Component effects on crystallization of RE-containing aluminoborosilicate glass

Syazwani Mohd Fadzil,^{a,b1} Pavel Hrma,^{a,c} Michael J. Schweiger,^c & Brian J. Riley^c

^a*Division of Advanced Nuclear Engineering, Pohang University of Science and Technology, 790784 Pohang, South Korea*

^b*School of Applied Physics, Faculty of Science and Technology, The National University of Malaysia, 43650 Bandar Baru Bangi, Selangor, Malaysia*

^c*Pacific Northwest National Laboratory, P.O. Box 999, Richland, Washington, United States*

ABSTRACT

Lanthanide-aluminoborosilicate (LABS) glass is one option for immobilizing rare earth (RE) oxide fission products generated during reprocessing of pyroprocessed fuel. This glass system can accommodate a high loading of RE oxides and has excellent chemical durability. The present study describes efforts to model equilibrium crystallinity as a function of glass composition and temperature as well as liquidus temperature (T_L) as a function of glass composition. The experimental method for determining T_L was ASTM C1720-11. Typically, three crystalline phases were formed in each glass: Ce-borosilicate ($\text{Ce}_3\text{BSi}_2\text{O}_{10}$), mullite ($\text{Al}_{10}\text{Si}_2\text{O}_{19}$), and corundum (Al_2O_3). Cerianite (CeO_2) was a common minor crystalline phase and Nd-silicate ($\text{Nd}_2\text{Si}_2\text{O}_7$) occurred in some of the glasses. In the composition region studied, T_L decreased as SiO_2 and B_2O_3 fractions increased and strongly increased with increasing fractions of RE oxides; Al_2O_3 had a moderate effect on the T_L but, as expected, it strongly affected the precipitation of Al-containing crystals.

Keywords: lanthanide borosilicate glass, pyroprocessing, liquidus temperature, crystalline phases

1. Introduction

The Korea Atomic Energy Research Institute as well as Korean Hydro and Nuclear Power are focusing research and development efforts on radioactive waste management in Korea [1]. The research covers waste treatment, disposal of high-level waste (HLW) as well as low- and intermediate-level wastes, and interim storage of spent nuclear fuel (SNF). The objectives are focused on waste treatment and volume reduction technology, low-level waste vitrification technology, low- and intermediate-level waste disposal and safety assessment technology, improvement of existing technology for SNF storage and transportation, and development of advanced SNF storage technology [1].

¹Corresponding author. Tel.: +6-03-89215929; fax: +6-03-89213777.
E-mail address: syazwanimf@ukm.edu.my (S. Mohd Fadzil).

After the Fukushima accident (March 11, 2011), SNF management has been an issue because, currently, SNF is kept in at-reactor storage that will reach maximum capacity in 2024. In 2008, the SNF management decided to develop pyroprocessing, a reprocessing technology for SNF with minimal proliferation risk [2,3]. Instead of direct disposal of SNF, which is a low-cost option, the fuel will be recycled by a treatment that increases resource utilization.

Korea is participating in collaborative projects using pyroprocessing technology [1]. According to the United States–Republic of Korea agreement on pyroprocessing, the first stage of the Korea Advanced Pyroprocessing Facility is planned for 2021 with commercial-scale demonstration by 2025 [1]. The technology has been under development at the Korea Atomic Energy Research Institute since 1997 and was tested in a pilot-scale study in 2007. The pyroprocessing technology is used to recycle SNF to integrate useful resources and reduce the waste volume in the repository by a factor of 100. The process includes pre-treatment, electroreduction, electrorefining, electrowinning, and waste salt treatment [1,3].

Pyroprocessing is a method for separating uranium and transuranic elements in an electroreduction process to be recycled in fast reactors as fuel [4,5]. During this process, the rare earth (RE) fission products, now separated for waste processing, are released into the electrorefiner and converted to chlorides [4-6]. The RE chlorides can be selectively turned into a mixture of oxides and oxychlorides by oxygen sparging [7]. The RE oxide/oxychloride mixture will have to be immobilized in a stable waste form such as a glass [8-10].

