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Product Consistency Test Leachate Data for Nepheline Scoping Study Glasses

K. M. Fox

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EXECUTIVE SUMMARY

In this report, the Savannah River National Laboratory provides chemical analysis for a series of leachates resulting from Product Consistency Tests of nepheline study glasses performed at the Pacific Northwest National Laboratory. A review of the standard solution data identified no issues with the analytical methods. The results of this scoping study will be used as part of an effort to better understand nepheline crystallization in high-level nuclear waste glasses with high concentrations of aluminum and sodium.

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LIST OF ABBREVIATIONS

DOE	U.S. Department of Energy
ICP-AES	Inductively Coupled Plasma – Atomic Emission Spectroscopy
HLW	High Level Waste
LAW	Low Activity Waste
ORP	Office of River Protection
PCT	Product Consistency Test
PNNL	Pacific Northwest National Laboratory
SRNL	Savannah River National Laboratory
TTQAP	Task Technical and Quality Assurance Plan
WTP	Hanford Tank Waste Treatment and Immobilization Plant
%RSD	Percent Relative Standard Deviation

1.0 Introduction

The U.S. Department of Energy (DOE) Office of River Protection (ORP) has requested that the Savannah River National Laboratory (SRNL) provide expert evaluation and experimental work in support of the River Protection Project vitrification technology development. DOE is building the Hanford Tank Waste Treatment and Immobilization Plant (WTP) at the Hanford Site in Washington to remediate 55 million gallons of radioactive waste that is temporarily stored in 177 underground tanks. The low-activity waste (LAW) fraction will be partitioned from the high-level waste (HLW). Both the LAW and HLW will then be vitrified into borosilicate glass using Joule-heated ceramic melters.

Efforts are being made to increase the loading of Hanford tank wastes in the glass while conforming to processing requirements and product quality regulations. DOE-ORP has requested that SRNL support the advancement of glass formulations and process control strategies in key technical areas, as defined in the Task Technical and Quality Assurance Plan (TTQAP).[1] Two of these areas are enhancing waste glass property/composition models and broadening the compositional regions over which those models are applicable.

In this report, SRNL provides analysis of Product Consistency Test (PCT) leachates resulting from a series of glasses fabricated at the Pacific Northwest National Laboratory (PNNL) to evaluate the effects of nepheline crystallization on chemical durability over varying time and temperature conditions. The results of this scoping study will be used as part of an effort to better understand nepheline crystallization in HLW glasses with high concentrations of aluminum and sodium. The data will be used in the development of improved property/composition models for HLW glass.

2.0 Experimental Procedure

2.1 Quality Assurance

Requirements for performing reviews of technical reports and the extent of review are established in Savannah River Site Manual E7, Procedure 2.60. SRNL documents the extent and type of review using the SRNL Technical Report Design Checklist contained in WSRC-IM-2002-00011, Rev. 2. Laboratory data for this study were recorded in the SRNL Electronic Laboratory Notebook system, experiment C3489-00079-28. The glasses provided by PNNL were fabricated following Test Instruction EWG-TI-0068.

2.2 Samples and Analysis

A series of leachates resulting from PCTs[2] performed at PNNL was received at SRNL for analysis. The identifier assigned by PNNL to each leachate and the corresponding SRNL analysis identifiers are given in Table 2-1. Each leachate was submitted to the SRNL Process Science Analytical Laboratory for analysis via Inductively Coupled Plasma – Atomic Emission Spectroscopy (ICP-AES). The samples were divided between two measurement blocks. The sample measurement run order is given in Table 2-2. Samples of a multi-element, standard solution[a] were included in the analytical plan as a check on the accuracy of the ICP-AES instrument used for these measurements. The standard solution samples are denoted in Table 2-2 by "std-i-j" where i=1 and 2 represents the block number and j=1, 2, and 3 represents the position in the block. The concentrations of Al, B, Li, Na, and Si in each leachate sample were measured.

[a] ICP multi-element custom solution, product number SM-744-013, High Purity Standards, Charleston, SC.

