.04	4.07	0.03	0.27	95.49
MN-15	17.79	4.15	0.39	0.00	1.67	1.41	0.17	0.27	17.53	0.63	3.08	44.52	4.69	0.03	0.33	96.65
MN-16	18.06	5.85	0.38	0.00	1.61	1.36	0.84	0.26	17.27	0.59	1.35	44.29	2.93	0.02	0.29	95.08
MN-17	18.16	0.38	5.96	0.00	1.67	1.54	0.00	0.26	15.63	0.61	2.48	47.30	5.14	0.02	0.36	99.50
MN-18	15.77	0.18	10.23	0.00	1.53	1.36	0.01	0.22	17.96	0.56	2.86	41.66	4.17	0.02	0.31	96.86
MN-19	14.48	0.42	13.54	0.00	1.56	1.30	0.46	0.27	20.80	0.50	1.17	38.14	4.10	0.02	0.30	97.07
MN-20	17.63	0.07	5.79	0.00	1.73	1.58	4.77	0.25	9.29	0.58	2.69	46.51	4.90	0.02	0.31	96.11
MN-21	15.89	0.06	11.62	0.00	1.66	1.38	10.48	0.28	9.02	0.55	1.74	41.30	4.02	0.03	0.30	98.32
MN-22	15.62	0.11	14.35	0.00	1.55	1.49	13.69	0.27	7.92	0.51	1.56	38.63	3.98	0.01	0.29	100.00
MN-23	19.19	0.07	11.70	0.00	1.83	1.44	0.16	0.26	9.90	0.61	2.60	46.43	4.91	0.02	0.35	99.46
MN-24	18.80	0.08	4.76	0.00	1.68	1.42	0.00	0.25	15.78	0.59	2.27	45.59	4.90	0.02	0.31	96.45
MN-25	15.98	0.04	6.30	0.00	1.62	1.26	0.14	0.23	21.08	0.56	1.87	42.26	4.34	0.02	0.31	96.01
MN-26	14.17	0.57	10.18	0.00	1.48	3.22	0.67	0.21	26.70	0.50	1.58	37.75	3.84	0.01	0.29	101.17
MHSi-1	19.93	7.83	0.42	0.00	1.77	2.01	0.02	0.24	9.71	0.51	1.60	47.80	3.82	0.01	0.35	96.02
MHSi-2	19.11	15.12	0.40	0.00	1.66	1.89	0.00	0.22	9.05	0.43	2.10	45.24	3.52	0.01	0.36	99.12
MHSi-3	16.97	23.46	0.38	0.00	1.58	1.69	0.01	0.21	7.99	0.44	1.55	41.18	3.18	0.01	0.33	98.98
MHSi-4	21.95	5.65	0.42	0.00	1.74	1.97	0.00	0.24	11.56	0.48	2.44	50.11	3.79	0.01	0.40	100.77
MHSi-5	19.36	11.95	0.38	0.00	1.68	1.83	0.00	0.22	13.58	0.62	1.82	44.53	3.39	0.01	0.36	99.72
MHSi-6	19.43	15.79	0.33	0.00	1.67	1.66	0.02	0.20	13.30	0.40	2.19	40.51	3.03	0.09	0.33	98.94
MHSi-11	24.36	7.15	0.39	0.00	2.00	1.74	6.82	0.21	7.27	0.42	2.34	44.54	3.36	0.04	0.37	100.99
MLSi-1	20.70	11.16	0.33	0.00	1.55	1.13	0.40	0.24	12.26	0.72	1.56	38.98	5.66	0.04	0.45	95.18
MLSi-2	19.26	23.20	0.41	0.00	1.48	0.97	0.07	0.22	10.50	0.66	1.17	35.35	4.89	0.03	0.56	98.75
MLSi-3	15.44	32.62	0.25	0.00	1.43	0.87	0.06	0.19	9.43	0.60	1.27	31.44	4.32	0.02	0.43	98.37
MLSi-4	25.04	8.78	0.31	0.00	1.48	1.01	0.02	0.23	13.60	0.69	2.01	37.86	5.24	0.03	0.58	96.86
MLSi-5	20.94	14.32	0.30	0.00	1.53	0.94	0.04	0.22	13.45	0.67	1.96	36.45	4.55	0.02	0.57	95.95
MLSi-7	24.87	5.73	0.37	0.00	1.49	0.98	0.00	0.24	17.15	0.68	1.13	37.79	5.45	0.02	0.33	96.22
MLSi-8	20.91	15.67	0.23	0.00	1.34	0.77	0.01	0.18	22.12	0.78	0.45	35.89	3.57	0.02	0.47	102.39
MLSi-9	22.18	14.47	0.25	0.00	1.26	0.70	0.00	0.18	27.09	0.51	0.67	28.61	3.88	0.01	0.28	100.08
MLSi-11	21.33	9.34	0.32	0.00	1.51	0.98	9.30	0.21	9.23	0.75	1.12	39.91	5.25	0.02	0.23	99.51

Sample ID	Al_2O_3	B_2O_3	CaO	Cs_2O	Fe_2O_3	K_2O	Li_2O	MgO	Na_2O	NiO	P_2O_5	SiO_2	U_3O_8	ZrO_2	Others†	Oxides
MIC4-144	12.75	10.81	0.68	0.00	3.28	0.92	3.40	1.06	16.55	0.64	2.40	37.73	2.85	1.59	1.03	95.71
MIC4-149A	14.38	10.53	0.77	0.00	2.61	1.09	3.93	1.05	16.84	0.78	2.76	38.93	3.45	0.78	1.1	98.99
MIC4-149C	14.11	10.01	0.76	0.00	2.48	1.09	3.87	0.84	16.38	0.75	2.64	37.02	3.53	0.78	1.39	95.64
MIC5-9A	14.74	9.53	0.71	0.00	2.07	1.66	4.15	0.86	13.55	0.67	3.07	39.40	2.53	1.13	1.00	95.06
MIC5-13A	14.58	9.08	0.64	0.00	1.69	1.82	4.21	0.63	13.16	0.63	3.60	41.93	1.75	1.73	1.20	96.65
MIC5-21A	14.00	9.01	0.58	0.00	1.58	2.12	4.03	0.40	12.48	0.58	3.69	43.09	1.50	1.01	1.16	95.21
MIC5-42A	15.06	9.09	0.56	0.00	1.58	2.23	4.41	0.34	13.52	0.61	3.97	44.24	1.42	0.27	1.07	98.38
MIC5-42C	15.30	9.23	0.54	0.00	1.53	2.18	4.48	0.32	13.46	0.62	4.02	45.14	1.35	0.29	1.19	99.64
12X	0.00	18.26	0.00	0.00	0.00	0.00	0.00	0.00	17.36	0.00	0.00	64.38	0.00	0.00	0.00	100.00
12X-HT	0.00	18.11	0.00	0.00	0.00	0.00	0.00	0.00	17.42	0.00	0.00	62.60	0.00	0.00	0.00	98.13
12XP3	0.00	18.11	0.00	0.00	0.00	0.00	0.00	0.00	16.42	0.00	2.87	62.60	0.00	0.00	0.00	100.00
12XP3-HT	0.00	18.75	0.00	0.00	0.00	0.00	0.00	0.00	17.66	0.00	2.68	60.91	0.00	0.00	0.00	100.00
12XP5(Pt)	0.00	18.88	0.00	0.00	0.00	0.00	0.00	0.00	16.49	0.00	4.99	59.64	0.00	0.00	0.00	100.00
12XP5HT	0.00	19.49	0.00	0.00	0.00	0.00	0.00	0.00	15.98	0.00	4.83	59.70	0.00	0.00	0.00	100.00
12XP5(SiO2)	0.00	17.62	0.00	0.00	0.00	0.00	0.00	0.00	16.21	0.00	5.05	61.12	0.00	0.00	0.00	100.00
12XP5-HT(SiO2)	0.00	18.18	0.00	0.00	0.00	0.00	0.00	0.00	16.05	0.00	4.94	60.84	0.00	0.00	0.00	100.01
PFP-1	8.98	14.11	0.88	0.00	0.00	0.00	7.44	0.31	4.36	0.08	0.00	54.04	0.00	0.06	3.82	94.08
PFP-2	8.67	13.63	0.85	0.00	0.00	0.00	7.18	0.30	4.21	0.08	3.33	52.18	0.00	0.06	3.73	94.22
PFP-3	8.49	13.34	0.83	0.00	0.00	0.00	7.03	0.29	4.12	0.08	5.33	51.10	0.00	0.06	3.68	94.35
PFP-4	8.31	13.06	0.81	0.00	0.00	0.00	6.89	0.29	4.03	0.08	7.33	50.02	0.00	0.05	3.63	94.50

† The Others column consists of the concentrations of BaO, Ce_2O_3, Cr_2O_3, CuO, Cu_2O, La_2O_3, MnO, MoO_3, Nd_2O_3, PbO, SrO, TiO_2, and ZnO.