In a previous study, a series of borosilicate glasses were fabricated with several RE oxides representative of RE fission products found in the electrorefiner following pyroprocessing [10]. The liquidus temperatures (T_L) and chemical durability of these glasses were determined. In this work, lanthanide-aluminoborosilicate (LABS) glasses were chosen for their high waste loading and excellent durability. A similar glass with SrO included in the composition was proposed for the immobilization of americium/curium waste stored at the Savannah River Site in F-Canyon Tank 17.1 [11,12].

Waste glass compositions vary widely and so do waste glass properties, including crystal types, crystal fractions, and the T_L . Depending on the melter type, both crystal fractions and the T_L can limit the waste loading because crystal formation can change the viscosity of the melt, can result in clogging of the melter pour spout, and can pull out critical glass-forming components from the melt, thus can negatively affect chemical durability. Generally, the processing technology that allows a high fraction of crystallinity can produce durable waste forms with high waste loading. Unlike the conventional Joule-heated melters that operate at $\sim 1150^\circ\text{C}$, an advanced melter technology such as a

cold crucible induction melter can operate at temperatures as high as $\sim 1700^{\circ}\text{C}$, thus possibly avoiding crystallization problems altogether [22,23].

In this work, we designed a matrix of LABS glasses and experimentally determined their equilibrium crystal fractions as functions of temperature. Typically, test matrices to explore the glass composition regions include varying one chemical component at a time, replacing one component with another, and using a statistical design to vary several components at a time [20,21]. The experimental approach in this work is centered on the one-at-a-time variation of each component. Though this approach does not provide information about interactive effects of the components, it represents the simplest design for obtaining component effects over a large composition region and allows for helpful graphical visualization of experimental data. To relate equilibrium crystal fractions and the T_L to glass composition, we fitted to data an empirical model that is based on chemical thermodynamics. Various models for crystallinity as a function of glass composition and temperature, and T_L as a function of glass composition have been used for HLW glasses with various primary crystalline phases [12-21].

2. Methodology

Experimental steps for this study consisted of glass fabrication followed by heat treatment of glass samples to determine T_L by the crystal fraction extrapolation method. More details of the process are found in the ASTM procedure described elsewhere [24]. Analytical techniques used were optical microscopy, scanning electron microscopy, energy dispersive spectroscopy, and X-ray diffraction.

2.1 Glass fabrication

The 250-g batches of lanthanide-aluminoborosilicate (LABS) glasses (Table 1) were made from fine powders consisting of oxides (i.e., SiO_2 , Al_2O_3 , CeO_2 , and Nd_2O_3) and boric acid. The batches were mixed and homogenized in an agate vibratory mill for several minutes. Batches were melted at 1325 to 1475°C for 1 h in a Pt/10%Rh crucible covered tightly with a Pt lid. The melting temperatures (T_m) used in this study were determined by visual observations where a temperature of 1325°C was used and if the glass had not melted after a few minutes, the temperature was increased iteratively until the melt appeared.

The melts were poured on an Inconel[®] plate to air quench. The cooled glasses were ground in a tungsten carbide mill for several minutes, remelted for 1 h, and then quenched under the same conditions. Glass melting temperatures were selected within the range of 1300 to 1500°C to facilitate processing in a cold crucible induction melter. Glasses were observed with an optical microscope to determine if phase separation or crystallization occurred to verify that all glasses were free of crystals prior to testing. No attempt was made to measure the glass compositions after melting because the

glasses were batched using analytical grade chemicals and melted in covered crucibles. No volatility of glass components was observed during melting.