Table 2-1. Sample Identifiers for PCT Leachates

PNNL Identifier	SRNL Identifier
NP6-06-CCC-6_DAYS-90C-acidified analyte	S-8864
NP6-06-CCC-6_DAYS-40C-acidified analyte	S-8865
NP6-06-CCC-6_DAYS-RmT-acidified analyte	S-8866
Blank-7_days-90C-acidified analyte	S-8867
Standard-7_days-90C-acidified analyte	S-8868
Standard-7_days-40C-acidified analyte	S-8869
Standard-7_days-RmT-acidified analyte	S-8870
NP6-16-CCC-6_days-90C-acidified analyte	S-8871
NP6-16-CCC-6_days-40C-acidified analyte	S-8872
NP6-16-CCC-6_days-RmT-acidified analyte	S-8873
NP6-06-Q-6_days-90C-acidified analyte	S-8874
NP6-06-Q-4_days-90C-acidified analyte	S-8875
NP6-06-Q-2_days-90C-acidified analyte	S-8876
NP6-06-Q-6_days-40C-acidified analyte	S-8877
NP6-06-Q-4_days-40C-acidified analyte	S-8878
NP6-06-Q-2_days-40C-acidified analyte	S-8879
NP6-06-Q-6_days-RmT-acidified analyte	S-8880
NP6-06-Q-4_days-RmT-acidified analyte	S-8881
NP6-06-Q-2_days-RmT-acidified analyte	S-8882
NP6-16-Q-6_days-90C-acidified analyte	S-8883
NP6-16-Q-4_days-90C-acidified analyte	S-8884
NP6-16-Q-2_days-90C-acidified analyte	S-8885
NP6-16-Q-6_days-40C-acidified analyte	S-8886
NP6-16-Q-4_days-40C-acidified analyte	S-8887
NP6-16-Q-2_days-40C-acidified analyte	S-8888
NP6-16-Q-6_days-RmT-acidified analyte	S-8889
NP6-16-Q-4_days-RmT-acidified analyte	S-8890
NP6-16-Q-2_days-RmT-acidified analyte	S-8891

Table 2-2. Sample Measurement Blocks and Run Order

Block 1	Block 2
std-1-1	std-2-1
S-8864	S-8878
S-8865	S-8879
S-8866	S-8880
S-8867	S-8881
S-8868	S-8882
S-8869	S-8883
S-8870	S-8884
std-1-2	std-2-2
S-8871	S-8885
S-8872	S-8886
S-8873	S-8887
S-8874	S-8888
S-8875	S-8889
S-8876	S-8890
S-8877	S-8891
std-1-3	std-2-3

3.0 Results

Table 3-1 provides the concentration measurements for the leachate samples and standard solution samples. Measurements that were below the analytical detection limits are reported with a "<" symbol.

Table 3-2 provides a review of the measurements of the solution standard samples that were included in the analytical blocks. For each analytical block, the mean, standard deviation, and percent relative standard deviation (%RSD) are determined for each element present in the standard. Following the guidance in ASTM C1285, there were two primary evaluations conducted for these summary statistics: the mean value for each analytical block was found to be less than 10% from the reference value (i.e., a percent relative bias less than 10%) for the element in question, and the %RSD was less than 10% for the element in question. The results in Table 3-2 satisfy these criteria, and thus, the results for the solution standard suggest no significant issues with the analytical outcomes for the measurements of the leachate samples.