Table IV. Database of Borosilicate Glasses Containing P_2O_5: Durability Response

Sample ID	Mean Measured pH	Log NL(B) g/L	Log NL(Li) g/L	Log NL(Na) g/L	Log NL(Si) g/L	Log NL(K) g/L	Log NL(P) g/L	[SB]-[WA]
IG1-01	11.48	0.74	0.515318	0.658742	0.127089	-1.54752	0.50845	7.53
IG1-02	9.96	0.85	0.243913	0.776491	-0.07283	-1.86064	0.24768	-8.28
IG1-03	11.43	1.63	1.083531	1.518615	0.710851	-1.86114	1.62967	139.85
IG1-04	10.78	-0.12	-0.01555	0.078302	-0.3982	---	0.07902	4.32
IG1-05	10.07	0.91	-0.59135	0.650671	-0.09579	---	0.42773	-5.93
IG1-06	10.79	1.28	1.280384	1.252325	0.500573	-1.91913	---	-22.66
IG1-07	10.2	-0.25	-0.14206	-0.93903	-0.28843	---	---	-2.71
IG1-08	10.83	-0.45	-0.24717	-0.1687	-0.59751	-1.86114	-0.16091	2.31
IG1-09	10.23	0.76	0.347087	0.661228	-0.30202	---	0.50132	10.40
IG1-10	12.45	1.47	1.328698	1.27923	1.051953	---	---	67.01
IG1-11	11.94	-0.24	-0.11968	0.272815	-0.24746	---	---	8.70
IG1-12	11.04	0.96	---	0.903713	-0.50802	-1.91956	---	-2.30
IG1-13	12.7	1.77	1.667325	1.768626	1.60366	-1.91956	---	109.17
IG1-14	11.55	0	-0.24509	0.114519	-0.40989	-1.30295	-0.63774	3.64
IG1-15	10.99	-0.01	-0.10184	0.067145	-0.39112	-1.74972	-0.07802	2.01
IG1-16	11.64	-0.15	-0.36524	0.075279	-0.5041	-1.30295	-0.37620	3.82
IG1-17	11.08	-0.39	-0.88381	-0.17413	-0.62517	-1.27324	-0.51905	1.68
IG1-18	10.11	-0.2	-0.24145	-0.15177	-0.56695	-1.27324	-0.20364	0.36
IG1-19	10.06	-0.25	-0.2998	-0.18975	-0.60305	-1.27324	-0.23601	0.30
IG1-20	10.15	-0.3	-0.35723	-0.24914	-0.65939	-1.27324	-0.29858	0.22
IG1-21	11.49	0.25	0.168103	0.169491	-0.58576	-1.77887	-0.37781	2.94
IG1-22	10.5	-0.1	-0.35185	-0.10173	-0.58924	-1.27324	-0.29583	0.91
IG1-23	10.91	-0.46	-0.38333	-0.18298	-0.64871	-1.74064	-0.26646	1.71
IG1-24	11.49	0.32	0.159823	0.176573	-0.57528	-1.77887	-0.27498	1.89
IG1-25	11.7	-0.05	-0.09545	0.101429	-0.53897	-1.77887	-0.37422	4.01
IG1-26	11.5	0.79	0.559598	0.697732	0.157479	-1.54752	0.55159	8.10
IG1-27	10.94	-0.52	-0.23141	-0.17156	-0.62679	-1.86114	-0.17369	2.58
IG1-28	10.97	-0.45	-0.20205	-0.14698	-0.58723	-1.86114	-0.11722	2.66
IG1-29	10.83	-0.35	-0.14701	-0.45739	-0.38751	---	0.43410	-0.44
IG1-30	12.59	1.39	1.212621	1.254079	0.836097	---	0.79169	88.57
IG1-31	11.14	0.48	0.405468	0.195434	-0.27628	---	---	4.27
IG1-32	10.37	-0.41	-0.2389	-0.2708	-0.52274	-1.91913	-0.19564	0.98
IG1-33	12.49	1.07	1.286663	1.327485	0.933076	-1.91913	---	65.06
IG1-34	9.85	0.03	0.034918	-0.62393	-0.28921	---	-0.03816	-0.36
IG1-35	10.8	-0.06	1.565891	-0.14688	-0.51824	---	-0.73028	2.15
IG1-36	12.15	1.25	1.254658	1.252287	0.967117	-1.91913	---	63.76
IG1-37	9.9	1.19	-0.97893	1.104823	-0.01456	-1.61549	0.87843	-3.97
IG1-38	10.12	0.39	0.345961	0.168987	-0.20103	-1.31707	0.36654	-0.90
IG1-39	11.12	0.53	0.446287	0.334414	-0.46938	-1.79419	-0.39468	4.08
IG1-40	10.7	0.92	0.754279	0.860677	0.332697	-1.79419	0.51839	-2.06
IG1-41	11.73	1.61	1.21406	1.526705	1.102907	-1.79419	0.73005	-13.30
IG1-42	10.45	-0.4	-0.27761	-0.21255	-0.5985	-1.78483	-0.37739	1.26

Sample ID	Mean Measured pH	Log NL(B) g/L	Log NL(Li) g/L	Log NL(Na) g/L	Log NL(Si) g/L	Log NL(K) g/L	Log NL(P) g/L	[SB]-[WA]
IG1-43	10.22	0.16	-0.14965	0.043111	-0.6216	-1.31707	-0.50380	0.00
IG1-44	9.77	0.06	0.058648	-0.60078	-0.26072	.	-0.02201	-0.47
MN-1	8.66	-0.69	-0.30961	-0.56534	-0.76691	-0.99422	---	-1.00
MN-2	7.81	1.15	0.849125	0.522302	-0.70253	-0.47756	---	-80.29
MN-3	6.78	1.78	1.05955	1.12568	-0.48552	0.165006	---	-454.51
MN-4	8.36	-0.43	-0.88939	-0.44422	-0.60151	-0.99553	---	-1.19
MN-5	8.74	-0.46	-1.32307	-0.38111	-0.62232	-0.77963	---	-0.90
MN-6	9.03	1.14	0.98585	0.879653	-0.84576	0.131023	---	-37.44
MN-7	9.32	-0.48	-1.32307	-0.35467	-0.55969	-0.89837	---	-0.41
MN-8	8.11	1.03	-0.15176	0.520234	-0.73354	-0.45835	---	-50.93
MN-9A	9.8	0.8	-0.31236	0.729461	-0.69021	0.170715	---	2.31
MN-9B	9.8	0.82	-0.01133	0.687644	-0.67784	0.171899	---	2.31
MN-10	11.51	-1.37	-3.34837	0.462679	-0.45617	-0.16505	---	6.57
MN-11	11.17	0.15	0.121775	0.01512	-0.17538	-0.56218	---	3.32
MN-12	11.86	0.77	0.735957	0.571601	0.175261	-0.07936	---	29.54
MN-13	11.54	0.09	0.105139	0.07868	-0.12825	-0.3927	---	5.55
MN-14	11.94	0.67	0.67561	0.586283	0.303781	-0.08125	---	40.80
MN-15	10.32	-0.34	-0.9276	-0.12268	-0.48184	-0.81852	---	1.34
MN-16	10.02	0.01	-0.46765	0.029393	-0.53597	-0.62092	---	2.37
MN-17	10.09	-0.87	-1.32307	-0.36391	-0.78617	-0.9106	---	0.92
MN-18	10.72	-0.16	-1.58836	0.081112	-0.65754	-0.52322	---	5.51
MN-19	11.02	-0.14	0.01444	0.276949	-0.52413	-0.31102	---	11.17
MN-20	10.12	-1.24	-0.33634	-0.38129	-0.64488	-0.68868	---	1.03
MN-21	10.94	-0.3	0.157696	0.080102	-0.49056	-0.37286	---	11.48
MN-22	11.08	-0.12	0.512185	0.353971	-0.11955	-0.19847	---	30.87
MN-23	10.01	-1.32	-1.22652	-0.65418	-1.08527	-1.01961	---	0.11
MN-24	10.33	-0.71	-1.32307	-0.11955	-0.67341	-0.8145	---	2.30
MN-25	10.82	-0.68	-0.54716	0.331962	-0.25069	-0.17227	---	10.86
MN-26	11.34	-0.36	0.727859	0.937818	0.077372	0.187891	---	70.53
MHSi-1	8.39	-0.71	-2.16817	-0.55493	-0.74475	-1.11864	---	-0.96
MHSi-2	8.5	-0.31	-1.19042	-0.42067	-0.70769	-1.25575	---	-2.47
MHSi-3	7.76	1.32	0.426891	0.65859	-0.70732	-0.37584	---	-131.04
MHSi-4	8.53	-0.63	-1.6241	-0.492	-0.69336	-1.10204	---	-0.84
MHSi-5	8.7	-0.33	-1.32307	-0.34181	-0.62632	-0.90568	---	-1.30
MHSi-6	8.73	0.31	-2.27731	0.111424	-0.60407	-0.70689	---	-5.31
MHSi-11	10.38	-0.29	-0.20008	-0.53098	-0.4063	-0.55481	---	-0.29
MLSi-1	8.59	-0.5	-0.16356	-0.48597	-0.60112	-0.75816	---	-1.11
MLSi-2	8.39	1.08	0.863417	0.607622	-0.76291	-0.21374	---	-66.60
MLSi-3	7.02	1.87	1.613212	1.421663	-0.4686	0.658102	---	-611.10
MLSi-4	8.69	-0.35	-2.32307	-0.31139	-0.55764	-0.6396	---	-0.67
MLSi-5	9.01	0.48	0.031388	0.303884	-0.6166	-0.3513	---	-5.09
MLSi-7	9.33	-0.14	-1.6241	-0.11845	-0.47182	-0.48871	---	0.96
MLSi-8	9.31	1.06	-0.57311	0.633218	-0.83616	0.217571	---	-21.42
MLSi-9	10.2	0.95	-1.32307	0.725601	-0.78503	0.391596	---	9.36

Sample ID	Mean Measured pH	Log NL(B) g/L	Log NL(Li) g/L	Log NL(Na) g/L	Log NL(Si) g/L	Log NL(K) g/L	Log NL(P) g/L	[SB]-[WA]
MLSi-11	11.28	0.41	0.290433	0.248985	-0.2155	-0.32523	---	6.62
MIC4-144	11	-0.26	-0.29297	-0.16549	-0.49502	-0.76619	---	1.12
MIC4-149A	11.1	-0.16	-0.27128	-0.08464	-0.46316	-0.71548	---	1.61
MIC4-149C	11.3	-0.17	-0.24202	-0.08837	-0.45006	-0.70437	---	1.73
MIC5-9A	10.87	-0.39	-0.20485	-0.22466	-0.53899	-0.73438	---	1.38
MIC5-13A	10.99	-0.45	-0.21592	-0.3094	-0.57575	-0.71864	---	1.10
MIC5-21A	10.76	-0.45	-0.12532	-0.28877	-0.55346	-0.71768	---	1.25
MIC5-42A	10.67	-0.43	-0.11933	-0.29911	-0.521	-0.6867	---	1.34
MIC5-42C	10.75	-0.43	-0.10988	-0.3012	-0.52616	-0.67401	---	1.38
COZZI 12X	10.35	-0.67	---	-1.02986	-2.12848	---	---	-0.67
COZZI 12X-HT	10.37	-0.6	---	-0.97136	-2.2063	---	---	-0.77
COZZI 12XP3	10.35	-0.28	---	-0.92568	-1.9363	---	---	-2.22
COZZI 12XP3-HT	10.03	-0.25	---	-0.9473	-2.08442	---	---	-2.50
COZZI 12XP5	no data	no data	---	no data	no data	---	---	no data
COZZI 12XP5 HT	no data	no data	---	no data	no data	---	---	no data
COZZI 12XP5	9.74	-0.3	---	-0.96009	-2.32591	---	---	-2.02
COZZI 12XP5-HT	9.71	-0.29	---	-0.90578	-2.23392	---	---	-2.08
PFP-1	.no data	no data	no data	no data	no data	. no data	no data	no data
PFP-2	.no data	no data	no data	no data	no data	. no data	no data	no data
PFP-3	.no data	no data	no data	no data	no data	. no data	no data	no data
PFP-4	.no data	no data	no data	no data	no data	. no data	no data	no data

Table V. Borosilicate Glasses Containing Li_2O and Exhibiting CPS

P_2O_5 (wt%)	B_2O_3 (wt%)	ΣM_2O (wt%)[‡]	SiO_2/B_2O_3	Major Alkali	CPS Phase	Glass + Melt Temp(°C)	Ref.
1.94-4.0	5.68-10.65	15.83-25.74	3.56-8.09	Na_2O + Li_2O	8 with Li_3PO_4 + 3 with SiO_2 or Silicate	11 M-Area MLLW Glasses Various Temps	This study
3.62-13.12	4.37-13.13	14.10-25.59	2.60-12.00	Li_2O + Na_2O	24 with Li_3PO_4 and Li_2NaPO_4	24 Idaho HAW Glasses @ 1150°C	This study; Staples, et. al (1998)[Error! Bookmark not defined.]

