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Title: Final Letter Report on PCT/monolithic Long-Term Glass Ceramic Corrosion Tests
M3NT-18SR030105147

Introduction

The Savannah River National Laboratory (SRNL) is collaborating with the Pacific Northwest National Laboratory (PNNL) to study advanced waste form glass ceramics for immobilization of waste from Used Nuclear Fuel (UNF) separations processes. This work is part of the United States Department of Energy Nuclear Engineering (USDOE-NE) program on Material Recovery & Waste Form Development (MRWFD) (MRWFD 2015, MRWFD 2016). The glass ceramic waste forms take advantage of both crystalline and glassy phases where 'troublesome' elements (e.g., low solubility in glass or very long-lived) partition to highly durable ceramic phases with the remainder of elements residing in the glassy phase. The ceramic phases are tailored to create certain minerals or unique crystalline structures that can host the radionuclides by binding them in their specific crystalline network while not adversely impacting the residual glass network (Crum et al., 2011). Glass ceramics have been demonstrated using a scaled melter test performed in a pilot scale (1/4 scale) cold crucible induction melter (CCIM) (Crum et al., 2014; Maio et al., 2015). This report summarizes final long-term corrosion leach testing results from both the Phase I and Phase II bench scale tests involving crucible fabrication and chemical durability testing of glass ceramics using the Product Consistency Test (PCT). Preliminary results from both Phase I and Phase II bench scale tests involving statistically designed matrices have previously been reported (Crawford, 2013; Crawford, 2014; Crawford, 2015, Crawford, 2016).

Experimental

Details pertaining to the glass ceramic fabrication, chemical durability testing via the static powdered leach testing PCT (ASTM C-1285) and analytical instrumentation have been presented in earlier reports (Crawford, 2013; Crawford, 2104). ASTM Type I water was collected from an ARIES High Purity Water System with an indicated resistivity of greater than 18 MΩ·cm resistivity and was used as leachant in these studies. Leachates were analyzed using Inductively Coupled Plasma Atomic Emission Spectroscopy (ICP-AES) for all analytes except for Cs, which was analyzed by Inductively Coupled Plasma Mass Spectroscopy (ICP-MS). Three glass ceramic samples from Phase II were analyzed by Scanning Electron Microscopy/Energy Dispersive X-ray Spectroscopy (SEM/EDS). The leached particles were prepared per PCT guidance with ASTM Type I water rinse and drying at < 90 ºC. The SEM is a Hitachi SU8200 model with EDS mapping capabilities - the EDS detector for the SEM is manufactured by Oxford Instruments.

Phase II and Phase I Glass Ceramic PCT Data

Phase II

This memorandum presents PCT leachate data for the Phase II Glass Ceramics collected at 882 days. Previous data was reported through 64 weeks or 448 days (Crawford, 2016). Table 1 shows dilution-corrected leachate concentrations. All leachates for this and previous glass ceramic PCTs used a dilution factor of 6 mL filtered leachate added to 4 mL of nominal 0.4M ultrapure nitric acid to result in a 10 mL diluted leachate in an ~1 volume percent (vol %) acid for analysis. Table 2 shows the normalized concentrations (NC_i) in units of gram wasteform leached / liter. These data were generated using the measured chemical composition of the Phase II glass ceramic (Crawford, 2014; Crawford, 2015) and the dilution-corrected leachate values. Appendix A shows the as-analyzed 882-day leachate concentrations and the ambient temperature measured pH values for the leachates. The ‘I’, ‘O’ and ‘C’ designations in the sample identifications represent Inner, Outer and Centroid compositions in the statistically designed Phase II glass ceramic matrix. The targeted Phase II and Phase I glass ceramic chemical compositions with the statistical designation identifications are shown in Appendix B.

The NC_i equation is given below.

$$NC_i = C_i(\text{sample}) / f_i$$

where:

NC_i = normalized concentration, g waste form/L leachant,
C_i(sample) = concentration of element “*i*” in the solution, g_i/L,
f_i = mass fraction of element “*i*” in the unleached waste form (unitless)

The NC_i data shown in Table 2 for the compositions of I29 and O22 are highlighted since these two compositions produced higher NC_i values than the other compositions (see figures and discussion below). All of the NC_i values presented for the Phase II glass ceramics can be compared to the Environmental Assessment (EA) glass (Jantzen et al., 1993) mentioned in the Department of Energy (DOE) Waste Acceptance Product Specifications (WAPS, 2012 for Vitrified High-Level Waste (HLW)) as an upper limit benchmark glass. The 7-day published reference values for normalized concentrations of EA boron and sodium release are 16.695 +/- 1.222 g/L and 13.346 +/- 0.902 g/L, respectively. Thus these data show that even at the longest duration of 882 days, the NC_i values for these Phase II glass ceramics are lower than the shorter 7-day data for the reference EA glass (Jantzen et al., 1993). The 7-day Phase II glass ceramic data has been previously reported (Crawford, 2015).

Table 1. Phase II Dilution Corrected Leachate Concentrations from 882-Day PCT

Sample ID	Al	B	Ba	Ca	Li	Mo	Na	Si	Sr	Te	Cs
	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)
I29	2.70	208.33	2.87	3.45	49.33	156.83	275.54	129.20	1.75	<3.33	84.42
I30	1.85	11.15	<1.67	2.93	2.30	4.72	4.22	18.60	<1.67	<3.33	9.14
I31	<1.67	93.02	5.00	11.83	29.15	5.88	26.40	69.33	6.55	<3.33	52.79
I32	<1.67	22.17	4.58	5.00	4.58	33.65	52.29	27.22	3.27	<3.33	34.75
I33	<1.67	35.52	<1.67	2.50	8.93	6.45	9.70	32.32	<1.67	<3.33	24.40
I34	<1.67	33.33	2.20	<1.67	12.88	86.83	60.50	38.33	<1.67	<3.33	41.57
I35	<1.67	62.97	3.43	7.95	29.35	59.77	90.24	92.63	4.57	13.12	77.03
I36	<1.67	23.53	3.60	7.52	5.42	38.23	50.86	38.35	3.35	3.95	43.23
I40	<1.67	16.15	3.98	2.35	3.73	40.12	40.32	28.93	2.53	<3.33	26.42
O21	3.18	112.65	<1.67	25.43	<1.67	119.17	314.99	37.62	5.32	10.45	42.55
O22	<1.67	263.37	11.38	15.43	<1.67	17.15	177.63	135.23	7.73	<3.33	248.77
O23	<1.67	22.90	<1.67	5.15	13.45	8.42	6.94	47.48	<1.67	<3.33	29.77
O24	<1.67	100.98	5.72	5.28	<1.67	38.10	169.52	142.57	8.72	<3.33	163.27
O25	<1.67	42.55	16.28	11.82	24.28	11.27	5.60	137.33	11.08	<3.33	105.14
O26	2.15	27.82	1.67	3.43	11.60	23.98	2.91	12.12	3.93	<3.33	36.06
O27	<1.67	29.35	<1.67	13.18	<1.67	3.22	9.11	60.47	2.35	<3.33	70.12
O28	2.18	7.15	<1.67	<1.67	<1.67	4.92	5.37	15.63	<1.67	<3.33	9.76
O39	<1.67	54.68	10.93	7.75	34.70	19.53	5.71	160.97	7.53	<3.33	160.94
C41	<1.67	14.10	4.07	3.73	4.83	21.58	13.33	19.97	2.15	<3.33	16.35

Table 2. Phase II Normalized Concentrations from 882-Day PCT*

Sample ID	NC_{Al}	NC_{B*}	NC_{Ba}	NC_{Ca}	NC_{Li}	NC_{Mo}	NC_{Na*}	NC_{Si}	NC_{Sr}	NC_{Te}	NC_{Cs}
	(g/L)	(g/L)	(g/L)	(g/L)	(g/L)	(g/L)	(g/L)	(g/L)	(g/L)	(g/L)	(g/L)
O21	0.08	6.66	<0.06	0.44	NA	8.09	7.26	0.28	0.41	1.45	1.94
O23	<1.07	1.30	<0.06	0.10	1.65	0.56	1.87	0.30	<0.13	<0.25	0.89
O25	<0.97	2.09	<0.37	0.18	2.81	0.24	1.03	0.80	0.55	<0.29	2.09
O27	<0.06	0.84	<0.04	0.21	NA	0.20	0.87	0.43	0.12	<0.28	1.22
I29	0.19	6.77	<0.09	0.11	8.05	3.13	6.62	0.87	0.12	<0.38	2.30
I31	<0.13	2.81	<0.17	0.39	4.76	0.20	2.29	0.44	0.48	<0.36	1.35
I33	<0.06	1.10	<0.06	0.08	2.13	0.13	0.91	0.24	<0.13	<0.43	0.67
I35	<0.12	3.08	<0.10	0.15	5.07	2.55	3.00	0.66	0.28	1.31	2.05
I36	<0.10	1.11	<0.10	0.14	2.07	1.62	1.21	0.25	0.20	0.41	1.22
O22	<0.87	8.75	<0.46	0.27	NA	0.29	8.87	1.07	0.65	<0.46	8.01
O24	<1.38	2.80	<0.13	0.19	NA	2.34	2.93	0.84	0.42	<0.28	2.75
I32	<0.05	0.72	<0.11	0.20	0.61	2.41	1.27	0.21	0.18	<0.35	1.06
O28	0.06	0.46	<0.04	<0.07	NA	0.09	0.55	0.11	<0.09	<0.47	0.21
I30	0.06	0.54	<0.05	0.06	0.84	0.09	0.36	0.11	<0.12	<0.45	0.23
O26	0.15	1.46	<0.05	0.06	4.57	1.04	0.53	0.08	0.23	<0.37	0.69
I34	<0.13	1.63	<0.07	<0.05	2.85	1.96	1.54	0.29	<0.12	<0.43	1.44
O39	<0.77	3.10	<0.24	0.12	4.22	0.27	0.88	1.03	0.36	<0.30	2.84
I40	<0.13	0.85	<0.14	0.07	1.52	1.50	1.10	0.18	0.18	<0.43	0.76
C41	<0.07	0.57	<0.13	0.10	1.13	0.52	0.56	0.13	0.15	<0.40	0.40

NA – No Li was added to these glass ceramics
*Normalized concentrations for EA boron and sodium 7-day release are 16.695 +/- 1.222 g/L and 13.346 +/- 0.902 g/L, respectively

Normalized concentrations for the glass components (NC_B and NC_{Na}) as well as key waste components (NC_{Mo} and NC_{Cs}) are plotted in Figure 1 through Figure 4 for the Phase II PCT leachates over the various time intervals. Data for these figures derives from Table 2 from this report and previously reported NC_i values for the 7- and 28-day data sets (Crawford, 2015) and 112- and 448-day data sets (Crawford, 2016). Both the Phase I and Phase II data sets contain a few compositions that show significantly higher NC_B and NC_{Na} than other compositions in the respective sets. Phase I data sets will be presented/discussed later in this report. Figure 1 shows that for the compositions O22 and I29, the NC_B values are 8.7 g/L and 6.7 g/L at 882 days compared to all the other compositions that approach 3 g/L and below. Similar trends are shown for NC_{Na} in Figure 2. All of the NC_{Mo} and NC_{Cs} values plotted in Figure 3 and Figure 4 are at or below the 3 g/L level, except for the O22 matrix that shows NC_{Cs} values that approach 8 g/L at 882 days.

The normalized concentrations for glass ceramics show similar trends to those from long term leach studies on glass, e.g., the SON68 glass that was studied in previous work involving long term PCT leach studies from SRNL (Ebert et al., 2011). The generalized trend is a Stage I release in the initial 7 days, followed by a transition to Stage II waste glass corrosion with a measurable residual rate. The pH measurements from the ambient temperature final leachates are plotted in Figure 5. All of the pH values are in the range of 8.8 to 10.3, except for the final 882 day O21 composition, which has a pH value of 10.9. The O28 composition leachate pH also appears to be consistently below the other composition leachates at all the time intervals. Measured pH values for blank leachant solutions that were tested with the glass ceramics ranged from pH 6.6 to 8.4.

One of the long term static tests on glass ceramics (matrix O21) shows possible indication of a Stage III or resumption of rate that is more similar to the Stage I rate at this time. The various stages of waste glass corrosion are described in detail in recent publications (MRWFD 2015; Gin et al., 2015). Figure 6 shows normalized concentration plots for this O21 composition. Figure 6 shows NC_i values for B, Na and Mo that suggest a higher corrosion rate in the 448-day to 882-day regime than the 28-day to 112-day regime. The NC_{Cs} appears to show similar behavior but to a lesser extent.