Table 3-1. Measured Values (mg/L) for the Leachate and Standard Solution Samples

Block	SRNL ID	PNNL ID	Al	B	Li	Na	Si
1	std-1-1	std-1-1	3.94	20.2	10.2	81.0	49.7
	S-8864	NP6-06-CCC-6_DAYS-90C-acidified analyte	1.74	126	32.2	108	<1.00
	S-8865	NP6-06-CCC-6_DAYS-40C-acidified analyte	1.84	65.7	16.9	61.4	<1.00
	S-8866	NP6-06-CCC-6_DAYS-RmT-acidified analyte	<1.00	4.39	1.35	4.44	<1.00
	S-8867	Blank-7_days-90C-acidified analyte	<1.00	<1.00	<1.00	<1.00	<1.00
	S-8868	Standard-7_days-90C-acidified analyte	<1.00	59.4	20.1	160	85.8
	S-8869	Standard-7_days-40C-acidified analyte	<1.00	2.05	<1.00	6.99	1.18
	S-8870	Standard-7_days-RmT-acidified analyte	<1.00	<1.00	<1.00	1.81	<1.00
	std-1-2	std-1-2	3.74	18.9	9.98	78.9	46.7
	S-8871	NP6-16-CCC-6_days-90C-acidified analyte	<1.00	28.4	10.8	16.8	<1.00
	S-8872	NP6-16-CCC-6_days-40C-acidified analyte	2.19	7.84	3.35	6.25	<1.00
	S-8873	NP6-16-CCC-6_days-RmT-acidified analyte	<1.00	3.15	1.30	2.88	<1.00
	S-8874	NP6-06-Q-6_days-90C-acidified analyte	3.30	121	30.1	108	<1.00
	S-8875	NP6-06-Q-4_days-90C-acidified analyte	1.49	61.7	16.1	57.4	<1.00
	S-8876	NP6-06-Q-2_days-90C-acidified analyte	<1.00	17.2	4.76	15.9	<1.00
	S-8877	NP6-06-Q-6_days-40C-acidified analyte	<1.00	2.58	<1.00	2.89	<1.00
	std-1-3	std-1-3	3.61	21.6	9.55	75.6	48.8
2	std-2-1	std-2-1	3.98	20.7	10.1	83.3	50.5
	S-8878	NP6-06-Q-4_days-40C-acidified analyte	1.32	3.18	<1.00	3.14	<1.00
	S-8879	NP6-06-Q-2_days-40C-acidified analyte	1.54	2.55	<1.00	2.66	<1.00
	S-8880	NP6-06-Q-6_days-RmT-acidified analyte	<1.00	1.02	<1.00	1.07	<1.00
	S-8881	NP6-06-Q-4_days-RmT-acidified analyte	<1.00	<1.00	<1.00	<1.00	<1.00
	S-8882	NP6-06-Q-2_days-RmT-acidified analyte	<1.00	<1.00	<1.00	<1.00	<1.00
	S-8883	NP6-16-Q-6_days-90C-acidified analyte	<1.00	22.4	8.27	15.1	<1.00
	S-8884	NP6-16-Q-4_days-90C-acidified analyte	12.3	14.7	6.07	10.5	3.82
	std-2-2	std-2-2	3.82	19.4	10.0	80.9	48.8
	S-8885	NP6-16-Q-2_days-90C-acidified analyte	10.7	10.9	4.59	8.78	3.93
	S-8886	NP6-16-Q-6_days-40C-acidified analyte	<1.00	2.66	<1.00	2.67	<1.00
	S-8887	NP6-16-Q-4_days-40C-acidified analyte	1.08	2.61	<1.00	2.61	<1.00
	S-8888	NP6-16-Q-2_days-40C-acidified analyte	1.46	2.82	<1.00	2.27	<1.00
	S-8889	NP6-16-Q-6_days-RmT-acidified analyte	<1.00	1.13	<1.00	<1.00	<1.00
	S-8890	NP6-16-Q-4_days-RmT-acidified analyte	<1.00	<1.00	<1.00	<1.00	<1.00
	S-8891	NP6-16-Q-2_days-RmT-acidified analyte	<1.00	1.29	<1.00	1.26	<1.00
	std-2-3	std-2-3	3.79	19.4	9.84	79.4	48.3

Table 3-2. Results from Samples of the Multi-Element Solution Standard

Block	1	2	Reference Values (mg/L)
Mean (Al (mg/L))	3.76	3.86	4
Mean (B (mg/L))	20.24	19.86	20
Mean (Li (mg/L))	9.90	10.00	10
Mean (Na (mg/L))	78.48	81.21	81
Mean (Si (mg/L))	48.38	49.24	50
% relative bias (Al)	-5.9%	-3.4%	<10% per ASTM C1285
% relative bias (B)	1.2%	-0.7%	
% relative bias (Li)	-1.0%	0.0%	
% relative bias (Na)	-3.1%	0.3%	
% relative bias (Si)	-3.2%	-1.5%	
Std Dev (Al (mg/L))	0.170	0.099	
Std Dev (B (mg/L))	1.374	0.747	
Std Dev (Li (mg/L))	0.320	0.154	
Std Dev (Na (mg/L))	2.727	1.974	
Std Dev (Si (mg/L))	1.530	1.158	
%RSD (Al)	4.5%	2.6%	<10% per ASTM C1285
%RSD (B)	6.8%	3.8%	
%RSD (Li)	3.2%	1.5%	
%RSD (Na)	3.5%	2.4%	
%RSD (Si)	3.2%	2.4%	

4.0 Summary

In this report, SRNL provides chemical analysis for a series of leachates resulting from PCTs of nepheline study glasses performed at PNNL. A review of the standard solution data identified no issues with the analytical methods.

5.0 References

1. Fox, K. M., "Task Technical and Quality Assurance Plan for Hanford Waste Glass Development and Characterization," *U.S. Department of Energy Report SRNL-RP-2013-00692, Revision 1,* Savannah River National Laboratory, Aiken, SC (2016).

2. ASTM, "Standard Test Methods for Determining Chemical Durability of Nuclear Waste Glasses: The Product Consistency Test (PCT)," *ASTM C1285,* (2014).

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