5.1 CRYSTALLINE PHASE SEPARATION IN MIXED WASTE GLASSES: PILOT SCALE TESTING

A large amount of data on actual mixed waste glasses containing 1-5 wt% U_3O_8 was generated by SRTC while performing a mixed (hazardous and radioactive) waste treatability study which included a pilot scale melter demonstration. In particular, several tanks of this mixed waste in the Savannah River Site (SRS) M-Area contained high phosphate, while several tanks were almost phosphate free. A pilot scale melter run was performed under funding from the Mixed Waste Focus Area (EM-50).

The melter campaign was initiated with low phosphate tank waste. Subsequently, high phosphate containing waste was fed to the melter. The amount of phosphate in the final glass was analyzed chemically and the glasses were analyzed by XRD. The glasses made at the beginning of the campaign had 2.40 wt% P_2O_5 before the high P_2O_5 tank waste was fed to the melter. The 2.4 wt% P_2O_5 glass exhibited no crystalline phases at the beginning of the melter run (Figure 6a). As the campaign progressed, glasses containing ≥2.76 wt% P_2O_5 began to crystallize Li_3PO_4 upon cooling indicating that the crystalline phase separation begins at ~2.6 wt% P_2O_5 (see Table I and Figure 6a horizontal line) even when the Al_2O_3 content reached ~15 wt% (Figure 6b). This indicates that high Al_2O_3 does not suppress CPS in these phosphate containing glasses as it does in borosilicate systems that undergo APS.[13]

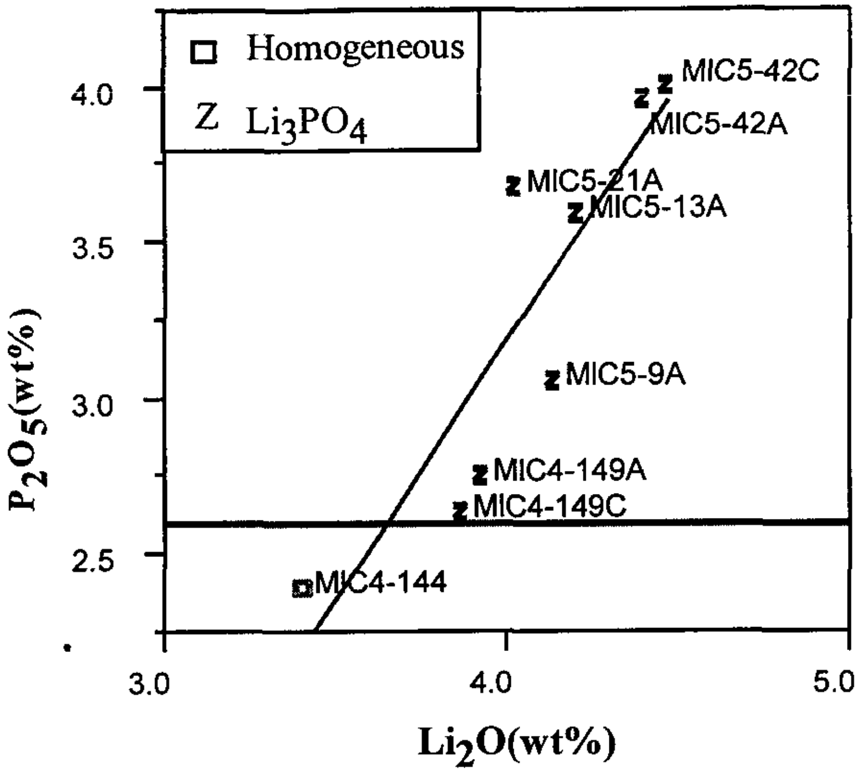

Figure 6a. Phosphate glass crystalline phase separation (CPS) began in glasses produced during a pilot scale melter runs of a mixed waste as a high P_2O_5 containing feed was fed to a melter of homogeneous glass. CPS started at about 2.6 wt% P_2O_5 the horizontal line indicated on the figure.

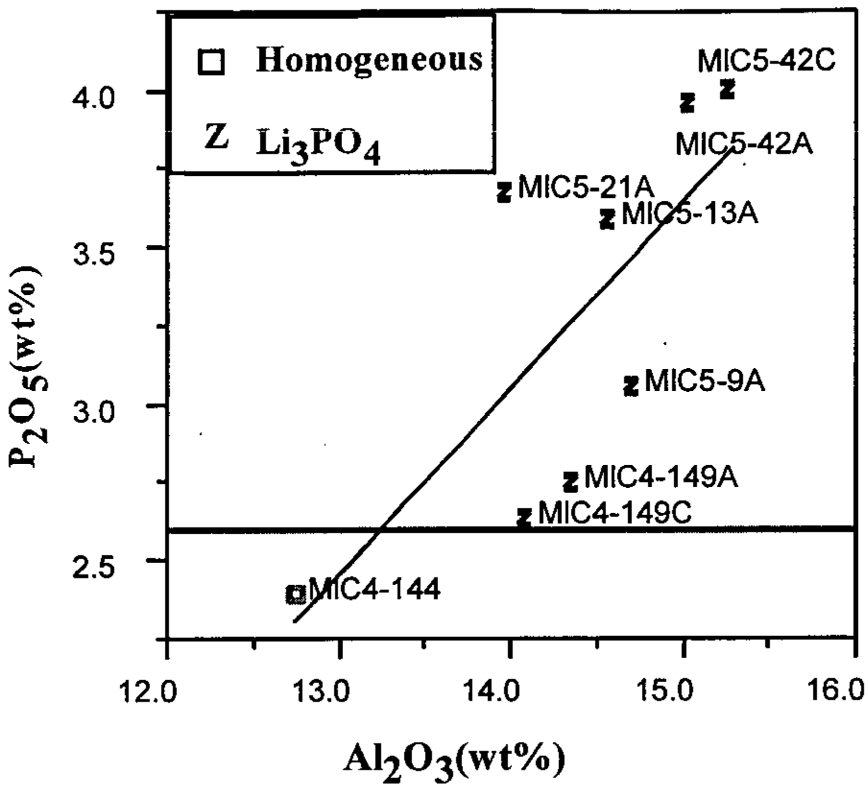

Figure 6b. Alumina contents as high as 15 wt% did not stabilize the glass against the CPS as it does in borosilicate glasses undergoing APS.

5.2 CRYSTALLINE PHASE SEPARATION IN BOROSILICATE GLASSES: CRUBIBLE AND PILOT SCALE TESTING

Analysis of the chemistry of the 83 crucible study glasses from the M-Area, INEEL, and the Cozzi studies were combined with the 8 glasses from the M-Area melter run. The combined data (91 glasses) supported the data from the melter campaign: glasses with greater than ~2.6 wt% P_2O_5 underwent some type of CPS (see Table II and Figure 7). Only eight glasses were reported to be homogeneous at >2.6 wt% P_2O_5: four of the simple glasses from the Cozzi study (shown in the ellipses in Figure 7a and 7b), three INEEL glasses (IG1-14, -35, -43), and one M-Area glass (MN-15). Conversley, several glasses were reported to have CPS (Z,X, and Y on Figure 7) at <2.6 wt% P_2O_5. These glasses were two of the Cozzi study simple glasses that crystallized only SiO_2 (12X and 12X-HT indicated by a Y in Figures 7a and 7b), one INEEL glass (IG1-30) that exceeded its solubility limit for ZrO_2, and two M-Area glasses (MN-5 and MHSi-11) which exceeded the solubility limit for Al_2O_3 (one crystallized α-Al_2O_3 while the other crystallized $LiAlO_2$).

The data shown in Figure 7 indicates that the Cozzi study simple glasses are poor representations of the complexity of the CPS in complex waste glasses. This may be attributed to the fact that the Cozzi study glasses contained only one alkali (Na_2O) while the INEEL and M-Area waste glasses contain complex combinations of alkalis and alkaline earth oxides. For the discussion of the effects of P_2O_5 type CPS on glass durability (Section 6.0), the 6 simple glasses, the IG1-30 glass, the MN-5, and the MHSi-11 glasses will be omitted from the database.