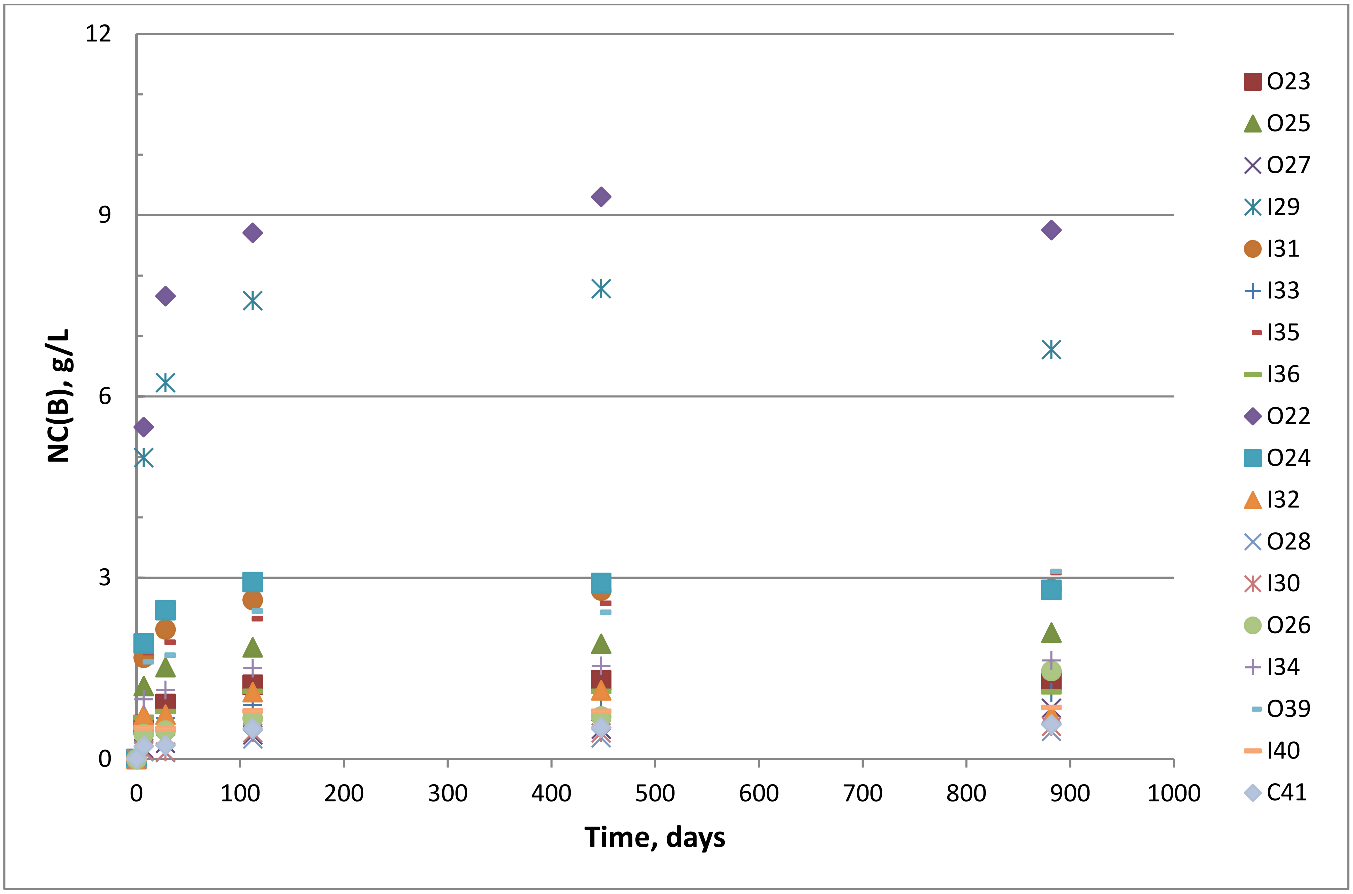


Figure 1. Phase II NC_B vs. Time

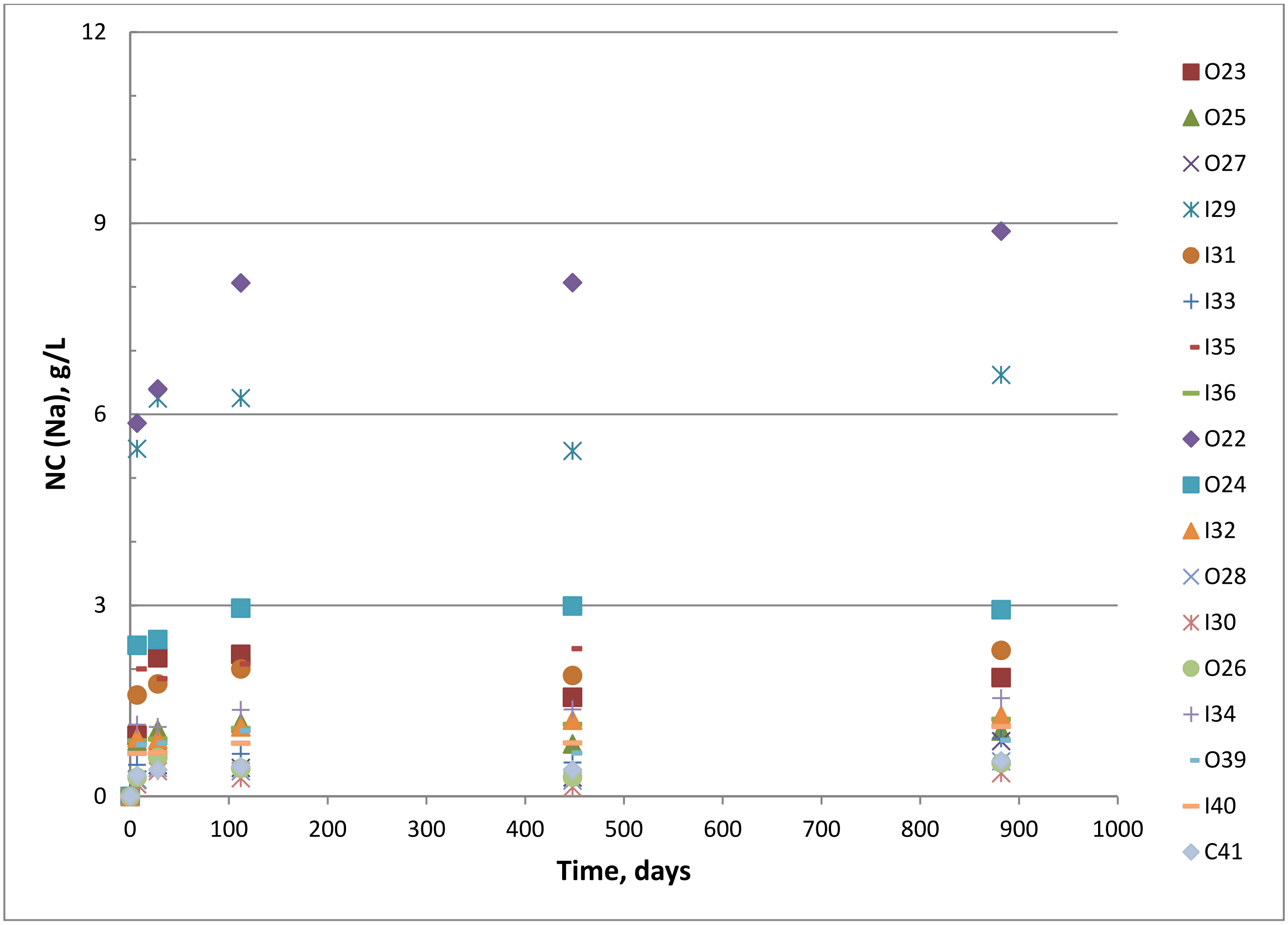


Figure 2. Phase II NC_{Na} vs. Time

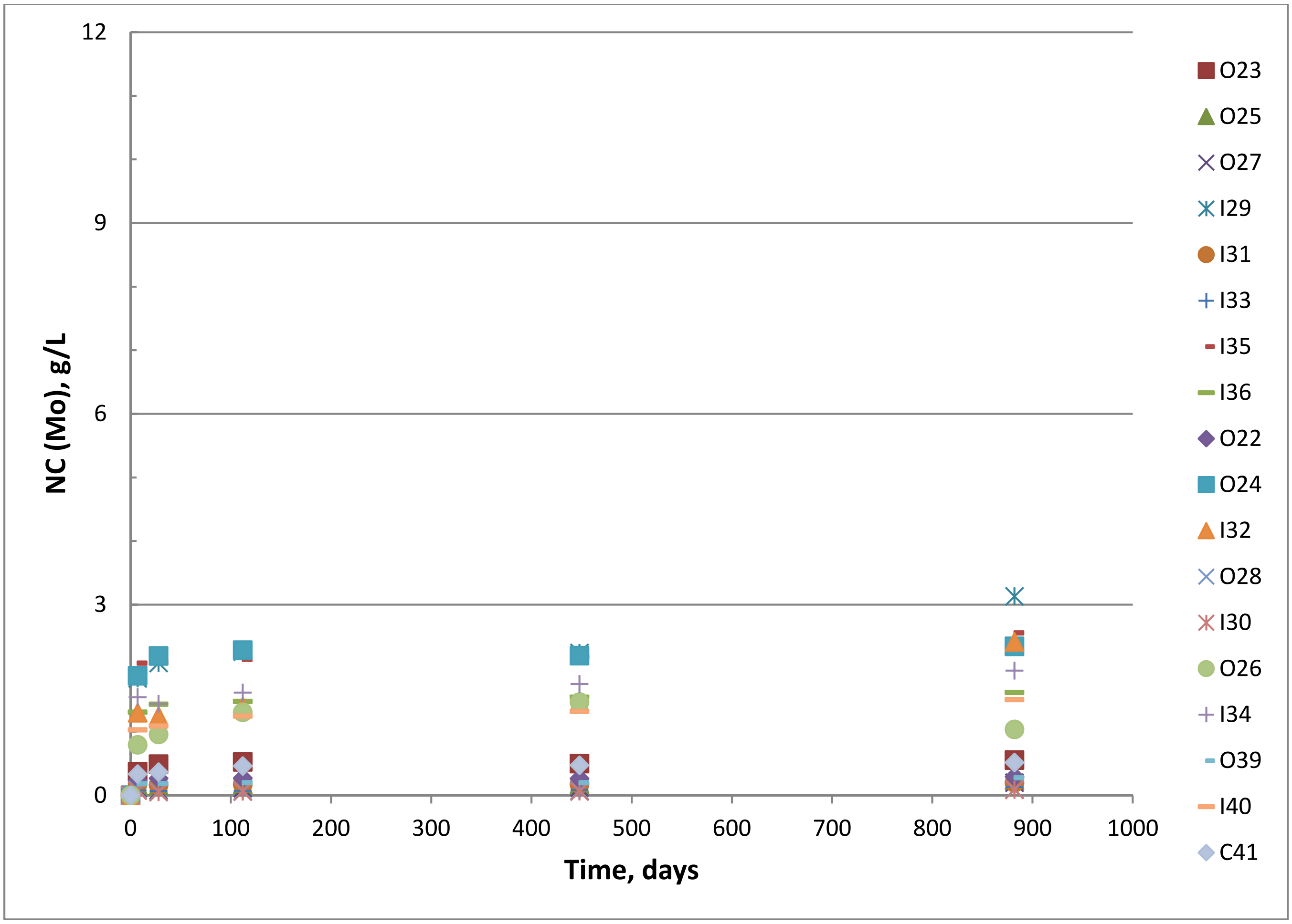


Figure 3. Phase II NC_{Mo} vs. Time

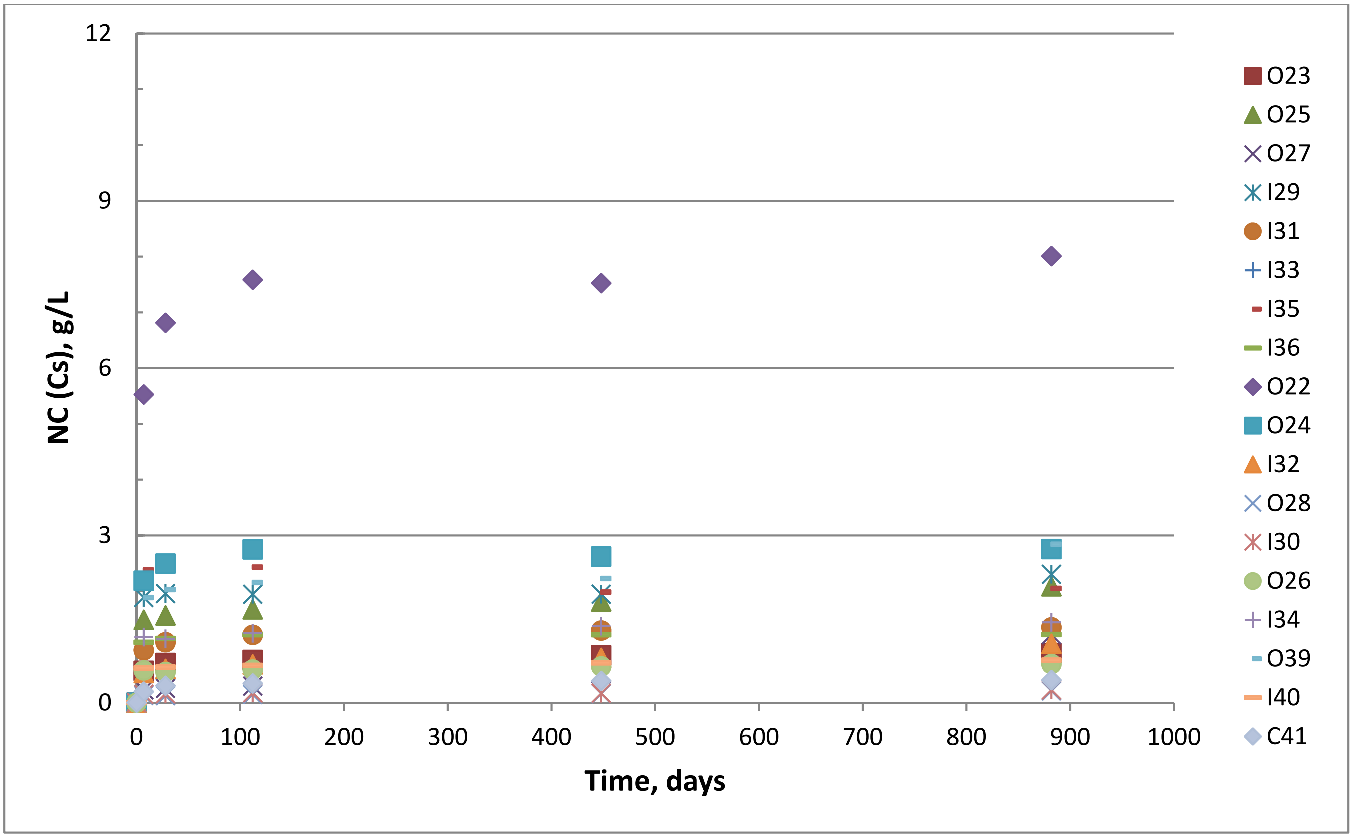


Figure 4. Phase II NC_{Cs} vs. Time

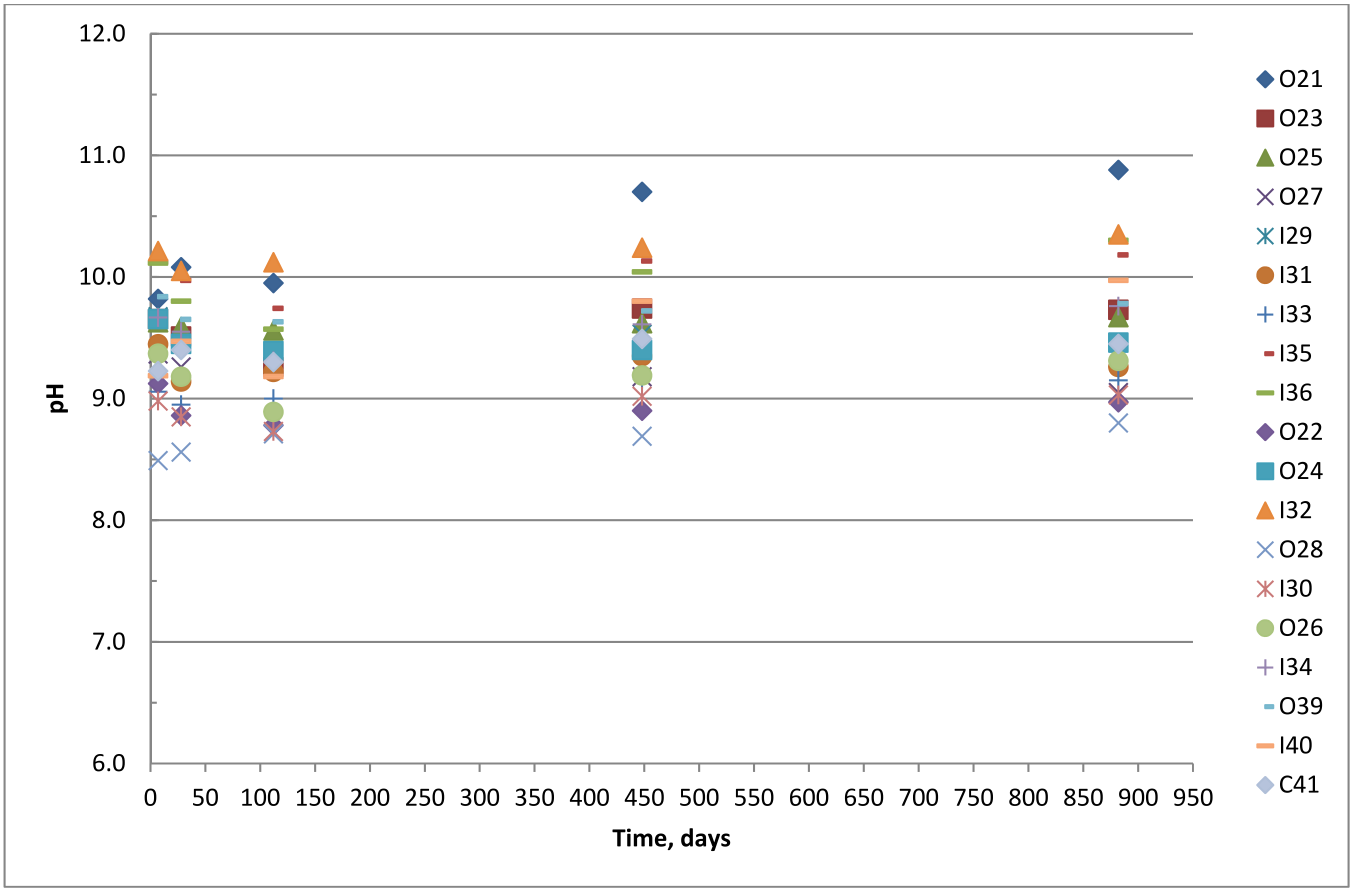


Figure 5. Phase II Leachate pH vs. Time

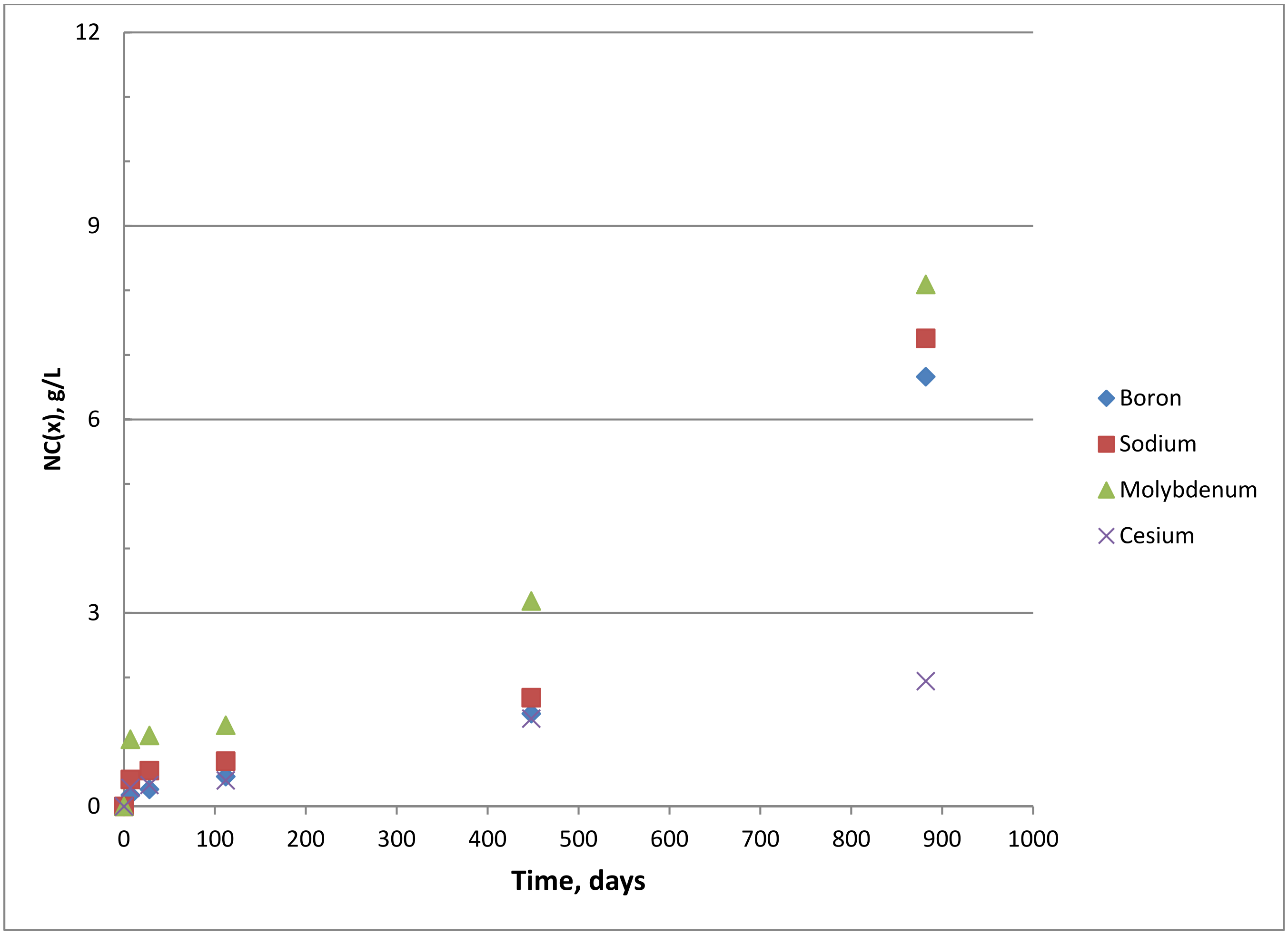


Figure 6. Phase II Normalized Concentration Sample O21 (B, Na, Mo, Cs)

Phase I

This memorandum presents PCT leachate data for the Phase I Glass Ceramics collected at 1217 and 1456 days. Previous data was reported through 64 weeks (Crawford, 2015). Table 7 shows dilution-corrected leachate concentrations for the 1217 day data and Table 8 shows the dilution-corrected leachate concentrations for the 1456-day data. The 1217 day data set was prepared by sampling a very small 0.25 mL aliquot of the leachate and diluting this volume into 10 mL of 1% nitric acid. The latter 1456-day set was prepared by using 6 mL filtered leachate added to 4 mL of nominal 0.4M ultrapure nitric acid to result in a 10 mL diluted leachate in an ~ 1 volume percent (vol%) acid for analysis.

Table 9 and Table 10 show the corresponding normalized concentrations (NC*i*) in units of gram waste form leached / liter. These data were generated using the measured chemical composition of the Phase I glass ceramic (Crawford, 2014; Crawford, 2015) and the dilution-corrected leachate values. Appendix A shows the as-analyzed 1217-day leachate concentrations and the 1456-day leachate concentrations the ambient temperature measured pH values for the leachates. No pH measurements were made for the 1217-day samples. The ‘I’, ‘O’ and ‘C’ designations in the sample identifications represent Inner, Outer and Centroid compositions in the statistically designed Phase I glass ceramic matrix shown in Appendix B.

Table 7. Dilution Corrected Leachate Concentration for 1217-Day PCT

Sample ID	Al	B	Ba	Ca	Li	Mo	Na	Si	Sr	Te	Cs
	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)
Blnk5	<41.00	40.14	<41.00	<41.00	<41.00	<41.00	<41.00	<41.00	<41.00	<41.00	<0.21
C1	<41.00	5.34	<41.00	<41.00	<41.00	15.46	<41.00	<41.00	<41.00	<41.00	9.69
C20	<41.00	4.58	<41.00	<41.00	<41.00	28.09	<41.00	<41.00	<41.00	<41.00	23.38
O3	<41.00	452.15	<41.00	7.63	123.66	565.70	720.34	154.79	<41.00	<41.00	182.42
O6	<41.00	23.20	<41.00	<41.00	<41.00	37.31	109.15	<41.00	<41.00	<41.00	78.84
O8	<41.00	61.56	<41.00	<41.00	<41.00	6.48	<41.00	<41.00	<41.00	<41.00	83.22
O10	<41.00	<1	<41.00	<41.00	<41.00	32.08	48.35	<41.00	<41.00	<41.00	20.51
O13	<41.00	42.40	<41.00	<41.00	<41.00	44.83	<41.00	<41.00	<41.00	<41.00	96.25
I14	<41.00	271.44	<41.00	<41.00	<41.00	4.84	177.54	<41.00	<41.00	<41.00	73.10
O15	<41.00	33.38	<41.00	<41.00	<41.00	<41.00	<41.00	<41.00	<41.00	<41.00	34.71
O16	<41.00	<1	<41.00	<41.00	<41.00	4.06	<41.00	<41.00	<41.00	<41.00	4.70

Table 8. Dilution Corrected Leachate Concentration for 1456-Day PCT

Sample ID	Al	B	Ba	Ca	Li	Mo	Na	Si	Sr	Te	Cs
	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)
Blnk5	<1.7	<1.7	<1.7	<1.7	<1.7	<1.7	2.22	3.33	<1.7	<3.3	<0.02
C1	<1.7	15.54	<1.7	3.76	8.47	21.00	15.53	34.33	<1.7	<3.3	10.42
C20	<1.7	18.62	2.33	4.21	7.20	36.00	20.30	29.70	2.13	<3.3	24.79
O3	11.53	434	2.95	2.87	141.93	461.50	725.75	262.50	1.85	3.83	161.93
O6	<1.7	37.28	4.36	6.38	<1.7	44.57	106.94	73.73	3.22	<3.3	79.29