Table 1 summarizes the glass compositions organized into five series by varied compositions. The baseline glass, L-53, was selected for high waste loading (53 mass%). Altogether, 21 homogeneous glasses were produced including baseline glass (L-53) by adding or removing 3 and 5 mass% of each component one at a time. The “H” and “L” letters in the glass identifications denote “high” and “low,” representing an increase or decrease of a component fraction from baseline composition, respectively, while “S,” “B,” “A,” “C,” and “N” letters denote the SiO₂, B₂O₃, Al₂O₃, CeO₂, and Nd₂O₃ oxide component, respectively. The numbers “3” and “5” at the end of each glass nomenclature denote the amount of each component changed from the baseline in mass%. The melting temperature was varied based on glass composition. The T_m values and waste (RE) loadings (in mass%) are included in Table 1, and component composition ranges are listed in Table 2.

2.2 Equilibrium crystal fractions, liquidus temperature, and phase analysis

Liquidus temperature was measured following ASTM C1720-11, specifically the crystal fraction extrapolation method [24]. Glass samples were heat-treated for 24±2 hrs at temperatures between 900°C and 1200°C. Selected samples were heat treated in duplicate at different times for reproducibility (the results were included as separate data points). After each heat treatment, the samples were air quenched and a portion was crushed in a tungsten carbide mill to a fine powder with a known amount of CaF₂ as an internal standard. The powdered samples were analyzed with a Bruker® D8 Advance (Bruker AXS Inc., Madison, WI) X-ray diffractometer so that the crystalline phases could be identified using the DIFFRAC^{plus} EVA (Bruker AXS) and quantified with whole-pattern fitting using TOPAS 4.2 (Bruker AXS) software. The uncertainty of temperature was around ±3°C while the crystal fraction extrapolation method has around ±0.5 mass% error.

2.3 Models

The equilibrium crystal fraction is generally not a linear function of temperature even for ideal binary mixtures for which

$$c = c_{\max} \left\{ 1 - \exp \left[-B_L \left(\frac{1}{T} - \frac{1}{T_L} \right) \right] \right\} \quad (1)$$

where c is the equilibrium crystal fraction, and $c_{\max}$, B_L , and T_L are coefficients that are independent of temperature, but are functions of composition.

Commonly, several crystalline phases coexist in a glass at equilibrium. If Equation (1) can fit c versus T data for more than one crystalline phase, we need to use a subscript for each phase:

$$c_j = c_{\max,j} \left\{ 1 - \exp \left[-B_{Lj} \left(\frac{1}{T} - \frac{1}{T_{Lj}} \right) \right] \right\} \quad (2)$$

where the subscript j stands for the j -th crystalline phase and c_j is the j -th crystalline phase mass fraction. Here, T_{Lj} , is the j -th crystalline phase “liquidus temperature,” where the quotes apply when the j -th crystalline phase is not the primary phase.

For a reasonably small composition region, such as that listed in Table 2, we can model T_{Lj} as linear function of composition using the relationship [12-21]:

$$T_{Lj} = \sum_{i=1}^N T_{Lij} g_i \quad (3)$$

where T_{Lij} is the i -th component coefficient for the j -th crystalline phase, N is the number of components in the model, and g_i is the mass fraction of the i -th component in glass.

Similarly, we can write for B_{Lj} and $c_{\max,j}$:

$$B_{Lj} = \sum_{i=1}^N B_{Lij} g_i \quad (4)$$

and

$$c_{\max,j} = \sum_{i=1}^N c_{\max,ij} g_i \quad (5)$$

Note that if the component coefficient value, such as T_{Lij} , B_{Lij} , or $c_{\max,ij}$, is lower than the minimum value of the associated property of glasses in the experimental dataset, such as T_{Lj} , B_{Lj} , or $c_{\max,j}$, an increase of the content of the i -th component decreases the value of this property in all glasses within the experimental composition region. If the component coefficient value is higher than the maximum value of the associated property, an increase of the content of the i -th component tends to increase the property value over the experimental composition region. Finally, if the component coefficient value lies between the minimum and maximum for the property values, the i -th component has little effect on the property [18].