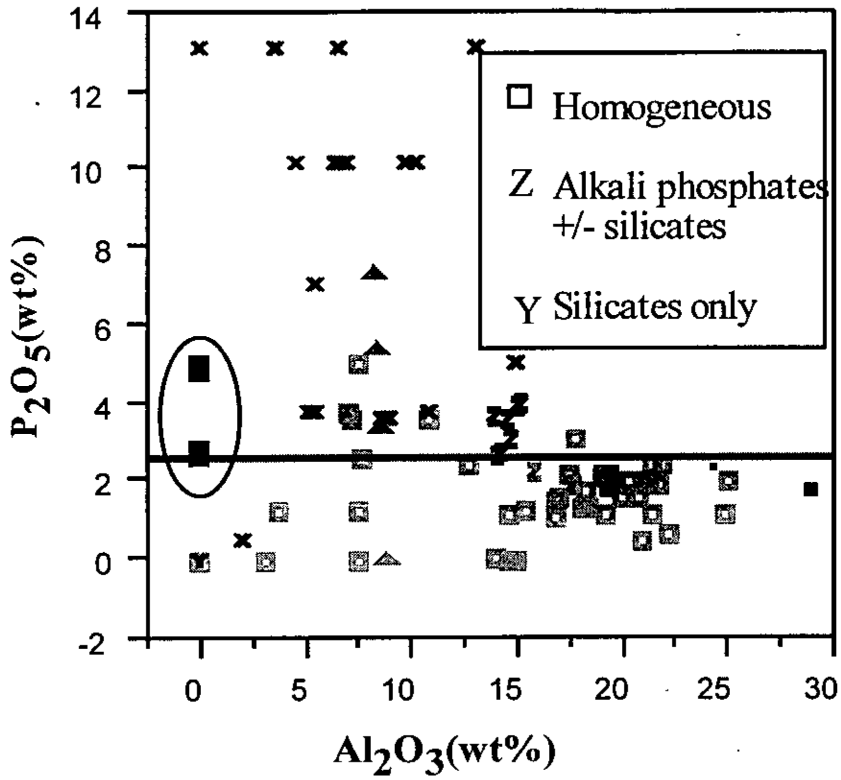

Figure 7a.Data from 91 borosilicate waste glasses containing varying amounts of P_2O_5 plotted against the amount of Al_2O_3 in each glass indicates that complex waste glasses with ≥2.6 wt% P_2O_5 (horizontal line) undergo crystalline phase separation of P_2O_5-rich and/or SiO_2-rich phases. The glasses in the ellipse are the simple four component glasses from Cozzi.[11]

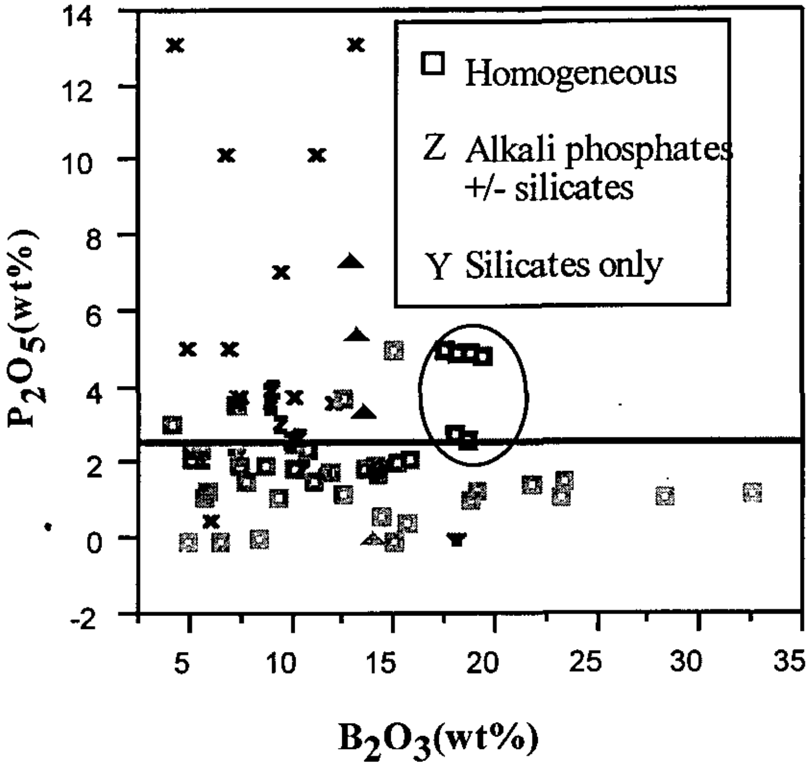

Figure 7a. Data from 91 borosilicate waste glasses containing varying amounts of P_2O_5 plotted against the amount of B_2O_3 in each glass indicates that complex waste glasses with ≥2.6 wt% P_2O_5 (horizontal line) undergo crystalline phase separation of P_2O_5-rich and/or SiO_2-rich phases. The glasses in the ellipse are the simple four component glasses from Cozzi.[11]

6.0 EFFECTS OF CRYSTALLINE PHASE SEPARATION ON GLASS DURABILITY

The relative durability of the 82 remaining P_2O_5 containing glasses in the database were examined as a function of their measured durability during PCT (ASTM C1285-97) testing. Normalized boron and sodium were chosen as the durability indicators because the slope relating these two parameters was ~1 (Figure 8) and the intercept was ~0 for the entire population of 82 glasses (R^2=0.76). This indicated that the B and Na were not involved in the CPS and could be used as a good indicator of the overall glass durability.

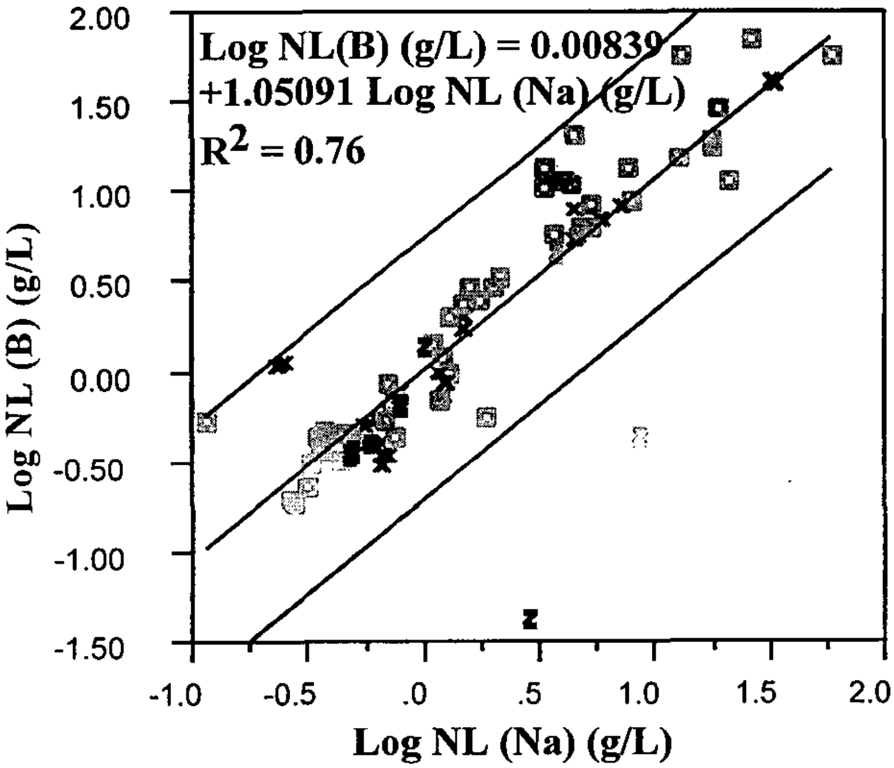

Figure 8. Plot of log NL(B) vs. log NL(Na) for 82 amorphous (indicated by squares) and CPS glasses (indicated by symbols x and z) showing that the leaching of the Na and B is congruent (slope of ~1 and intercept of ~0).

Conversely, a plot of the normalized release of boron verses the normalized release of lithium for the 44 glasses classified as homogeneous (visually and/or by x-ray analysis) gave a slope of less than unity, a non-zero intercept, and a very poor fit (R^2=0.44) indicating that Li was not a good indicator of overall glass durability (Figure 9a) for the homogeneous glasses or that some of the glasses classified as "homogeneous" were, indeed, phase separated (APS). The release of B and Li from the 37 glasses that had undergone CPS gave a slope of unity, a near zero intercept, and an R^2 of 0.73 (Figure 9b). Figure 9b indicates that Li is a good indicator of CPS. It also demonstrates that the normalized B release is somewhat greater than the Li release for the CPS glasses (perhaps since some Li was tied up in crystalline phases), making B a worst case indicator of the glass durability in the presence or absence of CPS.

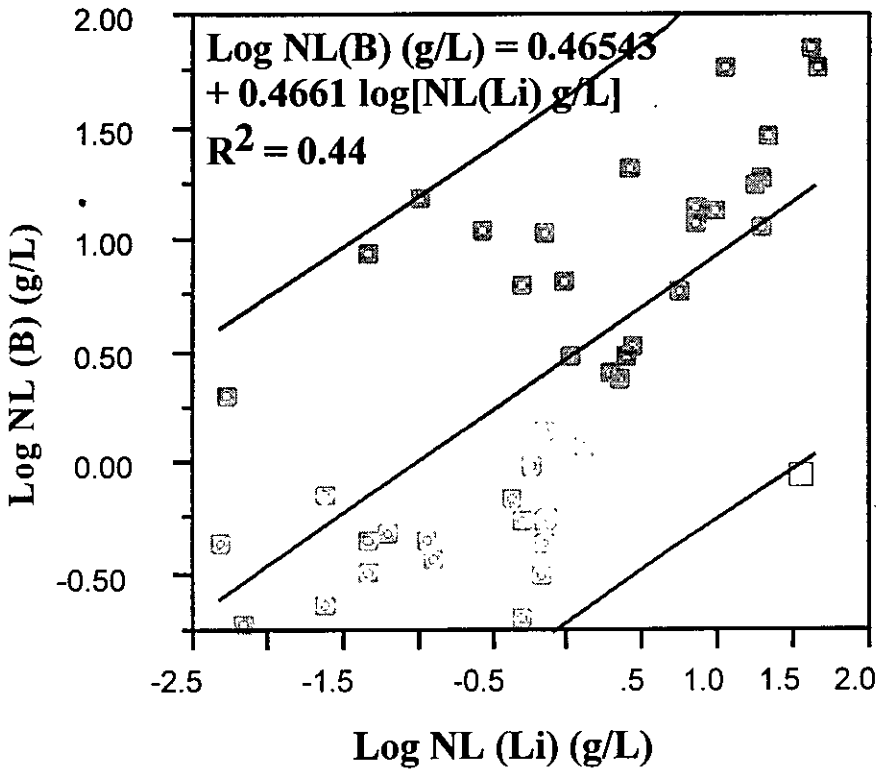

Figure 9a. Release of normalized B vs. Li for 50 homogeneous P_2O_5 glasses.