O8	<1.7	80.32	1.87	27.27	<1.7	11.45	23.59	45.50	2.33	<3.3	89.11
O10	<1.7	12.21	2.90	3.60	<1.7	39.20	39.79	29.95	3.32	<3.3	20.61
O13	<1.7	62.00	4.07	6.19	27.80	51.92	3.07	91.68	4.62	<3.3	98.28
I14	<1.7	277.33	<1.7	6.25	16.37	10.31	179.48	70.72	<1.7	<3.3	72.34
O15	<1.7	54.45	<1.7	8.66	13.56	3.54	2.98	58.25	<1.7	<3.3	38.22
O16	<1.7	16.80	<1.7	6.35	11.92	9.57	9.43	30.33	<1.7	<3.3	4.61

Table 9. Phase I Normalized Concentrations from 1217-Day PCT*

Sample ID	NC_{B*}	NC_{Ca}	NC_{Li}	NC_{Mo}	NC_{Na*}	NC_{Si}	NC_{Cs}
	(g/L)	(g/L)	(g/L)	(g/L)	(g/L)	(g/L)	(g/L)
C1	0.22	<1.00	<9.61	0.38	<1.61	<0.26	0.37
C20	0.17	<1.06	<9.83	0.72	<1.64	<0.30	0.57
O3	13.30	0.25	14.70	11.37	12.89	1.12	5.47
O6	1.23	<0.65	NA**	1.57	2.58	<0.28	2.82
O8	1.89	<1.70	NA**	0.14	<3.78	<0.32	1.68
O10	<0.1	<1.42	NA**	0.84	0.99	<0.25	0.58
O13	1.50	<1.72	<5.57	0.91	<6.05	<0.28	1.95
I14	5.47	<1.38	<27.61	0.13	5.95	<0.28	1.94
O15	0.66	<0.90	<5.17	<1.13	<7.67	<0.31	0.93
O16	<0.1	<0.60	<4.84	0.13	<2.75	<0.28	0.14

* Normalized concentrations for EA boron and sodium 7-day release are 16.695 +/- 1.222 g/L and 13.346 +/- 0.902 g/L, respectively.

** No Li added to this glass ceramic composition

Table 10. Phase I Normalized Concentrations from 1456-Day PCT*

Sample ID	NC_{Al}	NC_{B*}	NC_{Ba}	NC_{Ca}	NC_{Li}	NC_{Mo}	NC_{Na*}	NC_{Si}	NC_{Sr}	NC_{Te}	NC_{Cs}
	(g/L)	(g/L)	(g/L)	(g/L)	(g/L)	(g/L)	(g/L)	(g/L)	(g/L)	(g/L)	(g/L)
C1	<0.07	0.63	<0.05	0.09	1.99	0.52	0.61	0.22	<0.11	<0.16	0.39
C20	<0.07	0.68	0.07	0.11	1.73	0.93	0.81	0.22	0.14	<0.15	0.61
O3	5.68	12.76	0.09	0.10	16.88	9.28	12.99	1.90	0.13	0.17	4.85
O6	<0.91	1.98	0.12	0.10	NA**	1.88	2.53	0.51	0.18	<0.13	2.84
O8	<0.05	2.47	0.05	1.13	NA**	0.25	2.18	0.36	0.14	<0.13	1.80
O10	<0.04	0.64	0.10	0.12	NA**	1.03	0.82	0.18	0.26	<0.14	0.58
O13	<0.80	2.19	0.11	0.26	3.78	1.06	0.45	0.63	0.28	<0.11	2.00
I14	<0.09	5.59	0.06	0.21	11.02	0.27	6.01	0.47	<0.13	<0.15	1.92
O15	<0.04	1.07	0.06	0.19	1.71	0.10	0.56	0.44	<0.13	<0.15	1.02
O16	<0.04	0.84	0.06	0.09	1.41	0.31	0.63	0.20	<0.12	<0.15	0.13

* Normalized concentrations for EA boron and sodium 7-day release are 16.695 +/- 1.222 g/L and 13.346 +/- 0.902 g/L, respectively.

** No Li added to this glass ceramic composition

Normalized concentrations for the glass components (NC_B and NC_{Na}) as well as key waste components (NC_{Mo} and NC_{Cs}) are plotted in Figure 7 through Figure 10 for the Phase I PCT leachates over the various time intervals. Data for these figures derives from Tables 9 and 10 from this report and previously reported NC_i values for the 7-, 28-, 119-, and 448-day data sets (Crawford, 2015). Similar plots using the same vertical scale of 0 to 15 g/L were shown for the previous one year testing from the Phase I glass ceramics (Crawford, 2015). Both the Phase I and Phase II data sets contain a few compositions that show significantly higher NC_B and NC_{Na} than other compositions in the respective sets. Figure 7 shows that for the compositions O3 and I14, the NC_B values are 12 g/L and 6 g/L beyond 1200 days compared to all the other compositions that are less than 3 g/L. Similar trends are shown for NC_{Na} in Figure 8. The NC_{Mo} values plotted in Figure 9 are at or below the 3 g/L level, except for the O3 matrix that shows NC_{Mo} values in the range of 9 to 12 g/L at the longer leach intervals. The NC_{Cs} values plotted in Figure 10 are at or below 3 g/L, except for the O3 matrix that shows NC_i values in the range of 5 to 6 g/L at the longer leach intervals.