3. Results and Discussion

In this study, nearly 140 crystallinity data points were generated within the temperature range of 950°C to 1200°C. The crystalline phases observed were Ce-borosilicate ($\text{Ce}_3\text{BSi}_2\text{O}_{10}$), mullite ($\text{Al}_0\text{Si}_2\text{O}_{19}$), corundum (Al_2O_3), and cerianite (CeO_2). Stillwellite-Ce and Nd-silicate ($\text{Nd}_2\text{Si}_2\text{O}_7$) were only detected in a few samples. Fig. 1 depicts the micrograph of the L-53 glass heat-treated at 999°C for 23.8 h; the crystalline phases formed a dense network within the bulk glass. Crystal fractions for Ce-borosilicate, Al-containing crystalline phases (the sum of mullite plus corundum and corundum separately), and cerianite are plotted versus the heat-treatment temperature in Fig. 2 for the L-53 glass. Fig. 3 shows crystal fractions versus temperature for all other glasses. The Ce-borosilicate phase was present in 116 samples. Fractions of Al-containing crystalline phases consist of 102 data points for $\text{Al}_0\text{Si}_2\text{O}_{19}$ and 52 data points for Al_2O_3 . The sum of $\text{Al}_0\text{Si}_2\text{O}_{19}$ and Al_2O_3 fractions presents a significant trend in all data series. Corundum barely formed in high-viscosity glasses, such as HS5, HS3, and LA5.

Table 3 summarizes the values of c_{max} , B_L , and T_L for Ce-borosilicate and Al-containing crystalline phases for the glasses listed in Table 1. These values were obtained by fitting Equations (2) to (5) to data. The resulting component coefficients are listed in Table 4. The correlations coefficients were $R^2 = 0.941$ for the Ce-borosilicate phase and $R^2 = 0.922$ for the Al-containing phases. The root mean square errors were $s = 0.850$ and 0.437 , respectively. The lines in Figs. 2 and 3 represent the model, i.e., Equations (2) to (5) with coefficients listed in Table 4.

As Figs. 2 and 3 indicate, several outlying data for the Ce-borosilicate crystals were not used for model fitting. Three outlying data points (glasses HS5, HB5, and LN5) were crystal fractions obtained at temperatures close to T_L , where the sparse crystallinity could be missed by the tiny XRD sample. One outlier (glass HS3) occurred at a low temperature at which the heat-treatment time probably did not allow the sample to reach equilibrium. Finally, the more Ce-borosilicate crystals were produced in HB3 than the model would predict. This composition is located between L-53 and HB5 compositions. The discrepancy between HS3 and the neighboring compositions, L-53 and HB5, was probably caused by an error that we were unable to trace. However, misbatching is unlikely considering good agreement between models and data for Al-containing phases in HS3.

As the coefficients listed in Table 4 indicate, adding SiO_2 and B_2O_3 to glass has a tendency to decrease T_L for the Ce-borosilicate crystals, whereas an increased Al_2O_3 content has a moderate tendency to increase T_L , and CeO_2 and Nd_2O_3 have a strong tendency to increase T_L . As expected, T_L of Al-containing crystalline phases strongly increased as the Al_2O_3 content increased, to a lesser extent as CeO_2 and Nd_2O_3 were added to the glass, and decreased as SiO_2 and B_2O_3 fractions increased.

As Fig. 3 plots indicate, Al-containing crystals were typically present above the “liquidus” temperature of the Ce-borosilicate crystalline phase. However, as the temperature decreased below T_L , the Ce-borosilicate crystalline phase became dominant even for high-alumina compositions.

In the LB5 glass, a crystalline phase identified as $\text{Nd}_2\text{Si}_2\text{O}_7$ occurred at $T > 1100^\circ\text{C}$. The fraction of this phase increased as temperature increased while the total Al crystal fraction sharply decreased.