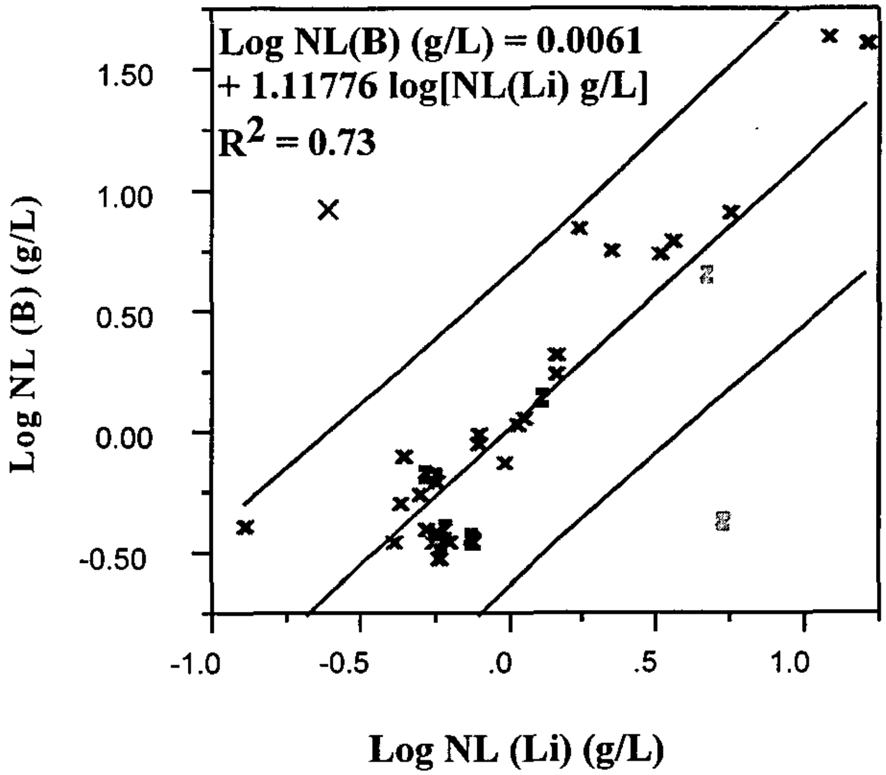

Figure 9b. Release of normalized B vs. Li for 37 crystallized CPS glasses.

Phosphate durability was available for the 44 glasses in the INEEL glass data set. There was no correlation between the log NL(P) in g/L and log NL(Li) in g/L for the homogeneous glasses. However, for the 24 of the 25 glasses undergoing CPS (one did not contain Li_2O as a component) there is a strong correlation between the normalized P and Li release (Figure 10). Therefore, normalized Li release can be used as an indicator of durability of the CPS phase (Figure 9b and 10) while normalized Na and B release can be used as an indicator of the overall glass durability (Figure 8)

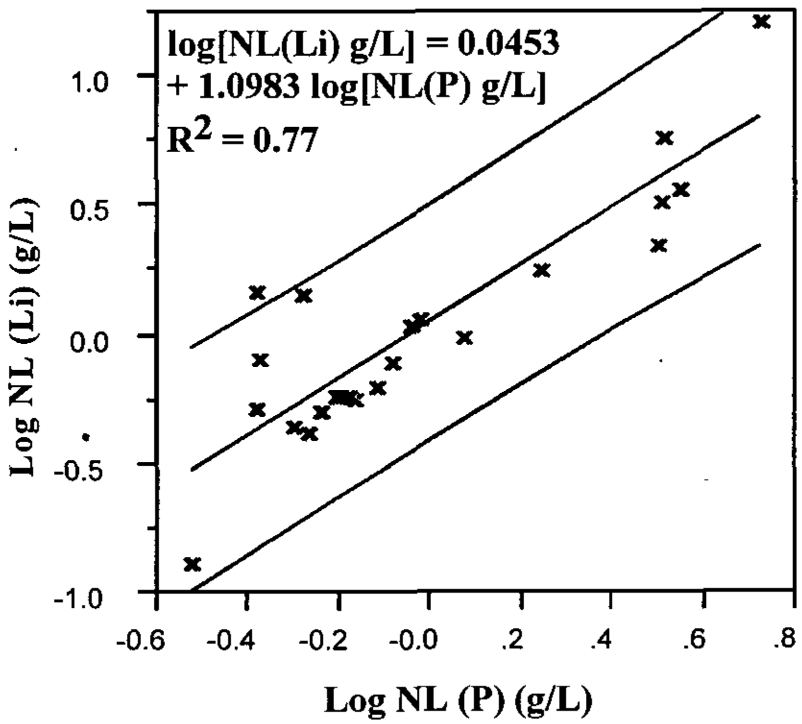

Figure 10. Release of normalized Li vs. P for 24 crystallized INEEL CPS glasses.

The durability of 82 P_2O_5 containing glasses was assessed against the $P_2O_5/(B_2O_3+SiO_2)$ molar ratio of the glasses (Figure 11). The distribution of the glass populations were thought to be the following:

- 32 had undergone CPS of a phosphate phase identified by XRD
- 50 visually homogeneous or homogeneous by XRD

It was recognized that many of the glasses that were classified as "homogeneous" visually or by XRD, may have undergone amorphous phase separation. The durability response shown in Figure 11 indicates that many of the high P_2O_5 containing CPS glasses were more durable than the low P_2O_5 containing homogeneous glasses circled in the figure. Many glasses classified as homogeneous were poorly durable and performed at or above EA glass limit in terms of their normalized B release (Figure 11a). Only a few of the high P_2O_5 containing CPS glasses were poorly durable and performed at or above EA glass limit (Figure 11a). The CPS does not appear to be the cause of the adverse effects on the waste glass durability (see discussion in next section).

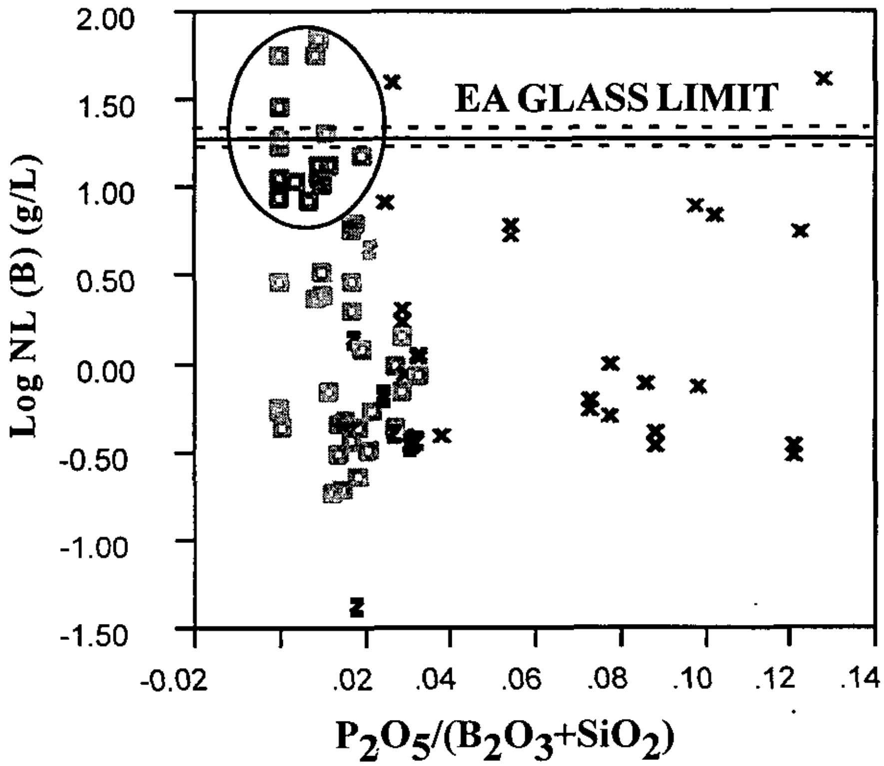

Figure 11a.The durability of the P_2O_5 containing borosilicate glasses as a function of the $P_2O_5/(SiO_2+B_2O_3)$ molar ratio. Borosilicate waste glasses that do not undergo CPS are shown as squares. Glasses that undergo CPS are indicated with darker text (x and z). Only a few CPS glasses and many APS glasses are less durable than the Environmental Assessment (EA) glass. The horizontal lines are the EA glass mean durability response[14] and the upper and lower 2σ durability limits.[15]

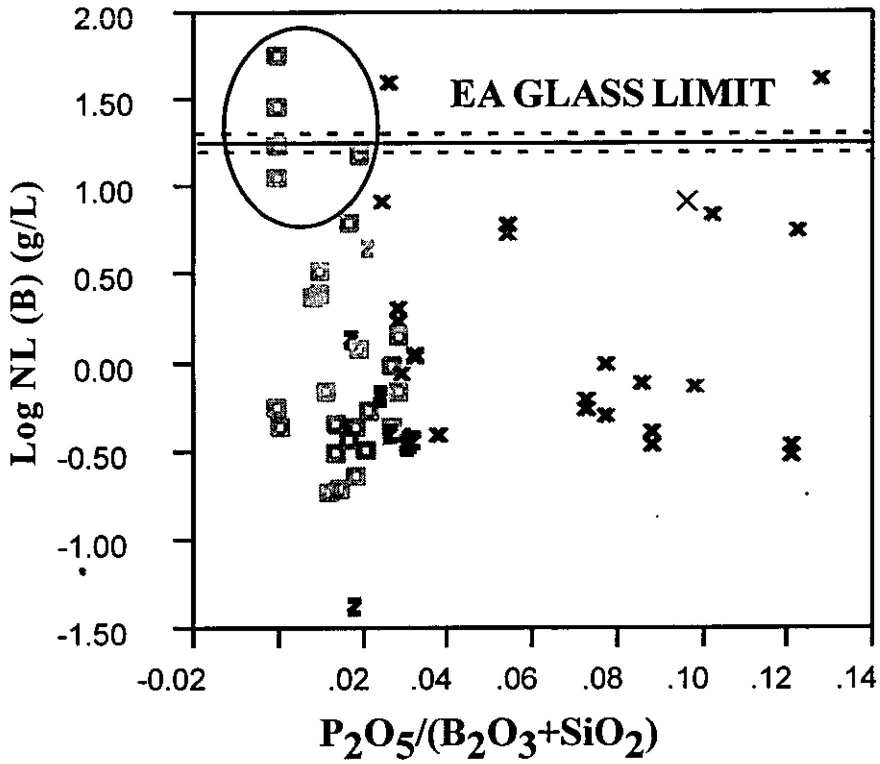

Figure 11b. Borosilicate glasses with ≥14 wt% B_2O_3 have been removed (20 glasses in all) since the data of Tovena et.al.[4] indicates that glasses with high B_2O_3 content undergo APS. This leaves only a few glasses within the ellipse that are less durable than the EA glass limit.

