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HANFORD LOW-LEVEL WASTE FORM PERFORMANCE FOR MEETING LAND DISPOSAL REQUIREMENTS

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ABSTRACT

Immobilized Low-activity waste (ILAW) from the Hanford site will be disposed of in near-surface burial grounds and must be processed into a chemically durable waste form to prevent release of hazardous constituents to the environment. To meet this goal, the LAW will be immobilized in borosilicate glass. The DOE Office of River Protection and the River Protection Project-Waste Treatment Plant (RPP-WTP) project have agreed on testing requirements that the immobilized LAW glass must meet to demonstrate chemical durability. Two of the tests are the Product Consistency Test (PCT)¹ and Environmental Protection Agency's (EPA) Toxicity Characteristic Leaching Procedure (TCLP).² This paper provides results of RPP-WTP PCT and TCLP testing on both actual radioactive and non-radioactive simulant LAW glasses to show they meet the associated land disposal requirements.

INTRODUCTION

The LAW glass produced from the planned RPP-WTP is to be stored in a near surface vault repository on the Hanford site. Detailed descriptions of existing disposal vaults as well as conceptual designs for new ILAW disposal

facilities at the Hanford site have been previously presented.³ Borosilicate glass is the selected Tri-party agreement waste form for immobilization of Hanford LAW. The borosilicate glass waste form has proven stability and durability. Per the Department of Energy (DOE) Office of River Protection and Bechtel National, Inc. contract,⁴ the ILAW glass normalized mass loss of sodium, silicon, and boron shall be less than 2.0 grams/m² using a seven-day ASTM- C1285-97 procedure¹ 'Product Consistency Test' run at 90 °C. The ILAW product shall also be acceptable for land disposal under Resource Conservation and Recovery Act (RCRA) Land Disposal Restrictions (LDR) 40CFR268,⁵ and thus must show compliance via the EPA's TCLP.

BACKGROUND AND OBJECTIVES

Four Hanford tank supernatant samples were processed through representative unit operations for High-Level Waste (HLW) pretreatment including entrained solids removal, strontium/transuranic (Sr/TRU) precipitation and filtration, and cesium and technetium removal via ion exchange. These processes remove most of the waste radioactivity. The resulting decontaminated supernatant samples were converted into LAW glass by consideration of their respective analytical characterizations. The LAW pretreated waste supernatants were blended with certain mineral glass formers to produce crucible-scale melter feeds. Vitreous State Laboratory (VSL) personnel of Catholic University of America (CUA) provided the target glass compositions and also fabricated simulated LAW glasses that were similar, but not exactly the same composition as the radioactive waste glasses. Two different Hanford tank wastes of the Envelope C group (Tanks 241-AN-107 & AN-102) and two different tank wastes of the Envelope A group (Tanks 241-AW-101 and AN-103) were used in this study. The overall objective of this work was to show compliance with the RPP-WTP contractual durability requirements via the PCT and TCLP testing of crucible melt glass produced from actual radioactive Hanford tank samples.

LAW GLASS FABRICATION AND CHARACTERIZATION

The radioactive LAW glasses produced for this study were made from melter feed slurries that were dried, calcined and melted in platinum crucibles at 1150 °C. The mineral glass formers used consisted of kyanite (Al₂SiO₃), orthoboric acid (H₃BO₃), wollastonite (CaSiO₃), red iron oxide pigment (Fe₂O₃), olivine (Mg₂SiO₄), silica sand (SiO₂), rutile ore (TiO₂), zinc oxide (ZnO), zircon sand (ZrSiO₄), lithium carbonate (LiCO₃). Sugar (C₁₂H₂₂O₁₁) was added to control glass oxidation state. Glass formulations considered sodium as the main waste component loading indicator with a minimum of 10 wt% Na₂O for the Env. C glasses and a minimum of 14 wt% Na₂O for the Env. A glasses. Further details describing the pretreatment of waste streams, fabrication of glasses and associated analytical characterization of the product glasses can be found in technical references.⁶

PRODUCT CONSISTENCY TEST

The glass durability test known as the PCT is an ASTM procedure (ASTM C1285-97) that tests the durability of crushed glasses over a seven-day test at 90 °C. The test uses glass particles sized between ~ 74 and 149 microns, or 100 to 200 mesh size. The sample size is typically > 1 g, with leachate volume to sample mass equal to 10X. Tests are performed in unsensitized Type 304L stainless steel. The initial and final pH values of the leachate solutions are measured and the filtered, acidified final product leachates are characterized for soluble components leached from the crushed glass. Results are reported as normalized elemental mass releases. Normalization considers the measured concentration of the element in the leachate and the concentration (wt%) of the same element in the glass. The PCT results from durability tests with glasses used in this study are presented in Table I. Table I shows normalized release data for the radioactive glasses and their respective surrogate glasses. Data is also shown for a nonradioactive reference glass, the Low-Activity Reference Material (LRM) glass.⁷ The data in Table I clearly show that normalized mass release for all analytes are well below the contract specified maximum of 2 g/m² for the seven-day PCT at 90 °C. Comparison of the surrogate glass leach data to their respective radioactive glasses also indicates good agreement even though the surrogate and radioactive compositions were not exactly the same.