Although the nonlinear model, Equation (2), was applicable to major crystalline phases, the CeO_2 phase was an exception. Its content was virtually independent of temperature, indicating that CeO_2 crystals might form during cooling. Table 5 lists the component coefficients of the function

$c_{\text{CeO}_2} = \sum_{i=1}^N c_{i\text{CeO}_2} g_i$. The coefficients are based on 136 data points. After omitting five outliers, the amount of observed CeO_2 ranged from 0.50 to 3.66 mass%; $R^2 = 0.77$ and $s = 0.33$. As the coefficient values indicate, CeO_2 crystal fraction decreased with increasing SiO_2 and B_2O_3 content in glass, and, as expected, increased with increasing CeO_2 content, whereas Al_2O_3 and Nd_2O_3 content had only moderate effects.

Fig. 4 compares the estimated Ce-borosilicate and Al-containing crystals fractions based on Equation 2 with the measured ones. The model used to estimate CeO_2 crystal fraction, shown in the previous paragraph, is also included in Fig. 4. The equality lines represent and the correlation coefficient are displayed for each crystalline phase (outliers were not considered).

Our results compare favorably with literature data for LABS glasses. With the Ce-borosilicate primary phase, the T_L values ranged from 1118 to 1248°C and for Al-containing crystalline phases, the T_L values ranged from 1138 to 1303°C. In most LABS-Sr glasses [11,12], the rare earth borosilicate ($\text{RE}_3\text{BSi}_2\text{O}_{10}$) crystalline phase was a clearly-recognizable primary crystalline phase, but Al_2O_3 and $\text{Al}_6\text{Si}_2\text{O}_{13}$ crystals appeared as the primary phases in a few glasses. The T_L values were between 1153 and 1405°C with either $\text{RE}_3\text{BSi}_2\text{O}_{10}$, Al_2O_3 , or $\text{Al}_6\text{Si}_2\text{O}_{13}$ as the primary phases [11,12]. Gasnier et al. [25] observed that the Ce-borosilicate phase was the primary crystalline phase even in Al-rich compositions. However in this study, the Nd^{3+} ion probably substituted for Ce^{3+} in the crystal structure of Ce-borosilicate phase because of similar ionic radii (98.3 and 101 pm, respectively) [26].

In HLW glasses, the primary crystalline phases are spinel and zirconium-containing minerals [12-21]. Interestingly, the relationship between c and T experimentally observed in HLW glasses, mostly for spinel crystals, was linear [13]. In this case, c can be approximated by a linear function, $c = b(T_L - T)$, where b is a constant coefficient. Values of the T_L estimated by linear extrapolation of the c versus T relationship over a temperature interval not far below the T_L are typically larger than carefully measured values, e.g., using multiple measurements in which the heat treatment is repeated until the

temperature comes as close to the T_L as possible [13] (the less laborious gradient temperature furnace method is subjected to a large uncertainty caused by the melt flow driven by surface tension gradients [27]). Thus, linear approximation of c versus T data is conservative for the case where crystals are not allowed to form. For any melters that tolerate crystals, the model can correctly represent c as a function of T and glass composition within the T interval appropriate for the melter performance [23] – an advanced melter technology can tolerate crystallinity at high temperature. Findings by Bräuchle and Huppertz [28] show that $\text{Pr}_3\text{BSi}_2\text{O}_{10}$ crystals, with a calculated density of 5.31 g cm^{-3} , is an analog to the Ce-borosilicate crystal ($\text{Ce}_3\text{BSi}_2\text{O}_{10}$). If the Ce-borosilicate crystal has a similar density to $\text{Pr}_3\text{BSi}_2\text{O}_{10}$, the crystal can potentially settle to form a sludge on the bottom of the melter. It has been shown that spinel with density of 5.2 g cm^{-3} can settle in the bottom of a conventional Joule-heated melter and this will decrease the melter lifetime [14]. This consequence is lessened if the crystallite sizes are small and because the settling rates are reduced and more of the crystals are discharged with the melt. Even though a nonlinear model, such as that of Equation (1), fits the data points better, the linear model could be preferable because it is easier for mathematical optimization. However, as Section 3 demonstrates, measured c versus T data for crystals in LABS compositions can be best fitted with the nonlinear model, Equation (1).