7.0 PREDICTING GLASS DURABILITY FOR HIGH P_2O_5 CONTAINING GLASSES WITH CRYSTALLINE PHASE SEPARATION (CPS)

The data of Tovena, et. al.[4] indicates that borosilicate glasses undergo APS when the B_2O_3 content of a glass is ≤2 wt% or between 10 and 15 wt%. If the glasses with ≤2 wt% B_2O_3 and ≥14 wt% B_2O_3 glasses are removed from the 82 glass database, then most of the poorly durable glasses within the ellipse shown on Figure 11a are removed (compare Figure 11a to 11b). Sixty-two glasses with B_2O_3 ranges between 4.15 and 13.7 remain in Figure 11b, e.g. glasses with SiO_2/B_2O_3 ratio (by wt%) of <3.81 are excluded (see Tables I and V). This removes most of the poorly durable glasses which are suspected to have undergone APS. These glasses all have ≤ 2.87 wt% P_2O_5. The glasses remaining in the poorly durable glass ellipse and above the EA glass limit shown in Figure 11b are poorly durable homogeneous glasses.

It is well known that glass compositions with less total alkali undergo less ion exchange and are relatively more durable. Glasses with high excess alkali create excess strong base in the leachates during static durability testing. Leachates with less alkali remain buffered in terms of leachate strong bases [SB] and weak acids [WA]. Plotting the [SB]-[WA] equilibria values from Table 2 for the remaining 62 glasses in Figure 12 demonstrates that glasses with high alkali are the glasses that have high leachate pH values where excess strong base [SB] is the dominant leachate species. Figure 12 demonstrates that the poor durability of the glasses in the ellipse is due to the excess [SB] and not the CPS.

For the DWPF, glasses with ≤19.3 wt% total alkali ($Na_2O + Li_2O + Cs_2O + K_2O$) have been found to remain durable because the leachates are buffered in terms of their [WA]-[SB] equilibria. Imposing the DWPF total alkali constraint to the remaining 62 glasses in the database demonstrates that the P_2O_5 containing borosilicate glasses remaining are very durable. Indeed, the usage of the 19.3 wt% total alkali DWPF limit appears to be overly conservative for these high Al_2O_3 and high ZrO_2 containing glasses as only 21 of the original 62 glasses in the database remain (Figure 13a). Optimizing the total alkali constraint for the P_2O_5 containing high Al_2O_3 and high ZrO_2 borosilicate glasses suggests a limit of ~22 wt% total alkali. This defines a durable glass population of 35 of the original 62 glasses (Figure 13b): some glasses are homogeneous (squares) and some glasses have undergone CPS (symbols x and z).

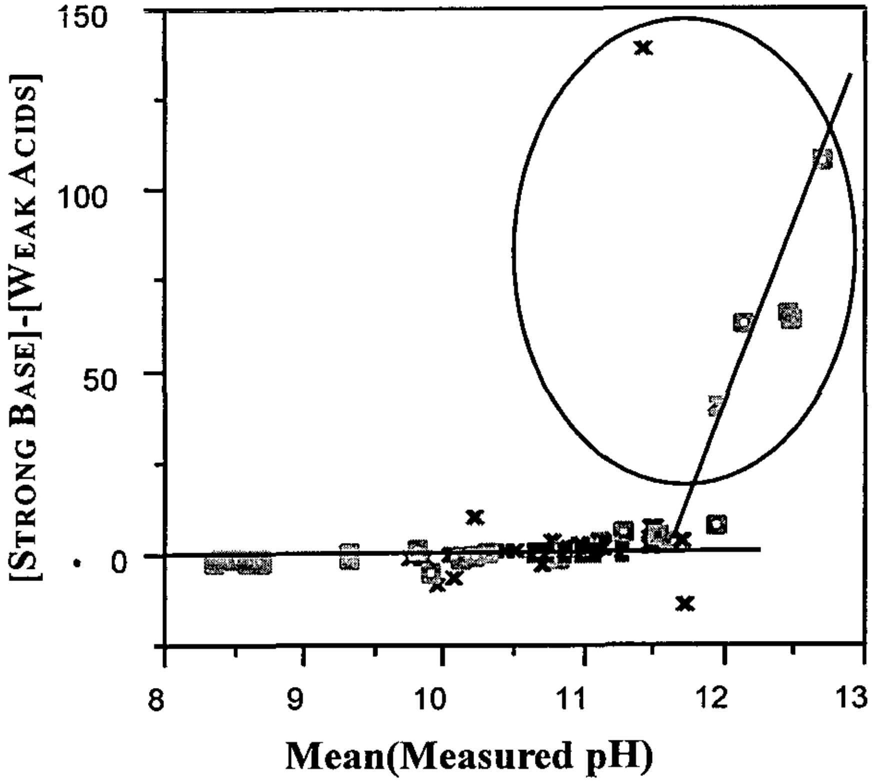

Figure 12. Glass durability is a function of the strong base and weak acid equilibria in the leachate and not a function of the CPS. Glasses indicated with squares are homogeneous. Glasses indicated with the symbols x and z have undergone CPS. Glasses in the ellipse have excess strong base, [SB], and are poorly durable.

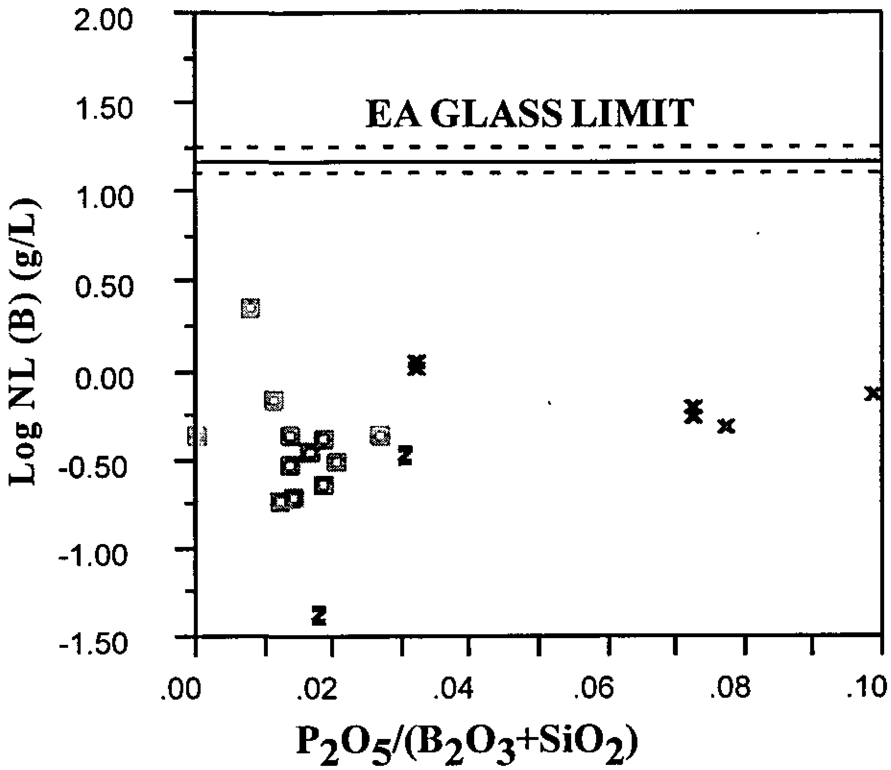

Figure 13a.Durable glass population when Σalkali oxides is limited to 19.3 wt%. Squares are homogeneous glasses while the symbols x and z indicate glasses that have undergone CPS.

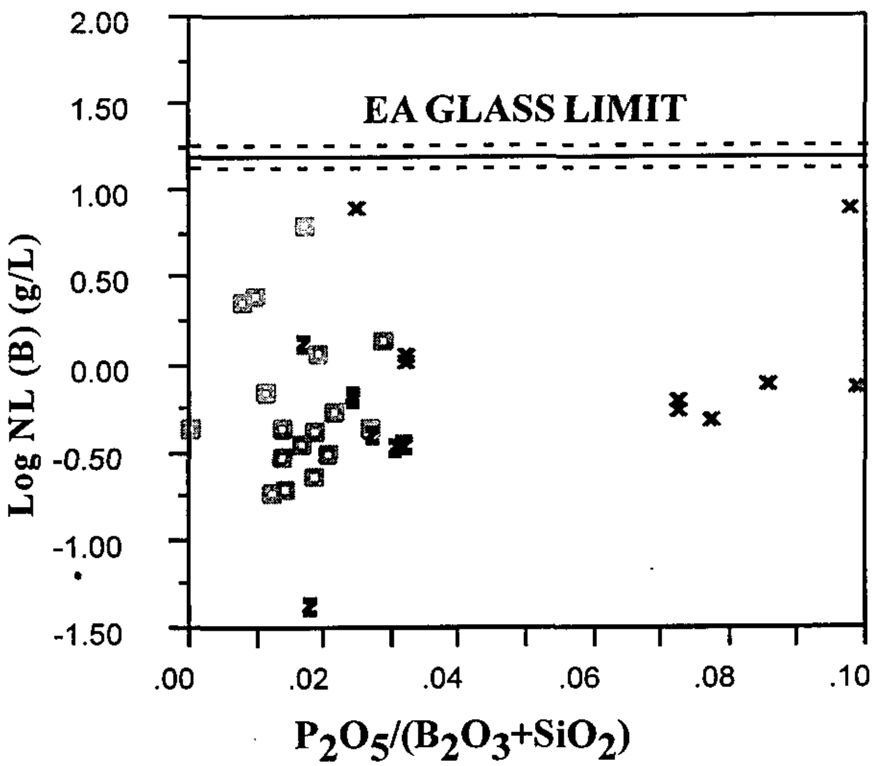

Figure 13b.Durable glass population when Σalkali oxides is limited to 22 wt%. Squares are homogeneous glasses while the symbols x and z indicate glasses that have undergone CPS.

The glass durability is a complex function of the B_2O_3 and total alkali in the glass. If the B_2O_3 limit causing APS is lowered from 14 wt% to 10 wt% then the total alkali limit can be raised from 22 wt% to 24 wt% and the same glass durability retained. In order to represent this graphically, the log normalized B release from Table II for the 82 glasses shown in Figure 11a were contoured (Figure 14a) on a plot of total alkali vs. B_2O_3 in wt%. Most of the durable glasses fall in the lower left quadrant defined by the horizontal line at 24 wt% total alkali (R_2O) and the vertical line 10 wt% B_2O_3.