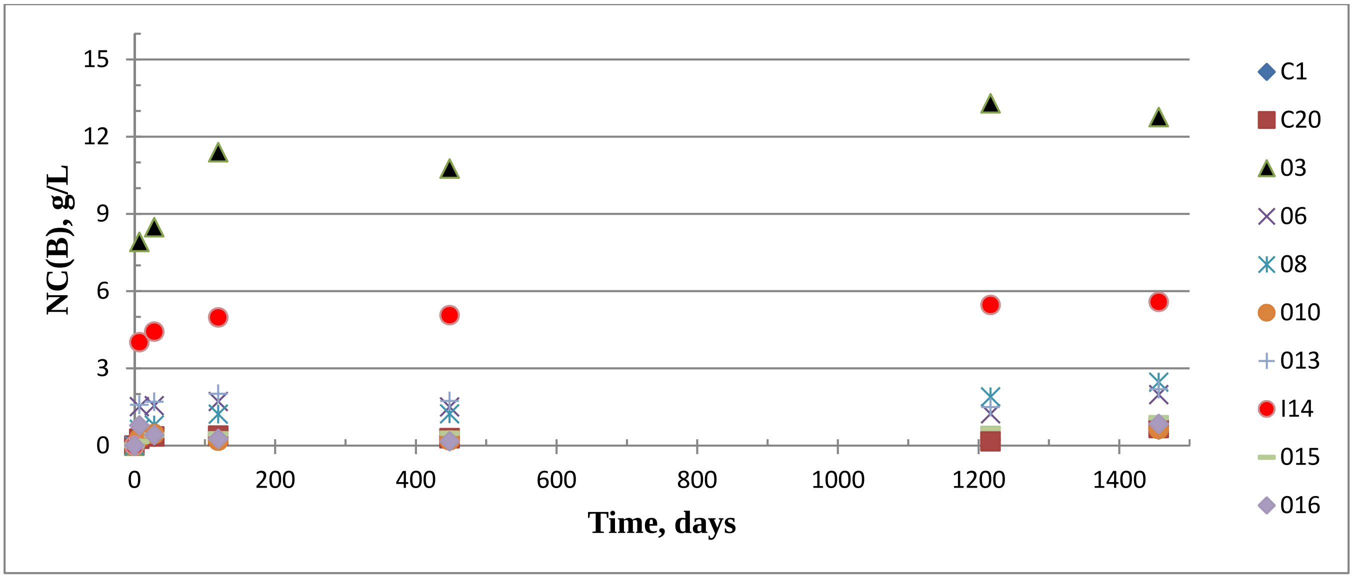


Figure 7. Phase II NC_B vs. Time

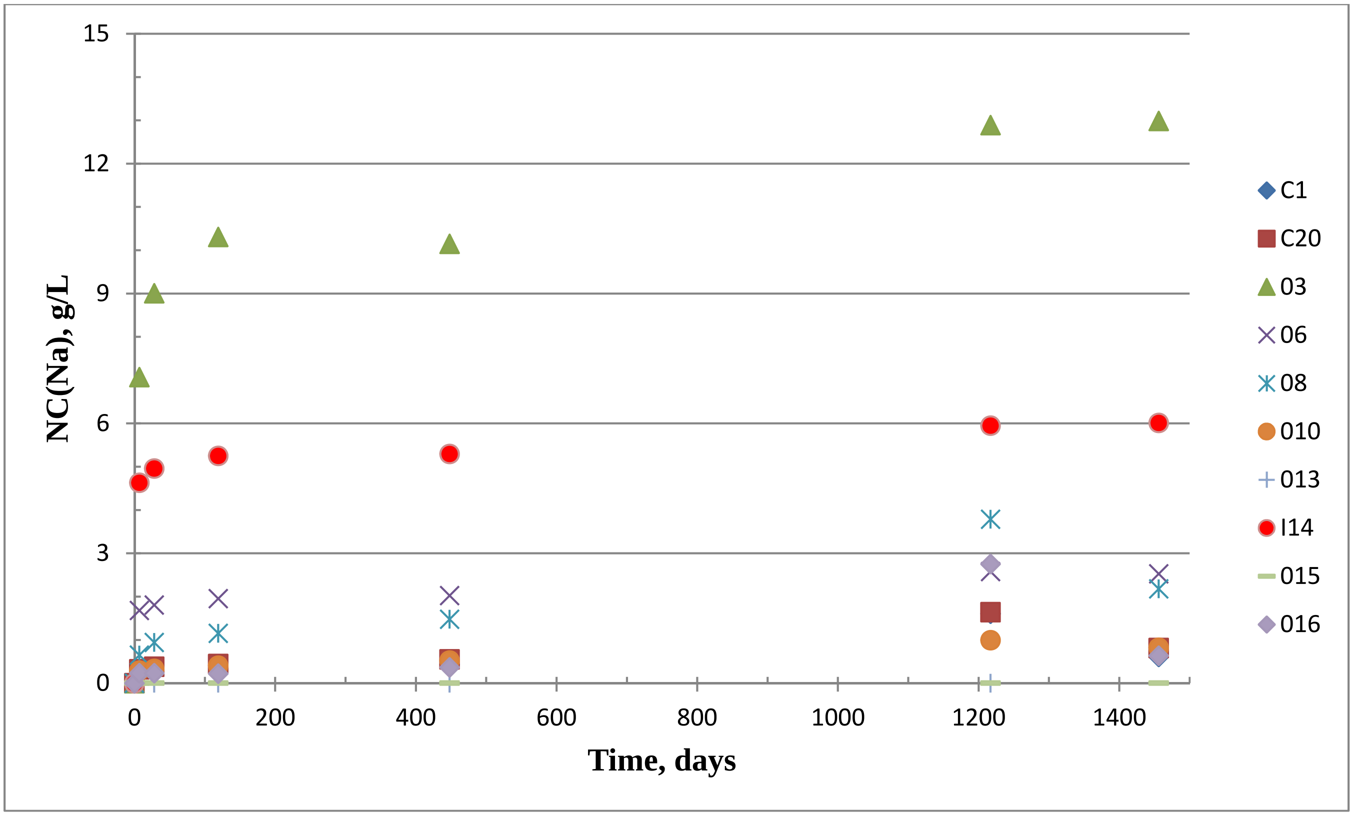


Figure 8. Phase II NC_{Na} vs. Time

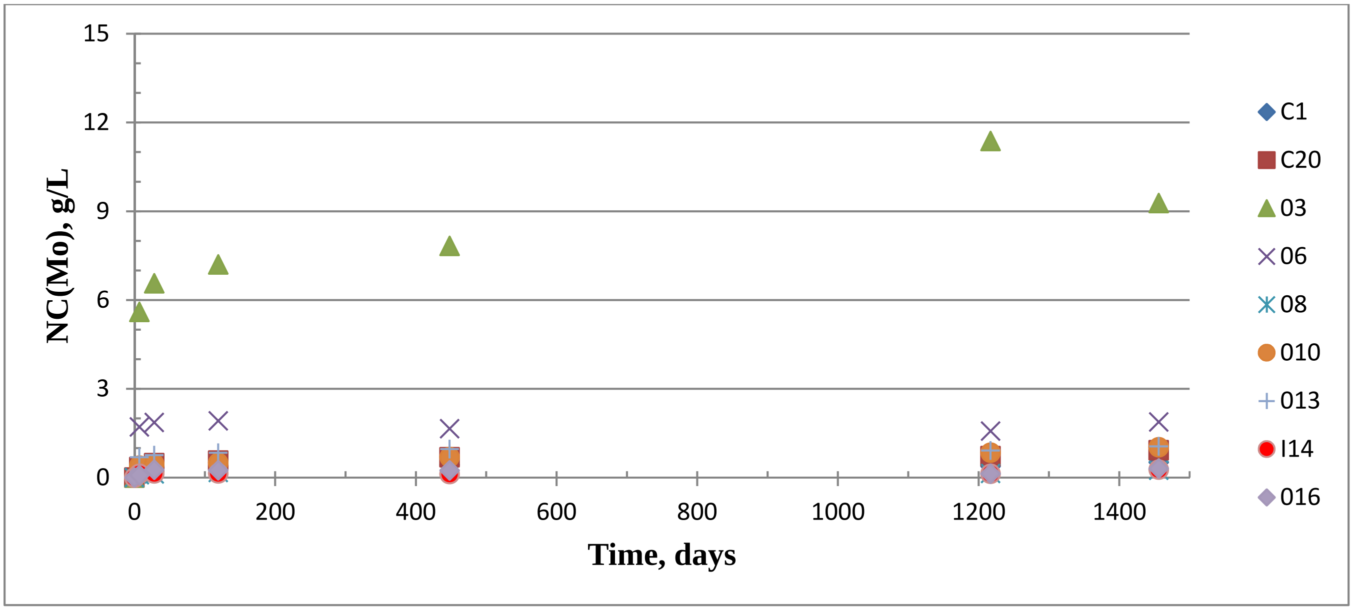


Figure 9. Phase II NC_{Mo} vs. Time

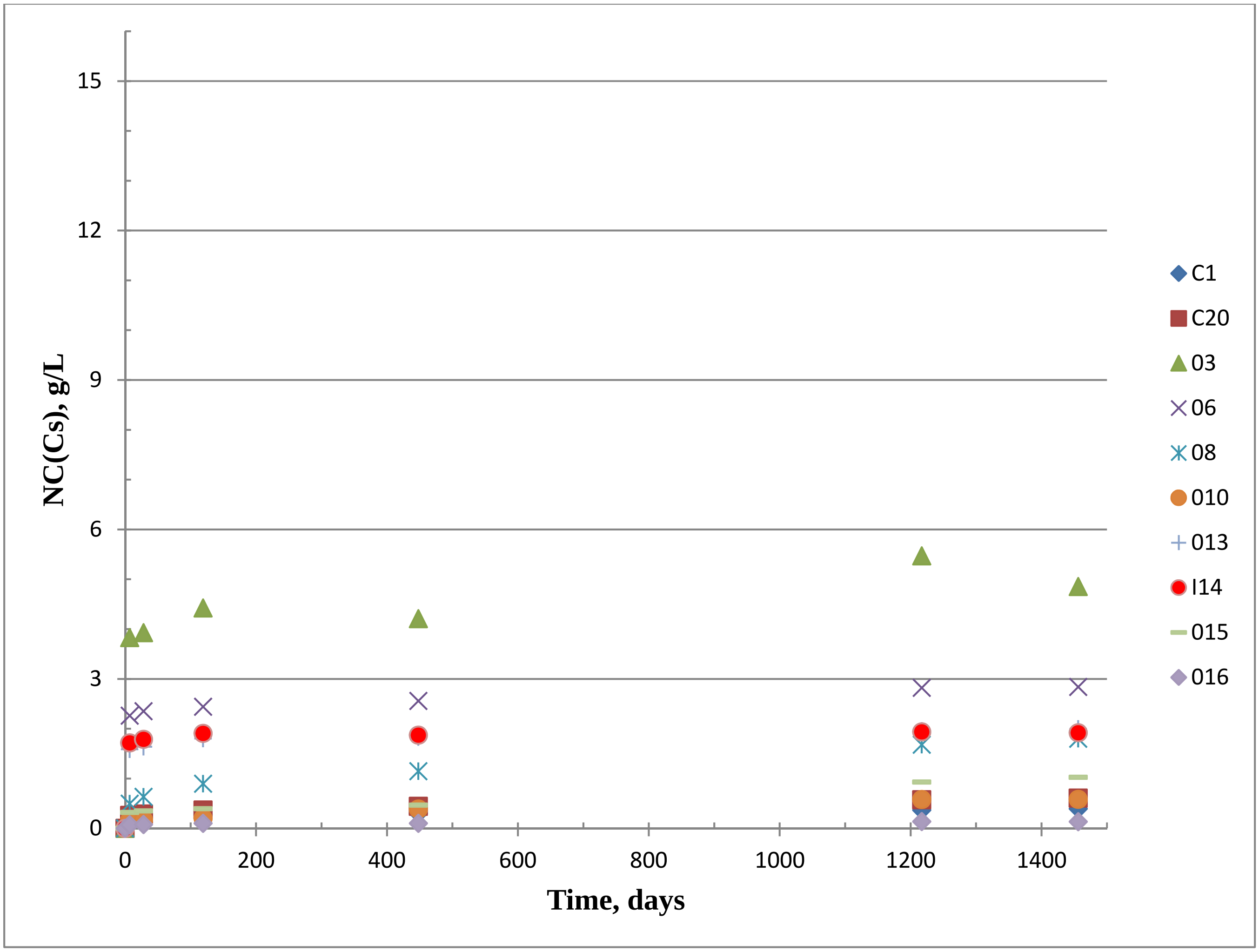
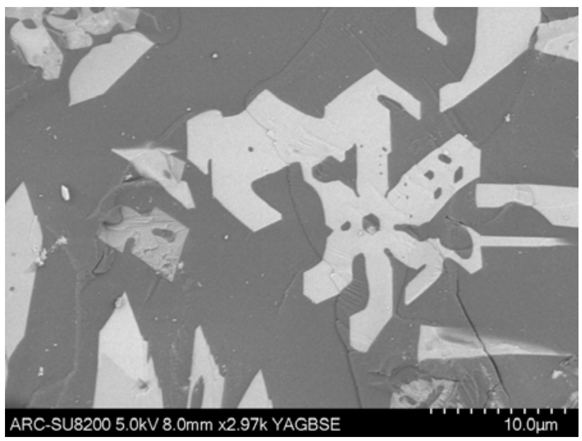


Figure 10. Phase II NC_{Cs} vs. Time

Microscopy on Phase II 64-week Leached Powders

Glass ceramic powders were examined by SEM. One sample, 'Centroid C41' was examined as representative of low corrosion. The 64 week NC(B) values were 0.52 g/L. Two other samples, 'I29' and 'O22' that showed highest corrosion with 64 week NL(B) in the range of 7.8 to 9.3 g/L, were also examined by SEM. Figure 11 shows a non-leached particle from sample C41 and Figure 12 shows leached particles from C41. In these and other SEM images of the glass ceramic particles, the ceramic phases are the lighter colored images that appear within the darker colored glassy matrix. Figure 13 shows an image of the non-leached I29 particle. Figure 14 shows Spectrum 1 indicating presence of oxyapatite ($YZSi_6O_{26}$, with with Y = alkaline earth, Z = Ln) and Figure 15 shows Spectrum 4 indicating powellite ($XMoO_4$ with with X=Ca, Ln, Sr, Ba).



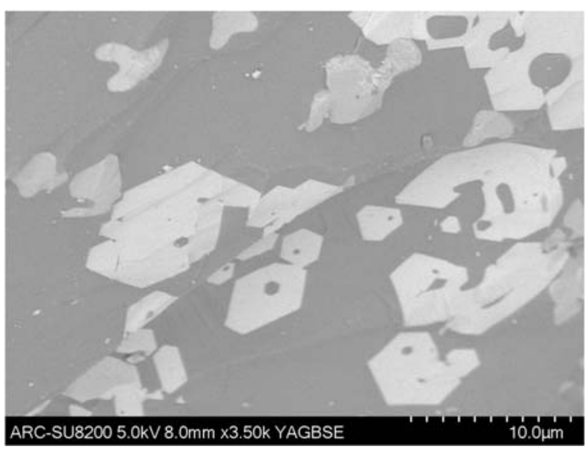
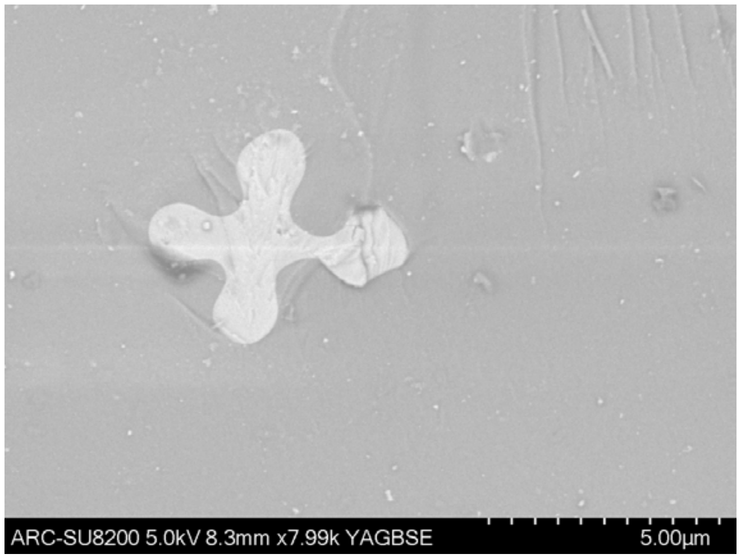


Figure 11. Images of Non-leached C41 Glass Ceramic Particles



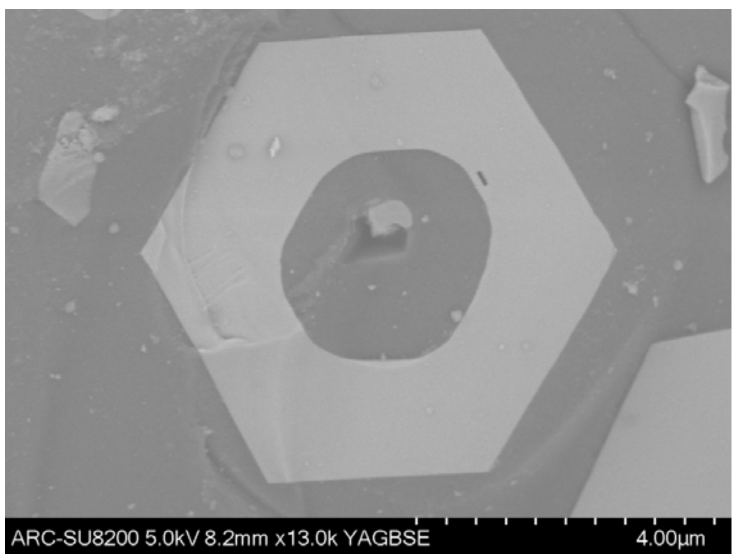


Figure 12. Images of Leached C41 Glass Ceramic Particle

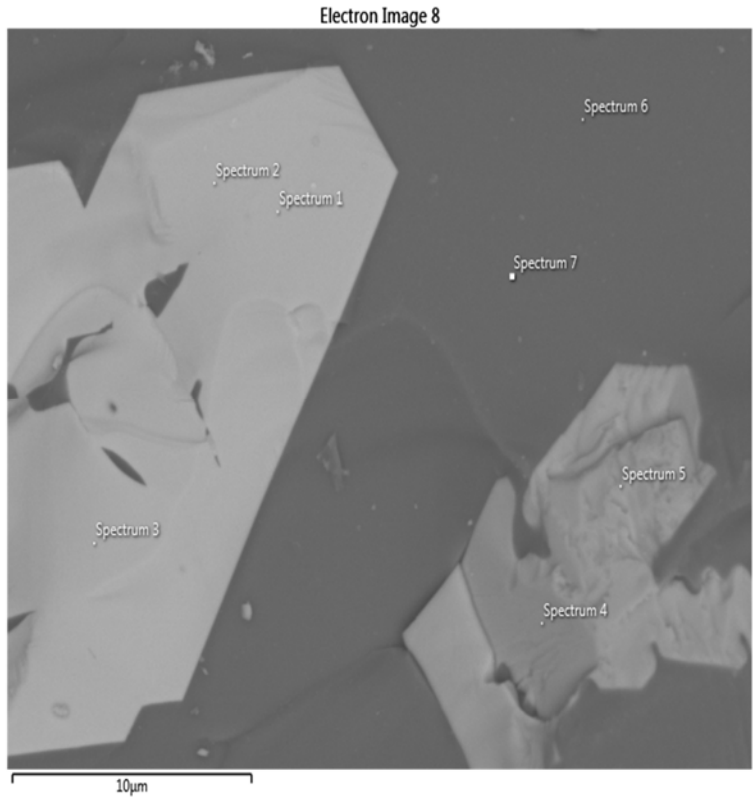


Figure 13. SEM image of Non-leached I29 Glass Ceramic particle.