4. Conclusion

The LABS glass has good chemical durability and is a feasible option to immobilize RE oxide fission products from reprocessing of pyroprocessed used nuclear fuel. Also, this glass has a high waste loading capacity of $>50 \text{ mass\%}$. This work was conducted to find the effect of glass components on T_L by varying the chosen baseline glass composition one component at a time. The LABS glasses precipitated $\text{Ce}_3\text{BSi}_2\text{O}_{10}$ as the major phase, conjointly with $\text{Al}_{10}\text{Si}_2\text{O}_{19}$ and CeO_2 . Some of the glasses precipitated Al_2O_3 , which affected the crystal fraction of $\text{Al}_{10}\text{Si}_2\text{O}_{19}$ in heat-treated samples. A few glasses contained CeBSiO_5 while one sample contained $\text{Nd}_2\text{Si}_2\text{O}_7$. This study may help with continued experimental efforts that would help Korean agencies to see value in using this waste form as it has high waste loading and can be of exceptional economic value.

A nonlinear model was fit to the dataset to obtain fractions of Ce-borosilicate and Al-containing crystalline phases as functions of temperature and glass composition in mass fractions of oxides. Generally, the T_L increased as SiO_2 and B_2O_3 fractions decreased and as the CeO_2 , Nd_2O_3 , and Al_2O_3 fractions increased. These components had a similar effect on the crystallization of CeO_2 , which, however, was not affected by temperature and is suspected to precipitate during glass quenching. The model treats the coexisting major crystalline phases individually (except Al-containing phases, for

which fractions were summed), ignoring their interactions through their effects on matrix glass composition.

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Figures and Tables

Fig. 1. SEM micrograph of L-53 heat-treated at 999°C for 23.8 h.

Fig. 2. Crystal mass fractions as a function of temperature for the baseline glass, L-53 (in duplicate).

Fig. 3. Crystal mass fractions as a function of temperature for the various series of glasses. The legend for all of the plots is shown at the top.

Fig. 4. Measured and predicted c_j where $j \equiv \text{Ce}_3\text{BSi}_2\text{O}_{10}$ and total Al crystals (fitted by Equation 2), or $j = \text{CeO}_2$ (fitted by $c_{\text{CeO}_2} = \sum_{i=1}^N c_{i\text{CeO}_2} g_i$). Correlation coefficients for the equality line are included on the plots, together with outliers.

Table 1. LABS glass compositions in mass fractions, melting temperature (T_m), and waste loading (in mass%).

Glass		SiO ₂	B ₂ O ₃	Al ₂ O ₃	CeO ₂	Nd ₂ O ₃	T_m (°C)	Waste loading
Baseline	L-53	0.1973	0.0797	0.1930	0.2024	0.3276	1325	53.0
SiO ₂ series	HS5	0.2473	0.0747	0.1810	0.1898	0.3072	1325	49.7
	HS3	0.2273	0.0767	0.1858	0.1949	0.3153	1325	51.0
	LS3	0.1673	0.0826	0.2002	0.2100	0.3398	1325	55.0
	LS5	0.1473	0.0846	0.2051	0.2150	0.3480	1350	56.3
B ₂ O ₃ series	HB5	0.1866	0.1297	0.1825	0.1914	0.3098	1350	50.1
	HB3	0.1909	0.1097	0.1867	0.1958	0.3169	1350	51.3
	LB3	0.2037	0.0497	0.1993	0.2090	0.3382	1365	54.7
	LB5	0.2080	0.0297	0.2035	0.2134	0.3453	1385	55.9
Al ₂ O ₃ series	HA5	0.1851	0.0747	0.2430	0.1899	0.3073	1475	49.7
	HA3	0.1900	0.0767	0.2230	0.1949	0.3154	1425	51.0
	LA3	0.2046	0.0826	0.1630	0.2100	0.3398	1365	55.0
	LA5	0.2095	0.0846	0.1430	0.2150	0.3479	1375	56.3
CeO ₂ series	HC5	0.1849	0.0747	0.1809	0.2524	0.3070	1375	55.9
	HC3	0.1899	0.0767	0.1858	0.2324	0.3153	1375	54.8
	LC3	0.2047	0.0827	0.2003	0.1724	0.3399	1375	51.2
	LC5	0.2097	0.0847	0.2051	0.1524	0.3481	1400	50.1
Nd ₂ O ₃ series	HN5	0.1826	0.0737	0.1787	0.1874	0.3776	1375	56.5
	HN3	0.1885	0.0761	0.1844	0.1934	0.3576	1375	55.1
	LN3	0.2061	0.0832	0.2016	0.2115	0.2976	1375	50.9
	LN5	0.2120	0.0856	0.2074	0.2175	0.2776	1385	49.5