Values of log NL(B) that are ≥1.0g/L are shown in Figure 14b. These values of log NL(B) are at or above the EA glass limit of 1.22 g/L or its lower 95% confidence band. The glasses contain $R_2O \geq 24$ wt% R_2O and ≥ 10 wt% B_2O_3. Note that there are glasses shown in Figure 14b that have acceptable durabilities (≤1.0 g/L log normalized B) at > 10 wt% B_2O_3. However, if the B_2O_3 value is raised to 14 wt% B_2O_3, then the R_2O level has to be lowered to 22 wt% R_2O to avoid the poorly durable glasses releasing 1.19 and 1.06 g/L log normalized B shown in Figure 14b.

The resulting durable composition domain for P_2O_5 containing borosilicate glasses, whether they are homogeneous or have undergone CPS, is given in Table VI as an indication of new composition regions to be assessed for INEEL waste glasses. In order to avoid glasses with excess [SB] that have poor glass durability the R_2O limit recommended in this paper is ≤22 wt% with a B_2O_3 limit of ≤14 wt%.

Table VI. Compositional Region Defined for Durable High P_2O_5 Containing Borosilicate Glasses Including Homogeneous and CPS Glasses

Glass Compositional Component(s)	Ranges (wt %)	Reason
B_2O_3	≥2 wt% and ≤14wt%	Avoid APS
$Na_2O + Li_2O + Cs_2O + K_2O$	≤22 wt%	Avoid poorly durable glasses that form excess [SB] during durability test
P_2O_5	0.06-13.25	Range tested in this study
Al_2O_3	0-25	Range tested in this study
ZrO_2	0-15	Range tested in this study
$Al_2O_3 + ZrO_2$	≥9	To improve durability and stabilize glass against APS in presence of excess alkali and B_2O_3

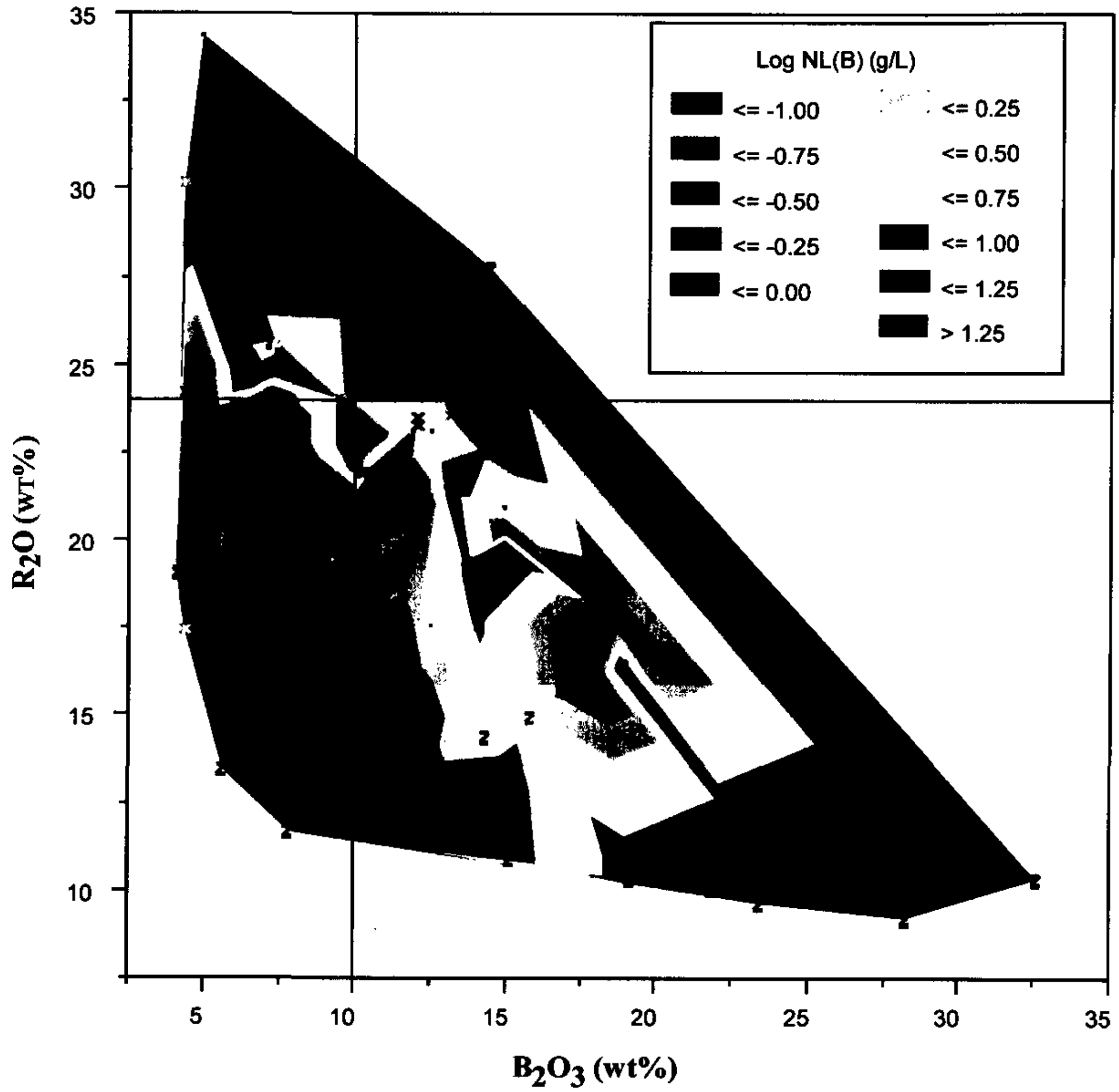

Figure 14a.Contour plot of the PCT leachate response, log normalized B in g/L, demonstrating that values of ≤1.0 g/L are released from P_2O_5 containing glasses with <10 wt% B_2O_3 and ≤24 wt% R_2O where $R_2O = \Sigma\ Na_2O + Cs_2O + K_2O$.

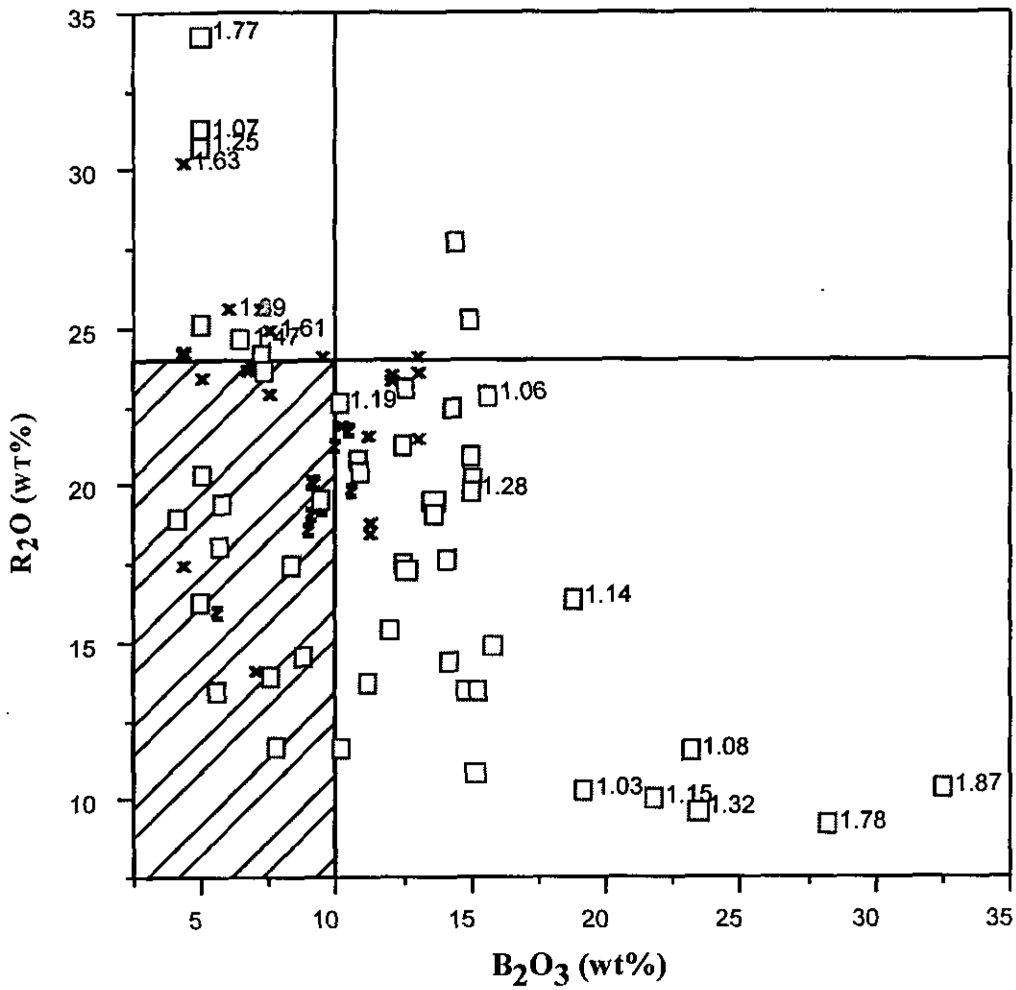

Figure 14b.Individual PCT leachate responses, log normalized B in g/L, demonstrating that values of ≤1.0 g/L are released from P_2O_5 containing glasses with <10 wt% B_2O_3 and ≤24 wt% R_2O where $R_2O = \Sigma Na_2O + Cs_2O + K_2O$. Note that if the B2O3 limit is raised to 14 wt% that the R2O must be lowered to 22 wt% to avoid the nondurable glasses releasing 1.19 and 1.06 log normalized B, respectivley. Glasses indicated with a square are homogeneous glasses while glasses indicted with a z or x have undergone crystalline phase separation. The shaded lower left quadrant defines the compositional region of durable glasses regardless of whether the glass is homogeneous or has undergone CPS.

8.0 CONCLUSIONS

- Two types of phase separation occur in the borosilicate waste glasses studied: amorphous phase separation (APS) is predicted in high B_2O_3 containing borosilicate waste glasses with ≤ 2.6wt% P_2O_5 while crystalline phase separation (CPS) occurs in borosilicate waste glasses with >2.6 wt% P_2O_5.

- In P_2O_5 the borosilicate waste glasses studied, the major glass constituents (alkalis and boria) control the glass durability and not the CPS.

- Based on the normalized B and Na release, APS appears to be detrimental to waste glass durability (as determined previously) while CPS does not.

- Due to the crystalline nature of the P_2O_5 phase separation and the fact that the CPS phases have little impact on glass durability, it is recommended that CPS should be treated as a crystallization effect rather than as a phase separation effect for waste acceptance. It remains to be demonstrated that the radionuclides do not preferentially partition into the CPS phase and leach at a greater rate than the B or Na. It also must be shown that the formation of the phosphate rich phase does not adversely effect the processability of the glass.