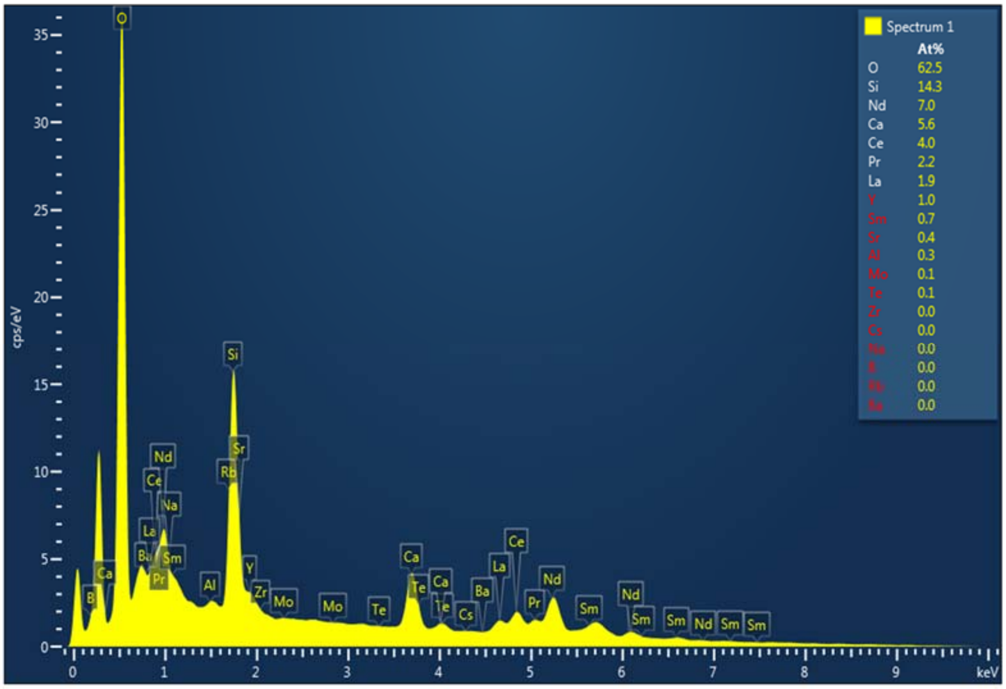


Figure 14. Spectrum 1 EDS indicating Oxyapatite

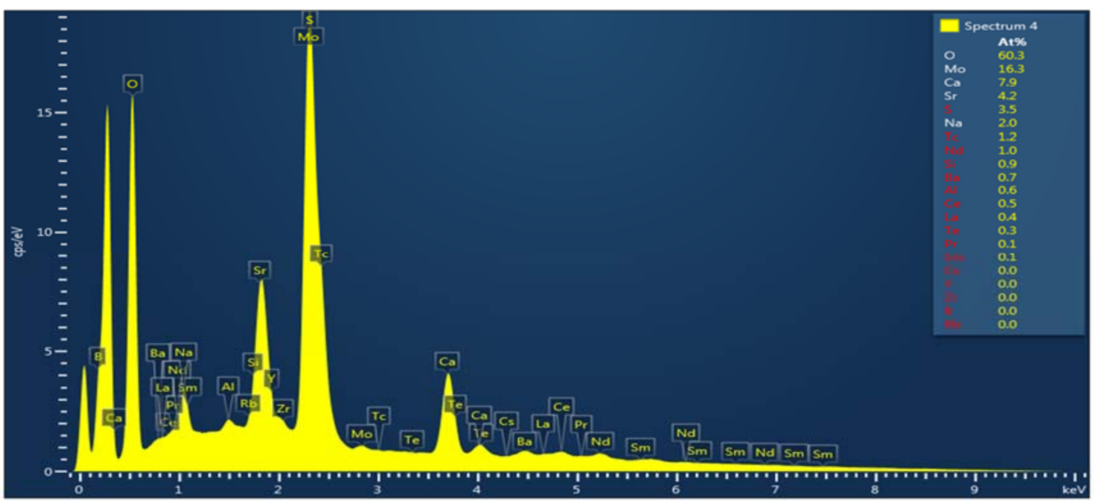


Figure 15. Spectrum 4 EDS indicating Powellite

Previous microscopy data from PNNL (see Figures 46 and 48 of MRWFD 2015) showed evidence of crystal-to-glass (C-to-G) interface corrosion on powders used in Single-Pass Flow-Through Tests. The appearance of ‘halos around the crystals’ were cited to indicate the C-to-G interface corrosion. Particles from the 64-week Phase II tests for samples I29 and O22 were examined to investigate the ceramic to glass transition. Figure 16 shows the I29 particle with no leaching and Figure 17 shows a similar I29 particle after the 64-week leaching. Both of these figures show the image scale bars of 10 microns. Some cracks in the glass/ceramic matrix are apparent in Figure 17 but there does not appear to be any ‘blurring’ or ‘halo’ evident around the ceramic/glass interface. Similar images of 64-week leached O22 particles are shown in Figure 18 at 30-micron resolution and Figure 19 at 10-micron resolution.

It is possible that the PNNL testing using SPFT (dilute) conditions (i.e., maintaining the forward corrosion rate throughout the test duration) were more aggressive corrosion conditions than these long-term static PCTs. SEM data obtained from PNNL were also of higher magnification (micron and sub-micron) than the SRNL SEM/EDAX scans. Other characteristics of the SEM analyses performed at the two different facilities could have also been different such as the beam acceleration and vacuum conditions of the chamber.

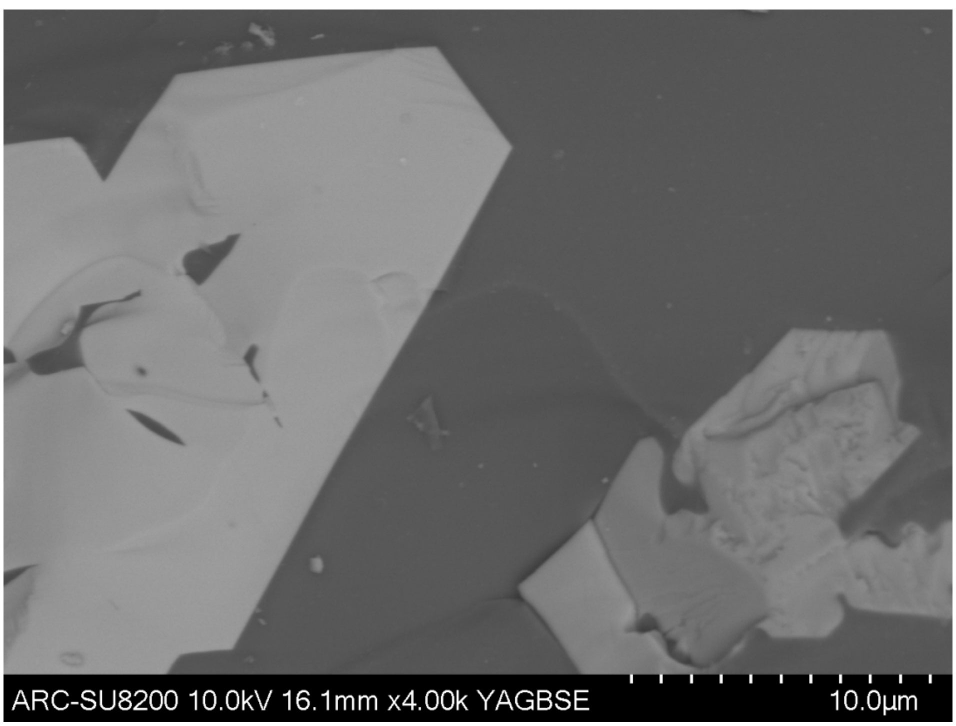


Figure 16. Image of I29 Non-Leached

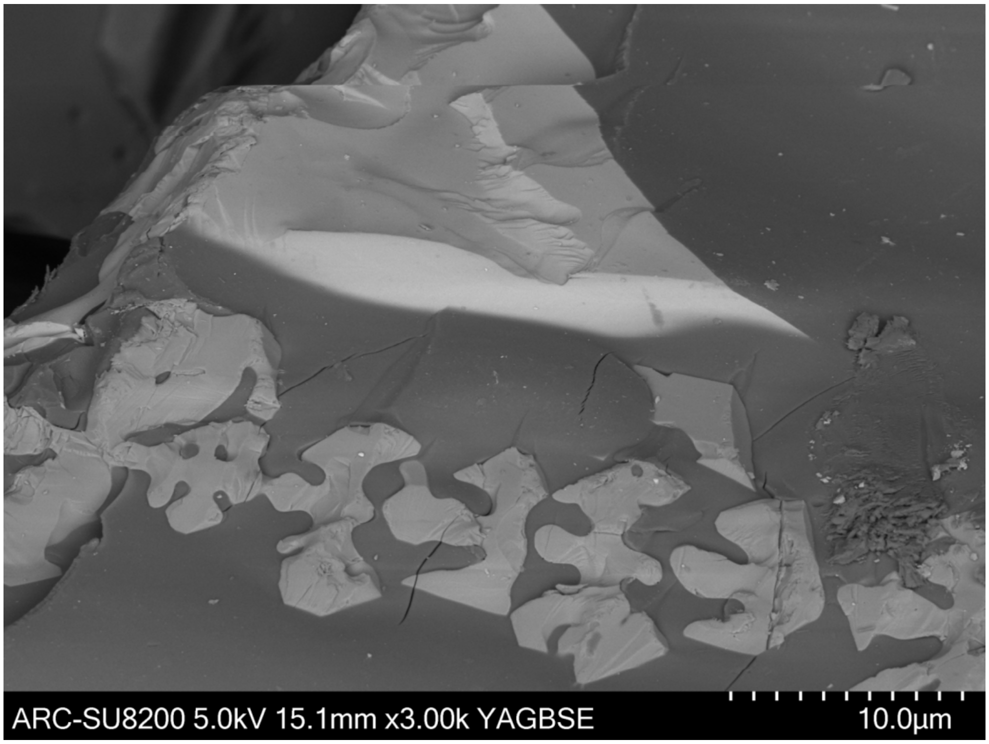


Figure 17. Image of I29 Leached

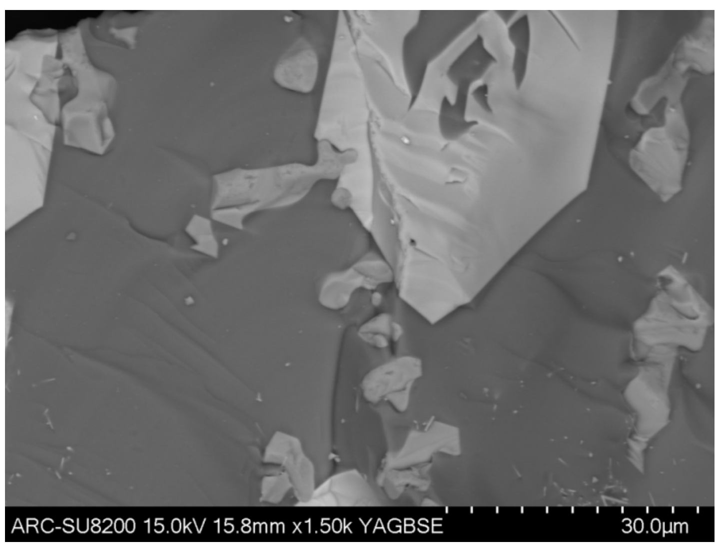


Figure 18. Image of O22 Leached

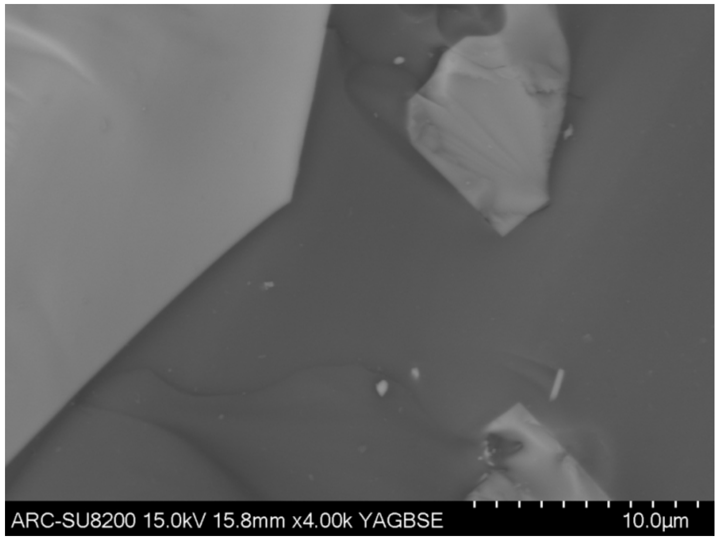


Figure 19. Image of O22 Leached

Coupon Measurements on Phase I Tests

As described in (Crawford, 2013) each Phase I PCT sample also contained a small monolithic glass ceramic specimen that was supported by a Teflon stand above the normal PCT powders. As each set of samples was removed at the various time intervals, these monoliths were weighed to track any changes in mass with leaching. Table 11 shows pre and post mass results for the various monoliths from Phase I testing over the 5 time intervals. The monoliths had a small spot of Si-adhesive added to create a non-leached 'island' on the surface in order to examine the particles in detail after leaching. Mass losses for the coupons from the O3 samples indicate a decrease in mass in the range of 3 to 9 milligrams from the initial to final samples, respectively. This mass loss equates to a percentage mass loss in the range of 1.1 to 3.0 wt%. All other coupon mass changes for all the time intervals were ± 0.002 g. All leached particles from Phase I testing were returned to PNNL for the purpose of inspection and archiving. Appendix C shows microscope images of the particles obtained after the final 1456-day Phase I leach testing (coupon set 5).

Table 11. Monolith Data

Sample	Coupon	Weight	Length	Width	Thickness	Wt. after Silicone	Wt. after PCT	Orig. - Leached	Leached / Orig.	Surf. Area
		(g)	(mm)	(mm)	(mm)	(g)	(g)	(g)		(cm^2)
C1	1	0.363	9.99	8.08	1.46	0.3659	0.366	-0.0001	1.00	2.14
	2	0.351	10.06	7.38	1.5	0.3543	0.3547	-0.0004	1.00	2.01
	3	0.35	9.94	7.34	1.56	0.3535	0.3534	0.0001	1.00	2.00
	4	0.329	9.79	7.26	1.47	0.3312	0.3312	0.0000	1.00	1.92
	5	0.33	9.79	7.34	1.46	0.3329	0.3326	0.0003	1.00	1.94
C20	1	0.725	13.64	10.55	1.61	0.7272	0.7273	-0.0001	1.00	3.66
	2	0.653	11.82	11.18	1.66	0.6577	0.6593	-0.0016	1.00	3.41
	3	0.457	12.1	10.59	1.09	0.4611	0.4609	0.0002	1.00	3.06
	4	0.595	11.29	9.65	1.89	0.5997	0.5996	0.0001	1.00	2.97
	5	0.285	11.38	8.2	0.99	0.2882	0.2878	0.0004	1.00	2.25
O3	1	0.269	8.97	7.95	1.25	0.2732	0.2701	0.0031	0.99	1.85
	2	0.286	9.01	7.72	1.35	0.2906	0.2875	0.0031	0.99	1.84
	3	0.287	8.92	8.37	1.29	0.2906	0.2859	0.0047	0.98	1.94
	4	0.271	9.03	7.61	1.285	0.2766	0.2724	0.0042	0.98	1.80
	5	0.299	8.78	8.33	1.32	0.3028	0.2938	0.0090	0.97	1.91
O6	1	0.11	7.08	5.78	0.78	0.112	0.1121	-0.0001	1.00	1.02
	2	0.098	7.07	5.82	0.72	0.1017	0.1018	-0.0001	1.00	1.01
	3	0.092	6.79	5.53	0.78	0.0937	0.0935	0.0002	1.00	0.94
	4	0.09	6.98	5.33	0.73	0.0921	0.0919	0.0002	1.00	0.92
	5	0.099	7	5.34	0.73	0.1009	0.1001	0.0008	0.99	0.93
O8	1	0.337	10.21	8.65	1.1	0.3384	0.3385	-0.0001	1.00	2.18
	2	0.357	10.12	8.64	1.19	0.3586	0.3591	-0.0005	1.00	2.20
	3	0.334	9.6	8.65	1.11	0.3349	0.3348	0.0001	1.00	2.07
	4	0.315	9.74	8.67	1.1	0.3171	0.317	0.0001	1.00	2.09
	5	0.317	8.76	8.66	1.18	0.3188	0.3176	0.0012	1.00	1.93
O10	1	0.323	9.01	8.5	1.38	0.3242	0.3244	-0.0002	1.00	2.01
	2	0.295	9.12	8.33	1.3	0.2978	0.298	-0.0002	1.00	1.97
	3	0.319	9.07	8.38	1.35	0.3203	0.3199	0.0004	1.00	1.99
	4	0.167	8.02	6.05	1.27	0.1697	0.1693	0.0004	1.00	1.33
	5	0.1	5.78	4.82	1.29	0.1005	0.1002	0.0003	1.00	0.83

Table 11. Monolith Data, continued

Sample	Coupon	Weight	Length	Width	Thickness	Wt. after Silicone	Wt. after PCT	Orig. - Leached	Leached / Orig.	Surf. Area
		(g)	(mm)	(mm)	(mm)	(g)	(g)	(g)		(cm^2)
O13	1	0.351	9.56	8.39	1.29	0.3524	0.3528	-0.0004	1.00	2.07
	2	0.323	9.24	8.12	1.28	0.3237	0.3238	-0.0001	1.00	1.94
	3	0.137	8.45	7.23	0.73	0.1399	0.139	0.0009	0.99	1.45
	4	0.155	7.28	4.58	1.3	0.157	0.1567	0.0003	1.00	0.98
	5	0.132	5.86	5.21	1.29	0.133	0.1325	0.0005	1.00	0.90
I14	1	0.141	8.07	7.86	0.765	0.1423	0.1439	-0.0016	1.01	1.51
	2	0.216	8.66	7.62	1.09	0.217	0.2174	-0.0004	1.00	1.67
	3	0.226	8.6	7.26	1.19	0.2279	0.2278	0.0001	1.00	1.63
	4	0.246	8.56	7.41	1.23	0.2483	0.2481	0.0002	1.00	1.66
	5	0.104	6.23	5.77	1.25	0.1048	0.1044	0.0004	1.00	1.02
O15	1	0.11	5.83	5.31	1.15	0.1109	0.1108	0.0001	1.00	0.88
	2	0.117	5.82	5.11	1.25	0.1187	0.1201	-0.0014	1.01	0.87
	3	0.123	5.84	5.15	1.29	0.1262	0.1262	0.0000	1.00	0.89
	4	0.097	5.3	4.96	1.13	0.0982	0.098	0.0002	1.00	0.76
	5	0.11	5.27	5.06	1.3	0.1123	0.1119	0.0004	1.00	0.80
O16	1	0.336	9.59	8.17	1.4	0.3378	0.3384	-0.0006	1.00	2.06
	2	0.178	7.36	5.56	1.33	0.179	0.1797	-0.0007	1.00	1.16
	3	0.299	7.96	7.77	1.57	0.3008	0.3007	0.0001	1.00	1.73
	4	0.388	9.36	9.01	1.66	0.3901	0.389	0.0011	1.00	2.30
	5	0.262	8.7	6.73	1.56	0.2642	0.2633	0.0009	1.00	1.65

Conclusions and Ongoing Testing

SRNL has successfully completed long term durability leach tests for 1456 days (3.9 years) on Phase I and 882 days (2.4 years) Phase II glass ceramics that were fabricated at both PNNL and SRNL. Conclusions from these tests are described below:

- Long term chemical durability tests performed through 448-day duration indicate that both the Phase I and Phase II glass ceramics behave similarly to durable HLW glasses. Leachate data indicates an early ‘stage I’ initial release, followed by a gradual slowing of release to some ‘stage II’ residual rate over time. One sample from the Phase II testing appears to exhibit resumption of a higher corrosion rate after 112 days based on leachate data from subsequent 448-day and 882-day test durations.