Table 2. Glass composition and waste loading ranges (in mass fraction).

Components	Min. fraction	Max. fraction
SiO ₂	0.1473	0.2473
B ₂ O ₃	0.0297	0.1297
Al ₂ O ₃	0.1430	0.2430
CeO ₂	0.1524	0.2524
Nd ₂ O ₃	0.2776	0.3776
Waste loading	0.4951	0.5650

Table 3. Values of c_{max} , B_L , and T_L for Ce-borosilicate and Al-containing crystalline phases.

Glass	Ce-borosilicate			Al-containing crystals		
	c_{max}	B_L (K)	T_L (°C)	c_{max}	B_L (K)	T_L (°C)
L-53	0.372	15521	1183	0.217	10497	1221
HA3	0.346	17217	1183	0.255	10417	1264
HA5	0.329	18349	1184	0.280	10365	1292
HB3	0.418	8526	1144	0.201	7319	1171
HB5	0.449	3868	1118	0.191	5203	1138
HC3	0.343	16541	1193	0.210	7346	1238
HC5	0.323	17223	1200	0.206	5243	1250
HN3	0.360	18532	1200	0.197	11838	1222
HN5	0.352	20537	1199	0.183	12731	1223
HS3	0.423	11861	1210	0.223	13397	1182
HS5	0.457	9421	1167	0.226	15330	1155
LA3	0.398	13821	1157	0.179	10576	1178
LA5	0.415	12689	1182	0.154	10629	1149
LB3	0.326	22499	1182	0.233	13667	1270
LB5	0.296	27157	1222	0.244	15783	1303
LC3	0.402	14497	1248	0.224	13655	1203
LC5	0.422	13816	1173	0.229	15758	1192
LN3	0.384	12517	1166	0.238	9159	1219
LN5	0.392	10512	1167	0.251	8266	1218
LS3	0.321	19181	1156	0.212	7596	1260
LS5	0.287	21621	1199	0.208	5663	1286

Table 4. Component coefficients for Ce-borosilicate and Al-containing crystalline phases.

	Ce-borosilicate			Al-containing crystals		
	$c_{max,ij}$	$10^{-3}B_{L,ij}$ (K)	$T_{L,ij}$ (°C)	$c_{max,ij}$	$10^{-3}B_{L,ij}$ (K)	$T_{L,ij}$ (°C)
SiO ₂	1.737	-82.41	1040	0.364	88.10	172
B ₂ O ₃	1.779	-198.82	252	-0.272	-86.88	-302
Al ₂ O ₃	-0.317	61.19	1470	1.237	8.36	2376
CeO ₂	-0.415	42.69	1726	0.035	-73.36	1684
Nd ₂ O ₃	0.100	82.93	1824	-0.240	40.52	1256

Table 5. Component coefficients for CeO₂ crystalline phase.

Component	c_i
SiO ₂	-0.084
B ₂ O ₃	-0.193
Al ₂ O ₃	0.013
CeO ₂	0.183
Nd ₂ O ₃	0.030