- U_3O_8, the only radionuclide studied, did not partition into any CPS phases.

- Alumina was not found to stabilize the glass against CPS in the manner in which it stabilizes borosilicate glasses without P_2O_5 from APS.

- Alumina and zirconia were found to stabilize the CPS glass matrix against APS and stabilize the homogeneous glasses against APS: all INEEL glasses with a combined concentration of Al_2O_3 and $ZrO_2 \geq 9$ wt% were found to have adequate durability.

9.0 ACKNOWLEDGEMENT

This paper was prepared in connection with work done under Contract No. DE-AC09-96SR18500 with the U.S. Department of Energy. This work was supported by the Tank Waste Focus Area (TFA) under Technical Task Plan (TTP) SR1-6-WT-31. The M-Area Glass Vitrification Studies were supported by the Mixed Waste Focus Area (EM-50) under TTP

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Title: Crystalline Phase Separation in Phosphate Containing Waste Glasses: Relevance to INEEL HAW (U)	Key Words (list 3): high activity waste, vitrification

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U. S. Department of Energy - Savannah River Operations Office

Dear Ms. Perrin:

REQUEST FOR APPROVAL TO RELEASE SCIENTIFIC/TECHNICAL INFORMATION

The attached document is submitted for classification and technical approvals for the purpose of external release. Please complete Part II of this letter and return the letter to the undersigned by 9/29/2000. The document has been reviewed for classification and export control by a WSRC Classification staff member and has been determined to be Unclassified.

Kevin Schmidt, WSRC STI Program Manager

I. DETAILS OF REQUEST FOR RELEASE

Document Number:	WSRC-TR-2000-00339,
Author's Name:	C. M. Jantzen
Location:	773-A Phone 5-2374
Department:	SRTC/Immobilization Technology Section
Document Title:	Crystalline Phase Separation in Phosphate Containing Waste Glasses: Relevance to INEEL HAW
Presentation/Publication:	
Meeting/Journal:	OSTI Reportable
Location:	
Meeting Date:	

II. DOE-SR ACTION

Date Received by TIO 09/14/2000

- [X] Approved for Release
- [ ] Approved Upon Completion of Changes
- [ ] Approved with Remarks
- [ ] Not Approved
- [ ] Revise and Resubmit to DOE-SR

Remarks: ______

W. F. Perrin, Technical Information Officer, DOE-SR   9/21/00 Date

OSR 25-82-W# (4-89)
Stores: 26-15460.00

DOE F 241.1
(7-98) p. 1 of 2
(Formerly DOE F 1332.15)
(All other versions are obsolete)

OMB Control No
1910-1400

US DEPARTMENT OF ENERGY
ANNOUNCEMENT OF U. S. DEPARTMENT OF ENERGY (DOE) SCIENTIFIC AND TECHNICAL INFORMATION (STI)

RECORD STATUS (select one):
X New ...... Revised Data .... Revised STI Product

Part I: STI PRODUCT DESCRIPTION

A. STI PRODUCT TYPE (select one)

X 1. **Technical Report**
a. Type: ☐ Topical ☐ Semiannual ☐ Annual ☒ Final Other (specify)
b. Reporting Period (mm/dd/yyyy) ..../..../.... thru ..../..../....

.... 2. **Conference**
a. Product Type: .... Conference Proceedings .... Conference Paper or Other (abstracts, excerpts, etc.) ___
b. Conference Information (title, location, dates) ___

.... 3. **Software Manual** (*The actual software package should be made available simultaneously. Follow instructions provided with ESTSC F 1 and ESTSC F 2.*)

.... 4. **Journal Article**
a. Type: X Announcement citation only ___ Preprint ___ Postprint
b. Journal Name
c. Volume ___ d. Issue ___ e. Serial identifier (*e.g., ISSN or CODEN*) ___

.... 5. **S&T Accomplishment Report**

.... 6. **Book**

.... 7. **Patent Application**
a. Date Filed (*mm/dd/yyyy*) ___/___/___
b. Date Priority (*mm/dd/yyyy*) ___/___/___
c. Patent Assignee ___

.... 8. **Thesis/Dissertation**

B. STI PRODUCT TITLE Crystalline Phase Separation in Phosphate Containing Waste Glasses: Relevance to INEEL HAW

C. AUTHOR(s) C. M. Jantzen
E-mail Address(*es*):

D. STI PRODUCT IDENTIFIER
___ 1. Report Number(s) WSRC-TR-2000-00339, Rev. 0
___ 2. DOE Contract Number(s) DE-AC09-96SR18500
___ 3. R&D Project ID(s)
___ 4. Other Identifying Number(s)

E. ORIGINATING RESEARCH ORGANIZATION Savannah River Site

F. DATE OF PUBLICATION (*mm/dd/yyyy*) 9/21/2000

G. LANGUAGE (*if non-English*) English

(*Grantees and Awardees: Skip to Description/Abstract section at the end of Part I*)

H. SPONSORING ORGANIZATION ___

I. PUBLISHER NAME AND LOCATION (if other than research organization) ___

Availability (*refer requests to [if applicable]*)

J. SUBJECT CATEGORIES (*list primary one first*) 12

Keywords high activity waste, vitrification, phase separation, phosphate glass, waste glass

K. DESCRIPTION/ABSTRACT

As part of the Tanks Focus Area's (TFA) effort to increase waste loading for high-level waste vitrification at various facilities in the Department of Energy (DOE) complex, the occurrence of phase separation in waste glasses spanning the Savannah River Site (SRS) and Idaho National Engineering and Environmental Laboratory (INEEL) composition ranges have been studied. The type of phase separation that occurs in the phosphate rich borosilicate waste glasses, such as those investigated for INEEL, crystallizes upon cooling. This type of phase separation mechanism is less well studied than amorphous phase separation in phosphate poor borosilicate waste glasses. Therefore, the type of phase separation, extent, and impact of phase separation on glass durability for a series of INEEL-type glasses were examined and the data statistically analyzed in this study.

US DEPARTMENT OF ENERGY

ANNOUNCEMENT OF U. S. DEPARTMENT OF ENERGY (DOE) SCIENTIFIC AND TECHNICAL INFORMATION (STI)

DOE F 241.1 (p. 2 of 2)

Part II: STI PRODUCT MEDIA/FORMAT and LOCATION/TRANSMISSION

A. MEDIA/FORMAT INFORMATION

1. Medium of STI product is: ...... Paper ...... Electronic document ...... Computer medium ...... Audiovisual material
2. Size of STI product ________
3. File format:
   a. If electronic document is posted at site, indicate: ☒ html ☐ pdfn ☐ sgml ☐ xml PDF Normal PDF Image
   b. If electronic document is transmitted to OSTI, indicate: ☐ html ☐ pdfn ☐ pdfi ☐ msword PDF Normal
   ___ TIFFG4 ___ WP–indicate Version (5.0 or greater) ______ platform/operating system ______
   ___ MS Word–indicate Version (5.0 or greater) ______ platform/operating system ______ ___ Postscript
4. If computer medium or audiovisual material:
   a. Quantity/type (*specify*) ______
   b. Machine compatibility (*specify*) ______ c. Sound: ___ (*yes*) d. Color ___ (*yes*) e. Tables/Graphics ___ (*yes*)
   f. Other information about product format a user needs to know: ______

B. LOCATION/TRANSMISSION INFORMATION

1. STI Product is available at site: Unique URL (*of specific STI Product*) http://www.srs.gov/general/sci-tech/fulltext/tr2000339/tr2000339.html
2. STI Product is being transmitted to OSTI:
   a. ___ Electronically via FTP
   b. ___ Via Mail or shipment (*e.g., Federal Express*) (*Paper products, electronic documents on CD-ROM, diskettes, videocassettes, etc.*)
3. Information Product Filename (*of transmitted electronic format*) ......

C. ADDITIONAL INFORMATION (concerning media/format or location/transmission; for OSTI internal use only): ______

(Grantees and Awardees: Skip to Contact section at the end of Part III)

Part III: STI PRODUCT REVIEW? RELEASE INFORMATION

A. ACCESS LIMITATION

X..... 1. **Unlimited Announcement** (*available to U.S. and non-U.S. public*)

...... 2. **OpenNet** (*use OpenNet guidance for below*):
   a. If additional source other than making it available through NTIS:
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      (2) Document Location ______
   b. Field Office Acronym ______
   c. Declassification date (mm/dd/yyyy) __/__/____
   d. Declassification Status: ___ Declassified ___ Sanitized ___ Never classified
   e. OpenNet Document Type ______
   f. OpenNet Document Keywords ______
   g. OpenNet Addressee ______

...... 3. **U.S. Dissemination Only**

...... 4. **Copyrighted material**; Are there any restrictions based on copyright? ___ yes ___ no. If yes, list the restrictions ______

...... 5. **Small Business Innovation Research** (*SBIR*) Release date (mm/dd/yyyy) ....../....../......

...... 6. **Small Business Technology Transfer** (*STTR*) Release date (mm/dd/yyyy) ....../....../......

...... 7. **Proprietary/Trade Secret**

...... 8. **Patent Pending**

...... 9. **Protected data** ___ CRADA ___ Other (specify) ______ Release date (mm/dd/yyyy) ....../....../......

...... 10. **Official Use Only** (*OUO*)

...... 11. **Program-Directed Special Handling** (*specify*) ______

...... 12. **Export Control/ITAR/EAR**

...... 13. **Unclassified Controlled Nuclear Information** (*UCNI*) ______

...... 14. **Classified** Classification Level/Category of:
   a. This form U
   b. The STI Product Unclassified

15. **Other information relevant to access** (*specify; for OSTI internal use only*) ______

B. OTHER (*information useful to include in published announcement record which is not suited for any other field on this form*)

C. CONTACT AND RELEASING OFFICIAL

1. Contact (*if appropriate, the organization or site contact to include in published citations who would receive any external questions about the content of the STI Product or the research information contained therein*)

Name and/or Position Kevin Schmidt, Manager STI Program & Site Support

E-mail ______ Phone (803) 725-2765

Organization Westinghouse Savannah River Company

2. Releasing Official X I verify that all necessary reviews have been completed (e.g. Patent, Copyright, ECI, UCNI, etc.)

Released by (*name*) Kevin Schmidt Date (*mm/dd/yyyy*) 9/21/2000

E-Mail ______ Phone (803) 725-2765