- Only one sample set monolith coupons appear to show any measurable mass loss over the time intervals for the Phase I testing. This O3 sample matrix also showed the highest normalized mass loss from leachate analyses.

- Normalized concentrations determined for the glass ceramics at the longest time intervals of 2.4 to 3.9 years are lower for B and Na than the reported 7-day normalized release for these elements from the benchmark Environmental Assessment glass.

- SEM images of leached powders from the 448-day Phase II glass ceramic particles do not show any obvious signs of corrosion other than some micro-cracks in the glass ceramic matrix.

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Appendix A

Glass Ceramic PCT Leachate Data

Table A-1. Phase II As-analyzed Leachate Concentrations and pH from 882-Day PCT*

Sample ID	Al	B	Ba	Ca	Li	Mo	Na	Si	Sr	Te	Cs	pH
	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	
Blank-1	<1.00	1.94	<1.00	<1.00	<1.00	<1.00	<1.00	<1.00	<1.00	<2.00	<0.01	8.42
O21	1.91	67.59	<1.00	15.26	<1.00	71.5	188.99	22.57	3.19	6.27	25.53	10.88
O23	<1.00	13.74	<1.00	3.09	8.07	5.05	4.16	28.49	<1.00	<2.00	17.86	9.73
O25	<1.00	25.53	9.77	7.09	14.57	6.76	3.36	82.4	6.65	<2.00	63.08	9.67
O27	<1.00	17.61	<1.00	7.91	<1.00	1.93	5.47	36.28	1.41	<2.00	42.07	9.05
I29	1.62	125	1.72	2.07	29.6	94.1	165.32	77.52	1.05	<2.00	50.65	9.35
I31	<1.00	55.81	3	7.1	17.49	3.53	15.84	41.6	3.93	<2.00	31.68	9.26
I33	<1.00	21.31	<1.00	1.5	5.36	3.87	5.82	19.39	<1.00	<2.00	14.64	9.15
I35	<1.00	37.78	2.06	4.77	17.61	35.86	54.14	55.58	2.74	7.87	46.22	10.18
I36	<1.00	14.12	2.16	4.51	3.25	22.94	30.52	23.01	2.01	2.37	25.94	10.30
O22	<1.00	158.02	6.83	9.26	<1.00	10.29	106.58	81.14	4.64	<2.00	149.26	8.97
O24	<1.00	60.59	3.43	3.17	<1.00	22.86	101.71	85.54	5.23	<2.00	97.96	9.46
I32	<1.00	13.3	2.75	3	2.75	20.19	31.37	16.33	1.96	<2.00	20.85	10.35
O28	1.31	4.29	<1.00	<1.00	<1.00	2.95	3.22	9.38	<1.00	<2.00	5.86	8.80
I30	1.11	6.69	<1.00	1.76	1.38	2.83	2.53	11.16	<1.00	<2.00	5.48	9.03
O26	1.29	16.69	1	2.06	6.96	14.39	1.74	7.27	2.36	<2.00	21.63	9.31
I34	<1.00	20	1.32	<1.00	7.73	52.1	36.30	23	<1.00	<2.00	24.94	9.76
O39	<1.00	32.81	6.56	4.65	20.82	11.72	3.42	96.58	4.52	<2.00	96.57	9.78
I40	<1.00	9.69	2.39	1.41	2.24	24.07	24.19	17.36	1.52	<2.00	15.85	9.97
C41	<1.00	8.46	2.44	2.24	2.9	12.95	8.00	11.98	1.29	<2.00	9.81	9.45

*Other elements of the glass ceramics were analyzed in the leachate but were not detected above the detection limit for any of the compositions (La, Nd, Pr, Sm, Y, Zr).

Table A-2. Phase I As-analyzed Leachate Concentrations and pH from 1217-Day PCT*

Sample ID	Al	B	Ba	Ca	Li	Mo	Na	Si	Sr	Te	Cs
	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L
Blnk5	<1.00	1.06	<1.00	<1.00	<1.00	<1.00	<1.00	<1.00	<1.00	<1.00	<0.01
Blnk5	<1.00	0.894	<1.00	<1.00	<1.00	<1.00	<1.00	<1.00	<1.00	<1.00	
C1	<1.00	1.14	<1.00	<1.00	<1.00	0.365	<1.00	<1.00	<1.00	<1.00	0.24
C1	<1.00	1.08	<1.00	<1.00	<1.00	0.389	<1.00	<1.00	<1.00	<1.00	
C20	<1.00	1.11	<1.00	<1.00	<1.00	0.680	<1.00	<1.00	<1.00	<1.00	0.57
C20	<1.00	1.07	<1.00	<1.00	<1.00	0.690	<1.00	<1.00	<1.00	<1.00	
O3	<1.00	12.0	<1.00	0.193	3.02	13.8	17.6	3.83	<1.00	<1.00	4.45
O3	<1.00	12.0	<1.00	0.179	3.01	13.8	17.5	3.72	<1.00	<1.00	
O6	<1.00	1.59	<1.00	<1.00	<1.00	0.936	2.65	<1.00	<1.00	<1.00	1.92
O6	<1.00	1.50	<1.00	<1.00	<1.00	0.884	2.67	<1.00	<1.00	<1.00	
O8	<1.00	2.48	<1.00	<1.00	<1.00	0.163	<1.00	<1.00	<1.00	<1.00	2.03
O8	<1.00	2.48	<1.00	<1.00	<1.00	0.153	<1.00	<1.00	<1.00	<1.00	
O10	<1.00	0.836	<1.00	<1.00	<1.00	0.775	1.17	<1.00	<1.00	<1.00	0.50
O10	<1.00	0.809	<1.00	<1.00	<1.00	0.790	1.19	<1.00	<1.00	<1.00	
O13	<1.00	2.02	<1.00	<1.00	<1.00	1.09	<1.00	<1.00	<1.00	<1.00	2.35
O13	<1.00	2.00	<1.00	<1.00	<1.00	1.10	<1.00	<1.00	<1.00	<1.00	
I14	<1.00	7.54	<1.00	<1.00	<1.00	0.124	4.35	<1.00	<1.00	<1.00	1.78
I14	<1.00	7.65	<1.00	<1.00	<1.00	0.112	4.31	<1.00	<1.00	<1.00	
O15	<1.00	1.82	<1.00	<1.00	<1.00	<1.00	<1.00	<1.00	<1.00	<1.00	0.85
O15	<1.00	1.77	<1.00	<1.00	<1.00	<1.00	<1.00	<1.00	<1.00	<1.00	
O16	<1.00	0.846	<1.00	<1.00	<1.00	0.099	<1.00	<1.00	<1.00	<1.00	0.11
O16	<1.00	0.817	<1.00	<1.00	<1.00	0.099	<1.00	<1.00	<1.00	<1.00	

*Other elements of the glass ceramics were analyzed in the leachate but were not detected above the detection limit for any of the compositions (La, Nd, Pr, Sm, Y, Zr).

Table A-3. Phase I As-analyzed Leachate Concentrations and pH from 1456-Day PCT*

Sample ID	Al	B	Ba	Ca	Li	Mo	Na	Si	Sr	Te	Cs	pH
	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	
Blnk5	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0	1.33	2.00	<1.0	<2.0	<0.01	7.35
C1	<1.0	9.32	<1.0	2.26	5.08	12.6	9.32	20.6	<1.0	<2.0	6.25	9.00
C20	<1.0	11.2	1.40	2.53	4.32	21.6	12.2	17.8	1.28	<2.0	14.88	9.16
O3	6.92	260	1.77	1.72	85.2	277	435	158	1.11	2.30	97.16	9.91
O6	<1.0	22.4	2.61	3.83	<1.0	26.7	64.2	44.2	1.93	<2.0	47.57	9.89
O8	<1.0	48.2	1.12	16.4	<1.0	6.87	14.2	27.3	1.40	<2.0	53.47	8.33
O10	<1.0	7.32	1.74	2.16	<1.0	23.5	23.9	18.0	1.99	<2.0	12.37	9.30
O13	<1.0	37.2	2.44	3.71	16.7	31.2	1.84	55.0	2.77	<2.0	58.97	9.32
I14	<1.0	166	<1.0	3.75	9.82	6.19	108	42.4	<1.0	<2.0	43.40	8.95
O15	<1.0	32.7	<1.0	5.20	8.13	2.12	1.79	35.0	<1.0	<2.0	22.93	8.77
O16	<1.0	10.1	<1.0	3.81	7.15	5.74	5.66	18.2	<1.0	<2.0	2.77	9.53

*Other elements of the glass ceramics were analyzed in the leachate but were not detected above the detection limit for any of the compositions (La, Nd, Pr, Sm, Y, Zr).

Appendix B

Phase I and Phase II Glass Ceramic Statistical Design Target Compositions

Table B-1. Phase I Statistical Design Chemical Compositions

Layer	Outer**	Outer**	Outer**	Outer**	Outer**	Outer**	Outer	Outer**	Outer**	Inner
Waste Loading	**0.4458**	**0.4260**	**0.5767**	**0.4191**	**0.5561**	**0.5384**	**0.4393**	**0.4458**	**0.4739**	**0.4582**
glass-ceramic ID	**O16-2013**	**O15-2013**	**O13-2013**	**O10-2013**	**O8-2013**	**O6-2013**	**O12-2013**	**O3-2013**	**C1-2013**	**I2-2013**
Al2O3 *	0.0742	0.0747	0.0000	0.0735	0.0536	0.0000	0.0000	0.0000	0.0418	0.0320
B2O3 *	0.0623	0.1177	0.0562	0.0618	0.0998	0.0571	0.1182	0.1200	0.0853	0.0781
CaO *	0.0879	0.0632	0.0339	0.0373	0.0321	0.0805	0.0889	0.0387	0.0512	0.0642
Li2O *	0.0201	0.0202	0.0181	0.0000	0.0000	0.0000	0.0000	0.0206	0.0105	0.0168
Na2O *	0.0139	0.0000	0.0000	0.0619	0.0059	0.0528	0.0000	0.0712	0.0273	0.0139
SiO2 *	0.2959	0.2981	0.3151	0.3465	0.2525	0.2712	0.3537	0.3037	0.3099	0.3369
MoO3	0.0620	0.0592	0.0802	0.0582	0.0773	0.0748	0.0610	0.0620	0.0659	0.0637
SrO	0.0156	0.0149	0.0201	0.0146	0.0194	0.0188	0.0153	0.0156	0.0165	0.0160
BaO	0.0350	0.0334	0.0452	0.0329	0.0436	0.0422	0.0345	0.0350	0.0372	0.0359
Rb2O	0.0067	0.0064	0.0087	0.0063	0.0084	0.0081	0.0066	0.0067	0.0071	0.0069
Cs2O	0.0456	0.0436	0.0590	0.0429	0.0569	0.0551	0.0450	0.0456	0.0485	0.0469
Ce2O3	0.0492	0.0470	0.0636	0.0462	0.0613	0.0594	0.0485	0.0492	0.0523	0.0505
Eu2O3	0.0027	0.0026	0.0035	0.0026	0.0034	0.0033	0.0027	0.0027	0.0029	0.0028
Gd2O3	0.0026	0.0025	0.0033	0.0024	0.0032	0.0031	0.0025	0.0026	0.0027	0.0026
La2O3	0.0251	0.0240	0.0325	0.0236	0.0313	0.0303	0.0247	0.0251	0.0267	0.0258
Nd2O3	0.0831	0.0794	0.1075	0.0781	0.1037	0.1004	0.0819	0.0831	0.0884	0.0854
Pr2O3	0.0230	0.0219	0.0297	0.0216	0.0286	0.0277	0.0226	0.0230	0.0244	0.0236
Sm2O3	0.0171	0.0163	0.0221	0.0160	0.0213	0.0206	0.0168	0.0171	0.0181	0.0175
Y2O3	0.0100	0.0095	0.0129	0.0094	0.0124	0.0120	0.0098	0.0100	0.0106	0.0102
ZrO2	0.0473	0.0452	0.0612	0.0445	0.0590	0.0572	0.0466	0.0473	0.0503	0.0486
PdO	0.0003	0.0003	0.0004	0.0003	0.0004	0.0003	0.0003	0.0003	0.0003	0.0003
RhO2	0.0012	0.0012	0.0016	0.0012	0.0015	0.0015	0.0012	0.0012	0.0013	0.0013
RuO2	0.0031	0.0030	0.0041	0.0029	0.0039	0.0038	0.0031	0.0031	0.0033	0.0032
Ag2O	0.0018	0.0017	0.0023	0.0017	0.0022	0.0021	0.0017	0.0018	0.0019	0.0018
CdO	0.0018	0.0017	0.0023	0.0017	0.0022	0.0021	0.0017	0.0018	0.0019	0.0018
Sb2O3	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
SeO2	0.0013	0.0013	0.0017	0.0012	0.0016	0.0016	0.0013	0.0013	0.0014	0.0013
SnO2	0.0011	0.0011	0.0014	0.0010	0.0014	0.0013	0.0011	0.0011	0.0012	0.0011
TeO2	0.0104	0.0099	0.0135	0.0098	0.0130	0.0126	0.0103	0.0104	0.0111	0.0107
Total	**1.0000**	**1.0000**	**1.0000**	**1.0000**	**1.0000**	**1.0000**	**1.0000**	**1.0000**	**1.0000**	**1.0000**

*Rows highlghted indicate glass components
**Columns highlighted indicated glass ceramics corrosion tested in this work

Table B-1. Phase I Statistical Design Chemical Compositions, continued.

Layer	Inner	Inner	Inner	Inner	Inner	Inner**	Outer	Centroid**	Inner	Inner
Waste Loading	0.4979	0.4478	0.4559	0.4686	0.4680	0.4504	0.4393	0.4739	0.4559	0.4680
glass-ceramic ID	I4-2013	I7-2013	I9-2013	I11-2013	I5-2013	I14-2013	O18-2013	C20-2013	I19-2013	I17-2013
Al2O3 *	0.0529	0.0559	0.0319	0.0544	0.0556	0.0315	0.0000	0.0418	0.0319	0.0556
B2O3 *	0.0722	0.0763	0.0948	0.0966	0.0760	0.0998	0.1182	0.0853	0.0948	0.0760
CaO *	0.0407	0.0645	0.0438	0.0419	0.0428	0.0433	0.0889	0.0512	0.0438	0.0428
Li2O *	0.0046	0.0049	0.0167	0.0048	0.0163	0.0049	0.0000	0.0105	0.0167	0.0163
Na2O *	0.0450	0.0476	0.0484	0.0132	0.0135	0.0389	0.0000	0.0273	0.0484	0.0135
SiO2 *	0.2867	0.3030	0.3085	0.3206	0.3278	0.3312	0.3537	0.3099	0.3085	0.3278
MoO3	0.0692	0.0622	0.0634	0.0651	0.0650	0.0626	0.0610	0.0659	0.0634	0.0650
SrO	0.0174	0.0156	0.0159	0.0164	0.0163	0.0157	0.0153	0.0165	0.0159	0.0163
BaO	0.0390	0.0351	0.0358	0.0368	0.0367	0.0353	0.0345	0.0372	0.0358	0.0367
Rb2O	0.0075	0.0067	0.0069	0.0071	0.0071	0.0068	0.0066	0.0071	0.0069	0.0071
Cs2O	0.0509	0.0458	0.0467	0.0479	0.0479	0.0461	0.0450	0.0485	0.0467	0.0479
Ce2O3	0.0549	0.0494	0.0503	0.0517	0.0516	0.0497	0.0485	0.0523	0.0503	0.0516
Eu2O3	0.0030	0.0027	0.0028	0.0029	0.0028	0.0027	0.0027	0.0029	0.0028	0.0028
Gd2O3	0.0029	0.0026	0.0026	0.0027	0.0027	0.0026	0.0025	0.0027	0.0026	0.0027
La2O3	0.0280	0.0252	0.0257	0.0264	0.0263	0.0253	0.0247	0.0267	0.0257	0.0263
Nd2O3	0.0928	0.0835	0.0850	0.0874	0.0873	0.0840	0.0819	0.0884	0.0850	0.0873
Pr2O3	0.0256	0.0231	0.0235	0.0241	0.0241	0.0232	0.0226	0.0244	0.0235	0.0241
Sm2O3	0.0191	0.0171	0.0175	0.0179	0.0179	0.0172	0.0168	0.0181	0.0175	0.0179
Y2O3	0.0111	0.0100	0.0102	0.0105	0.0105	0.0101	0.0098	0.0106	0.0102	0.0105
ZrO2	0.0529	0.0475	0.0484	0.0497	0.0497	0.0478	0.0466	0.0503	0.0484	0.0497
PdO	0.0003	0.0003	0.0003	0.0003	0.0003	0.0003	0.0003	0.0003	0.0003	0.0003
RhO2	0.0014	0.0012	0.0013	0.0013	0.0013	0.0012	0.0012	0.0013	0.0013	0.0013
RuO2	0.0035	0.0031	0.0032	0.0033	0.0033	0.0032	0.0031	0.0033	0.0032	0.0033
Ag2O	0.0020	0.0018	0.0018	0.0019	0.0019	0.0018	0.0017	0.0019	0.0018	0.0019
CdO	0.0020	0.0018	0.0018	0.0019	0.0018	0.0018	0.0017	0.0019	0.0018	0.0018
Sb2O3	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
SeO2	0.0015	0.0013	0.0013	0.0014	0.0014	0.0013	0.0013	0.0014	0.0013	0.0014
SnO2	0.0012	0.0011	0.0011	0.0012	0.0012	0.0011	0.0011	0.0012	0.0011	0.0012
TeO2	0.0116	0.0105	0.0106	0.0109	0.0109	0.0105	0.0103	0.0111	0.0106	0.0109
Total	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000

*Rows highlghted indicate glass components
**Columns highlighted indicated glass ceramics corrosion tested in this work

Table B-2. Phase II Statistical Design Chemical Compositions

Mass fraction	Al2O3*	B2O3*	CaO*	Li2O*	Na2O*	SiO2*	MoO3	SrO	BaO	Rb2O
O21-2014	0.0680	0.0571	0.0806	0.0000	0.0572	0.2713	0.0222	0.0134	0.0300	0.0058
O22-2014	0.0000	0.1052	0.0791	0.0000	0.0234	0.2663	0.0906	0.0131	0.0295	0.0057
O23-2014	0.0000	0.0584	0.0676	0.0188	0.0000	0.3276	0.0226	0.0137	0.0307	0.0059
O24-2014	0.0000	0.1191	0.0384	0.0000	0.0707	0.3528	0.0246	0.0235	0.0528	0.0102
O25-2014	0.0000	0.0628	0.0885	0.0202	0.0000	0.3370	0.1014	0.0233	0.0523	0.0100
O26-2014	0.0660	0.1040	0.0335	0.0179	0.0000	0.2663	0.0215	0.0205	0.0462	0.0089
O27-2014	0.0501	0.1140	0.0857	0.0000	0.0068	0.2886	0.0236	0.0225	0.0506	0.0097
O28-2014	0.0628	0.0528	0.0319	0.0000	0.0059	0.2946	0.0852	0.0195	0.0439	0.0084
I29-2014	0.0227	0.1025	0.0436	0.0149	0.0551	0.3005	0.0840	0.0157	0.0352	0.0068
I30-2014	0.0558	0.0686	0.0691	0.0065	0.0102	0.3356	0.0828	0.0155	0.0347	0.0067
I31-2014	0.0219	0.1045	0.0421	0.0144	0.0110	0.3286	0.0440	0.0151	0.0340	0.0065
I32-2014	0.0215	0.0660	0.0414	0.0063	0.0467	0.3229	0.0432	0.0149	0.0334	0.0064
I33-2014	0.0519	0.0993	0.0400	0.0099	0.0095	0.2754	0.0770	0.0144	0.0323	0.0062
I34-2014	0.0211	0.0728	0.0407	0.0113	0.0514	0.2801	0.0783	0.0146	0.0328	0.0063
I35-2014	0.0216	0.0683	0.0728	0.0143	0.0370	0.2865	0.0435	0.0184	0.0414	0.0079
I36-2014	0.0251	0.0699	0.0753	0.0066	0.0544	0.2965	0.0450	0.0190	0.0428	0.0082
O39-2014	0.0000	0.0628	0.0885	0.0202	0.0000	0.3370	0.1014	0.0233	0.0523	0.0100
I40-2014	0.0251	0.0699	0.0753	0.0066	0.0544	0.2965	0.0450	0.0190	0.0428	0.0082
C41-2014	0.0418	0.0853	0.0512	0.0105	0.0273	0.3099	0.0659	0.0165	0.0372	0.0071

* Rows highlghted indicate glass components

Table B-2. Phase II Statistical Design Chemical Compositions, continued

Mass fraction	Cs2O	Gd2O3	Ce2O3	Eu2O3	La2O3	Nd2O3	Pr2O3	Sm2O3	Y2O3	ZrO2
O21-2014	0.0392	0.0038	0.0736	0.0041	0.0375	0.1244	0.0344	0.0255	0.0149	0.0190
O22-2014	0.0385	0.0038	0.0722	0.0040	0.0369	0.1221	0.0337	0.0251	0.0146	0.0186
O23-2014	0.0400	0.0039	0.0752	0.0042	0.0384	0.1271	0.0351	0.0261	0.0152	0.0711
O24-2014	0.0689	0.0022	0.0430	0.0024	0.0220	0.0728	0.0201	0.0149	0.0087	0.0211
O25-2014	0.0682	0.0022	0.0426	0.0023	0.0217	0.0719	0.0199	0.0148	0.0086	0.0208
O26-2014	0.0602	0.0037	0.0714	0.0039	0.0364	0.1208	0.0334	0.0248	0.0145	0.0184
O27-2014	0.0660	0.0022	0.0412	0.0023	0.0210	0.0697	0.0192	0.0143	0.0083	0.0740
O28-2014	0.0573	0.0035	0.0680	0.0038	0.0347	0.1149	0.0317	0.0236	0.0138	0.0175
I29-2014	0.0460	0.0026	0.0503	0.0028	0.0257	0.0851	0.0235	0.0175	0.0102	0.0342
I30-2014	0.0453	0.0026	0.0496	0.0027	0.0253	0.0839	0.0232	0.0172	0.0100	0.0337
I31-2014	0.0444	0.0034	0.0648	0.0036	0.0330	0.1095	0.0302	0.0225	0.0131	0.0330
I32-2014	0.0436	0.0033	0.0636	0.0035	0.0325	0.1076	0.0297	0.0221	0.0129	0.0584
I33-2014	0.0421	0.0032	0.0615	0.0034	0.0314	0.1040	0.0287	0.0214	0.0125	0.0565
I34-2014	0.0429	0.0033	0.0626	0.0035	0.0319	0.1058	0.0292	0.0217	0.0127	0.0574
I35-2014	0.0540	0.0033	0.0640	0.0035	0.0327	0.1082	0.0299	0.0222	0.0130	0.0326
I36-2014	0.0558	0.0026	0.0497	0.0027	0.0253	0.0840	0.0232	0.0172	0.0101	0.0608
O39-2014	0.0682	0.0022	0.0426	0.0023	0.0217	0.0719	0.0199	0.0148	0.0086	0.0208
I40-2014	0.0558	0.0026	0.0497	0.0027	0.0253	0.0840	0.0232	0.0172	0.0101	0.0608
C41-2014	0.0485	0.0027	0.0523	0.0029	0.0267	0.0884	0.0244	0.0181	0.0106	0.0503

Table B-2. Phase II Statistical Design Chemical Compositions, continued

Mass fraction	PdO	RhO2	RuO2	Ag2O	CdO	Sb2O3	SeO2	SnO2	TeO2	SUM
O21-2014	0.0002	0.0011	0.0027	0.0015	0.0015	0.0000	0.0011	0.0010	0.0089	1.0000
O22-2014	0.0002	0.0010	0.0026	0.0015	0.0015	0.0000	0.0011	0.0009	0.0088	1.0000
O23-2014	0.0003	0.0011	0.0028	0.0016	0.0015	0.0000	0.0011	0.0010	0.0091	1.0000
O24-2014	0.0004	0.0019	0.0047	0.0027	0.0027	0.0000	0.0020	0.0017	0.0157	1.0000
O25-2014	0.0004	0.0018	0.0047	0.0026	0.0026	0.0000	0.0020	0.0017	0.0156	1.0000
O26-2014	0.0004	0.0016	0.0041	0.0023	0.0023	0.0000	0.0017	0.0015	0.0137	1.0000
O27-2014	0.0004	0.0018	0.0045	0.0026	0.0025	0.0000	0.0019	0.0016	0.0151	1.0000
O28-2014	0.0004	0.0015	0.0039	0.0022	0.0022	0.0000	0.0016	0.0014	0.0131	1.0000
I29-2014	0.0003	0.0012	0.0032	0.0018	0.0018	0.0000	0.0013	0.0011	0.0105	1.0000
I30-2014	0.0003	0.0012	0.0031	0.0018	0.0018	0.0000	0.0013	0.0011	0.0103	1.0000
I31-2014	0.0003	0.0012	0.0030	0.0017	0.0017	0.0000	0.0013	0.0011	0.0101	1.0000
I32-2014	0.0003	0.0012	0.0030	0.0017	0.0017	0.0000	0.0013	0.0011	0.0099	1.0000
I33-2014	0.0003	0.0011	0.0029	0.0016	0.0016	0.0000	0.0012	0.0010	0.0096	1.0000
I34-2014	0.0003	0.0012	0.0029	0.0017	0.0017	0.0000	0.0012	0.0010	0.0098	1.0000
I35-2014	0.0003	0.0015	0.0037	0.0021	0.0021	0.0000	0.0015	0.0013	0.0123	1.0000
I36-2014	0.0004	0.0015	0.0038	0.0022	0.0022	0.0000	0.0016	0.0014	0.0127	1.0000
O39-2014	0.0004	0.0018	0.0047	0.0026	0.0026	0.0000	0.0020	0.0017	0.0156	1.0000
I40-2014	0.0004	0.0015	0.0038	0.0022	0.0022	0.0000	0.0016	0.0014	0.0127	1.0000
C41-2014	0.0003	0.0013	0.0033	0.0019	0.0019	0.0000	0.0014	0.0012	0.0111	1.0000

Table B-2. Phase II Statistical Design Chemical Compositions, continued

Mass fraction	Sum Glass	Waste Load	MoO3:SrO	MoO3:BaO	MoO3:Cs2O
O21-2014	0.5342	0.4658	1.7	0.7	3.8
O22-2014	0.4739	0.5261	6.9	3.1	16.0
O23-2014	0.4724	0.5276	1.7	0.7	3.8
O24-2014	0.5810	0.4190	1.0	0.5	2.4
O25-2014	0.5086	0.4914	4.4	1.9	10.1
O26-2014	0.4877	0.5123	1.0	0.5	2.4
O27-2014	0.5451	0.4549	1.0	0.5	2.4
O28-2014	0.4479	0.5521	4.4	1.9	10.1
I29-2014	0.5393	0.4607	5.4	2.4	12.4
I30-2014	0.5459	0.4541	5.4	2.4	12.4
I31-2014	0.5224	0.4776	2.9	1.3	6.7
I32-2014	0.5048	0.4952	2.9	1.3	6.7
I33-2014	0.4860	0.5140	5.4	2.4	12.4
I34-2014	0.4773	0.5227	5.4	2.4	12.4
I35-2014	0.5006	0.4994	2.4	1.1	5.5
I36-2014	0.5278	0.4722	2.4	1.1	5.5
O39-2014	0.5086	0.4914	4.4	1.9	10.1
I40-2014	0.5278	0.4722	2.4	1.1	5.5
C41-2014	0.5261	0.4739	4.0	1.8	9.2

Appendix C

Phase I Monolith Images After 1456 Days of Leaching

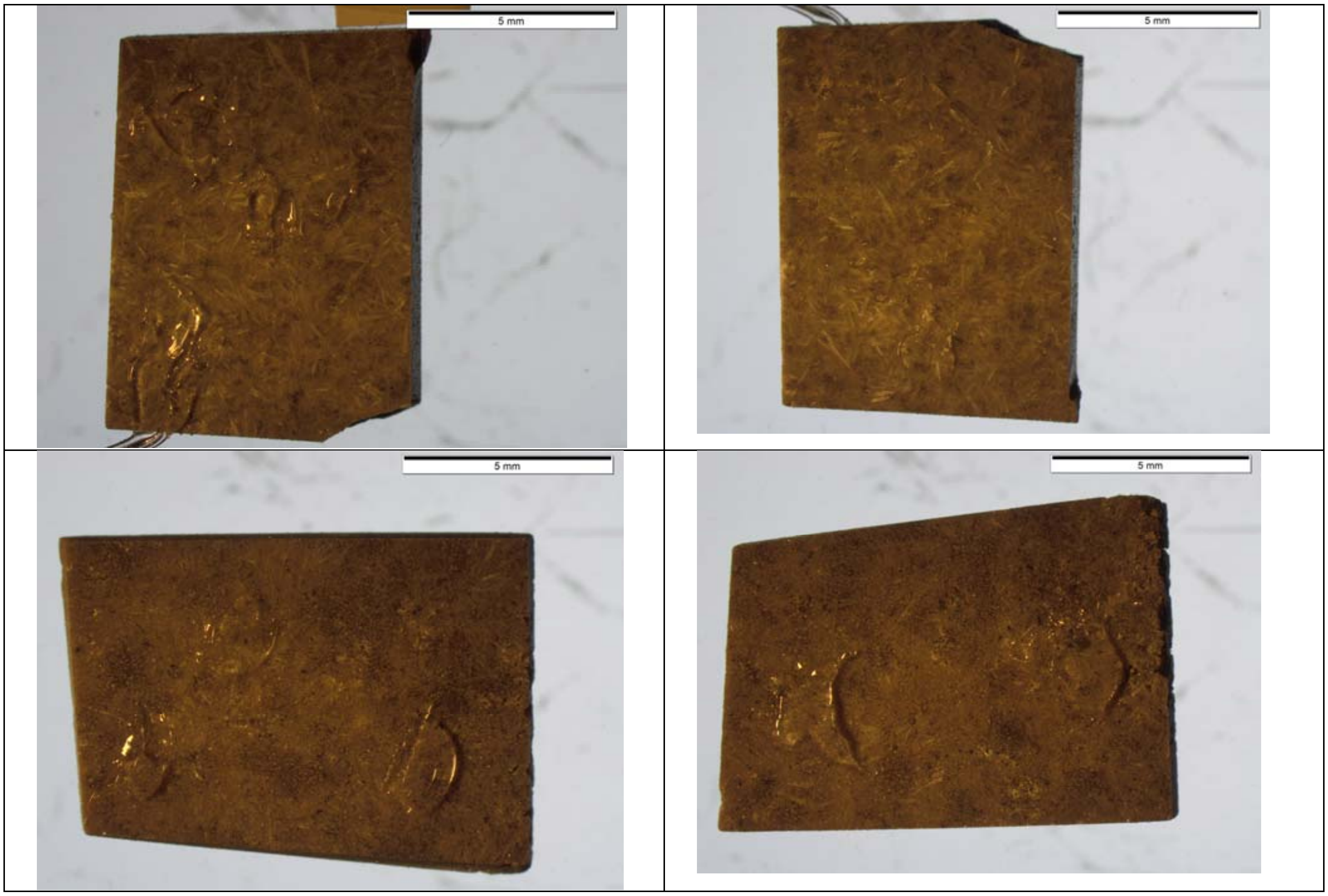

Figure C-1. Images of 1456 Day Leached Monolith Samples C1 (top) and C20 (bottom)

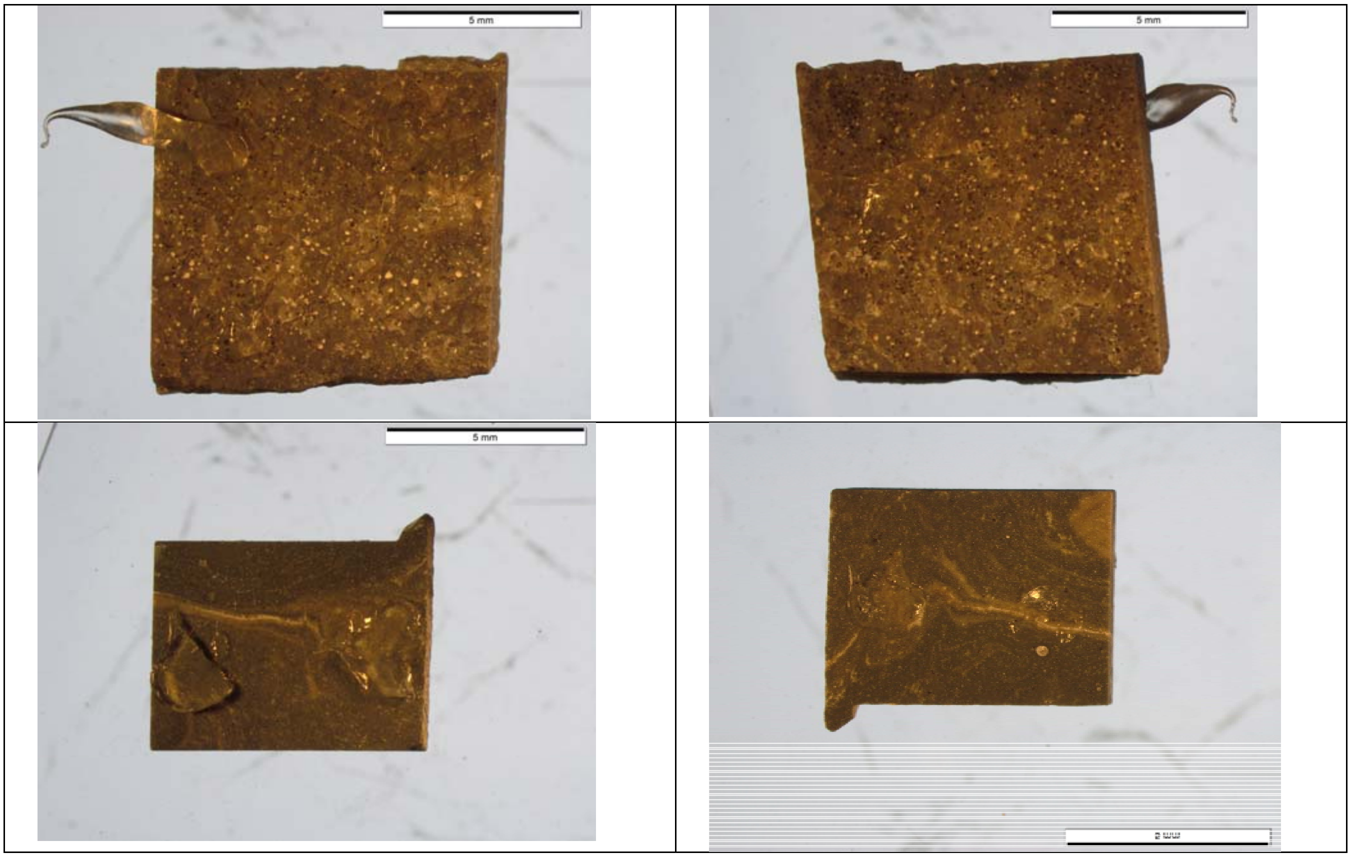

Figure C-2. Images of 1456 Day Leached Monolith Samples O3 (top) and O6 (bottom)

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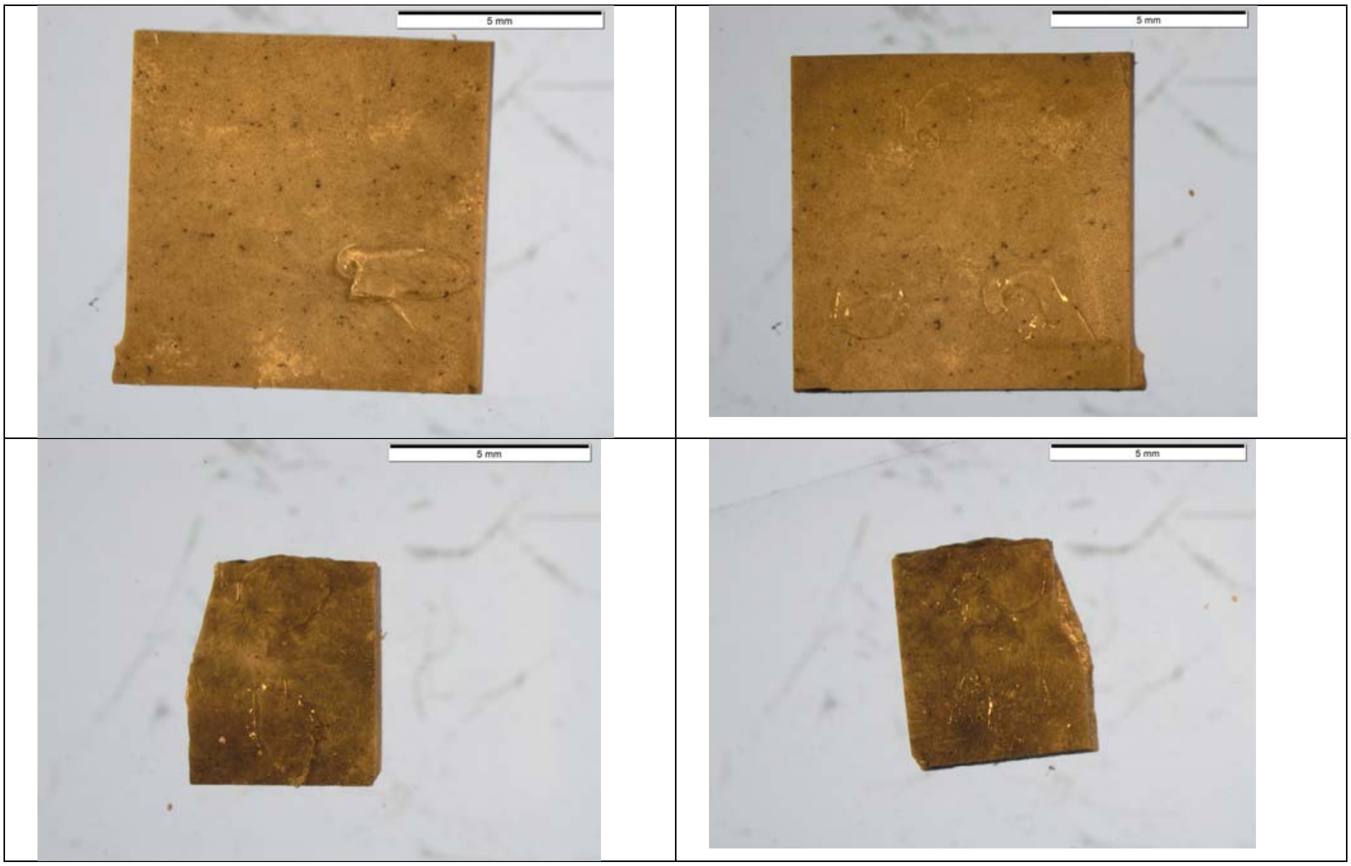

Figure C-3. Images of 1456 Day Leached Monolith Samples O8 (top) and O10 (bottom)

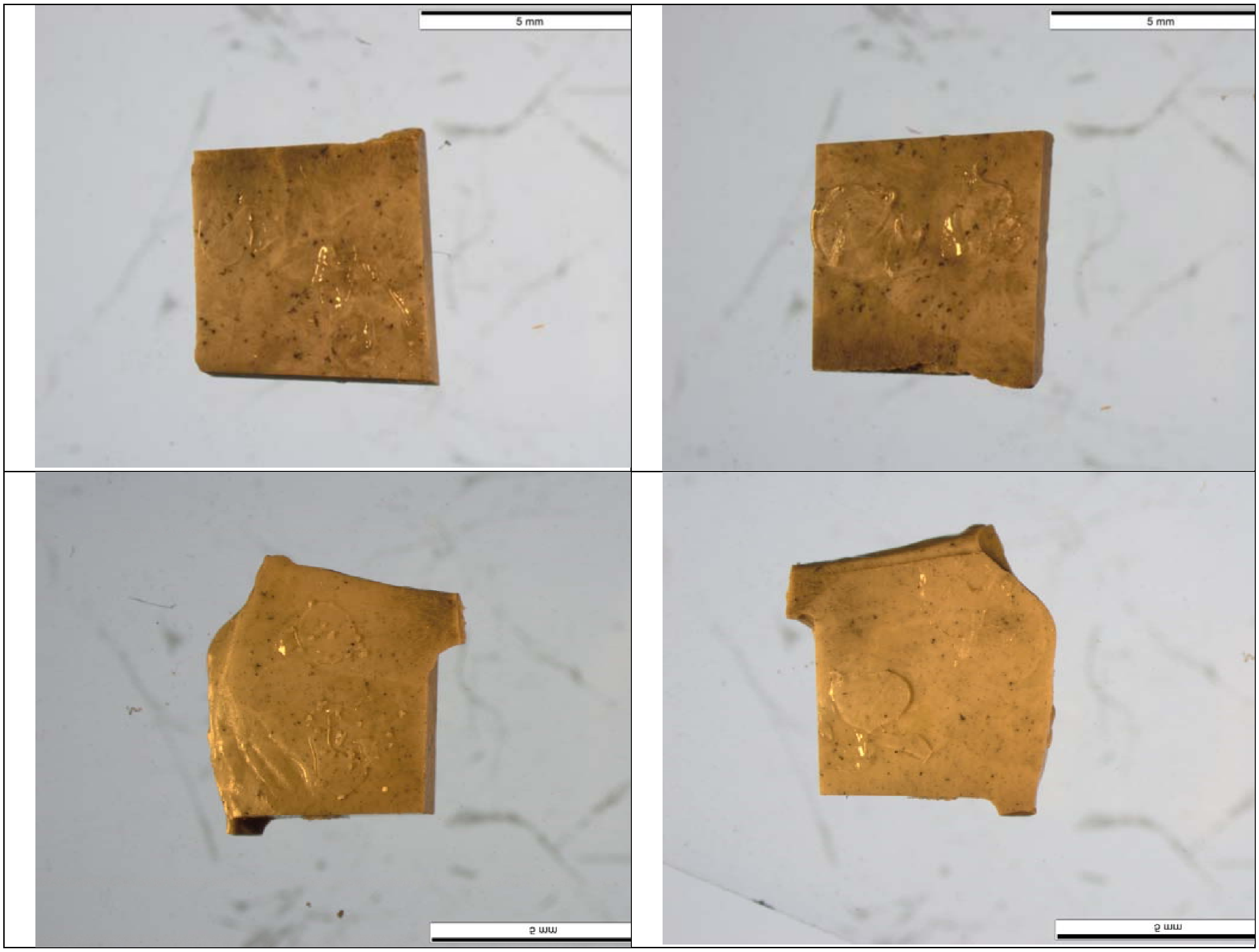

Figure C-4. Images of 1456 Day Leached Monolith Samples O13 (top) and I14 (bottom)

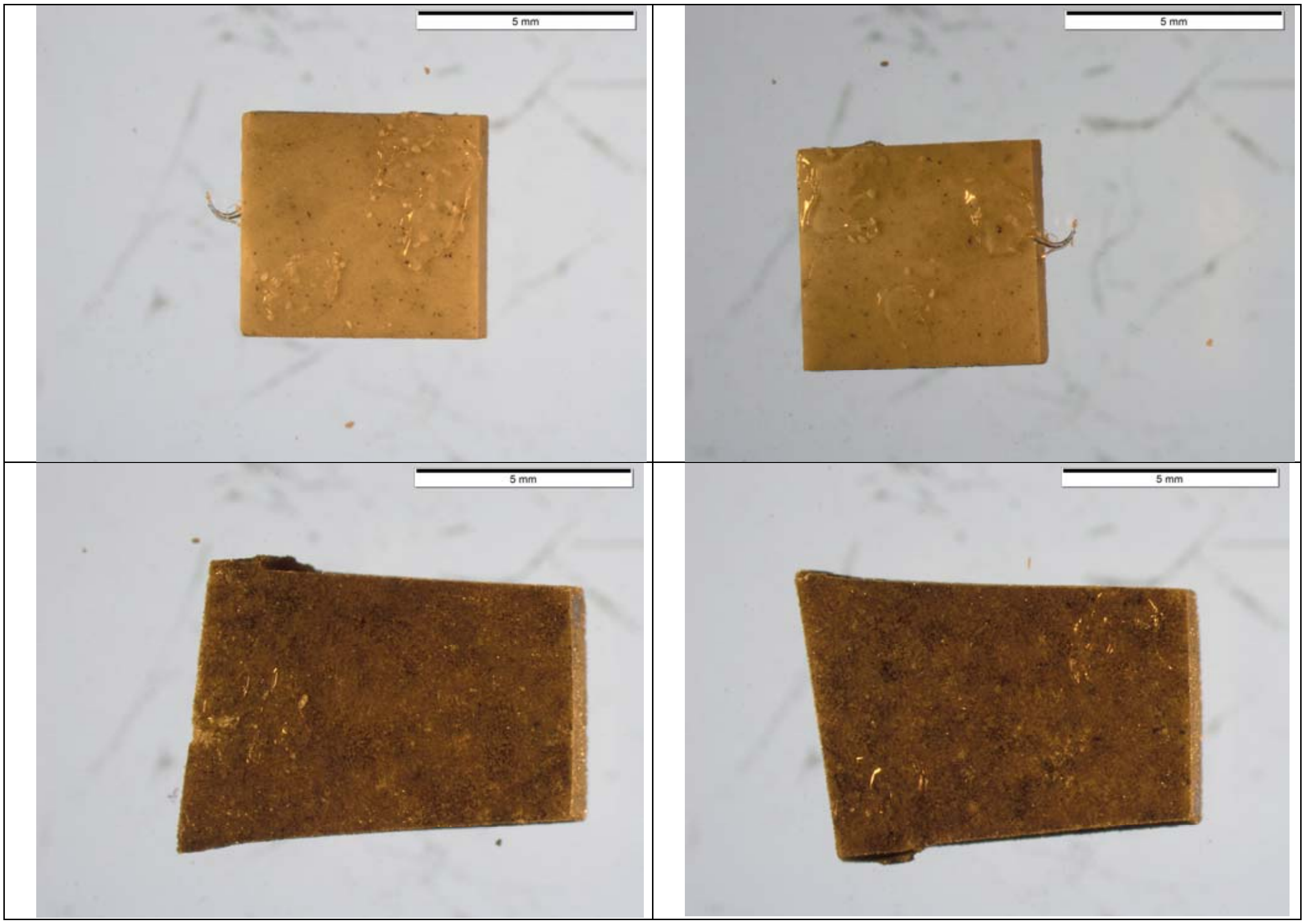

Figure C-5. Images of 1456 Day Leached Monolith Samples O15 (top) and O16 (bottom)