TOXICITY CHARACTERISTIC LEACHING PROCEDURE (TCLP)

The TCLP is specified by EPA SW-846 Method 1311 and uses glass particles that are capable of passing through a 9.5-mm (0.375-in.) standard sieve. The glass sample is placed into an extractor vessel with an extraction fluid mass equal to 20 times the mass of the glass sample. The extraction fluid used for these tests was TCLP extraction fluid #1 consisting of 5.7 mL glacial acetic acid, 64.3 mL of 1N NaOH in one liter of ASTM water with resulting pH of 4.93. The extractor vessel containing the sample is rotated end over end at 30 rpm for 18 hours at room temperature of 22 °C. The resulting liquid is then separated from the glass particles by filtration and the leachate is analyzed for the analytes of concern. Tables II and III present the TCLP test results for the four different radioactive glasses tested in this work. The four radioactive glasses were produced from pretreated alkaline supernatants that are generally very low in concentrations of RCRA metals. The data shown in these tables demonstrate that actual waste glasses easily meet Universal Treatment Standards (UTS).⁵ Similar data has been collected in tests with simulated ILAW glasses spiked with elevated concentrations of RCRA metals.⁸

Table I. PCT Results –Average 7-day PCT Normalized Mass Loss (g/m²)

Glass	B	Na	Si
LRM	0.51	0.50	0.17
LAWA41 Simulate	0.37	0.36	0.16
AN-103 Radioactive	0.37	0.40	0.17
LAWC21 Simulate	0.37	0.35	0.16
AN-102 Radioactive	0.30	0.35	0.12
LAWA88 Simulate	0.43	0.43	0.17
AW-101 Radioactive	0.57	0.59	0.20
LAWC15 Simulate	0.33	0.34	0.16
AN-107 Radioactive	0.35	0.42	0.19

Table II. TCLP Test Results for Tanks AW-101 and AN-107

	UTS (40CFR268)	AW-101		AN-107	
	TCLP Levels ug/mL	Leachate ug/mL	Glass ug/g	Leachate ug/mL	Glass ug/g
Analyte					
Ag*	0.14	0.0009	<0.0003	0.0011	<0.0003
As*	5.0	<0.006	<0.006	0.008	<0.006
Ba*	21	0.04	<0.03	0.04	240
Be	1.22	<0.03	<0.03	<0.03	<0.03
Cd*	0.11	<0.0003	<0.0003	0.013	40
Cr*	0.60	0.16	245	0.29	235
Ni	11	0.24	<0.08	0.33	430
Pb*	0.75	0.026	<0.003	0.028	38
Sb	1.15	<0.0007	<0.0007	<0.0007	<0.0007
Se #	1.0	<0.013	<0.013	<0.013	<0.013
Tl	0.20	<0.01	<0.01	<0.01	<0.01
Hg*	0.025	<0.001	<0.001	<0.001	<0.001

* Indicates vitrification is the LDR treatment standard for this metal – UTS shown for information only.

Se has a UTS (5.7 mg/L TCLP) that is greater than the toxicity characteristic, therefore the Se toxicity characteristic level of Washington State Acceptance Criteria (WAC) 173-303-090 is shown.

Table III. TCLP Test Results for Tanks AN-103 and AN-102

	UTS	AN-103	AN-102
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	(40CFR268)				
	TCLP Levels	Leachate	Glass	Leachate	Glass
Analyte	ug/mL	ug/mL	ug/g	ug/mL	ug/g
Ag*	0.14	<0.041	100	<0.0046	<800
As*	5.0	<0.005	<2	<0.02	<350
Ba*	21	2.9	37	2.46	65
Be	1.22	<0.002	<0.9	<0.0009	<0.45
Cd*	0.11	<0.012	26	<0.0013	<2.4
Cr*	0.60	<0.025	230	0.0084	300
Ni	11	<0.064	180	0.0094	140
Pb*	0.75	<0.3	150	<0.033	<85
Sb	1.15	<0.2	<77	<0.021	<38
Se #	1.0	0.015	<5	<0.016	<30
Tl	0.20	<0.06	<200	<0.067	<120
Hg*	0.025	<0.0001	0.44	<0.0005	<0.1

* Indicates vitrification is the LDR treatment standard for this metal – UTS shown for information only.

Se has a UTS (5.7 mg/L TCLP) that is greater than the toxicity characteristic, therefore the Se toxicity characteristic level of Washington State Acceptance Criteria (WAC) 173-303-090 is shown.

CONCLUSIONS

Data presented in this work address the overall objective aimed at showing compliance with the RPP-WTP contractual durability requirements via the PCT and TCLP testing of crucible melt glasses produced from actual radioactive Hanford tank samples. Comparison of normalized release rates from PCT's on both actual and surrogate LAW glasses to the contract specified maximum of 2 g/m² indicates that all of these glasses readily meet this durability standard. The ILAW glasses also effectively immobilize RCRA metals in actual waste samples.

REFERENCES

- ¹ ASTM C1285-97 (1998). *Standard Test Methods for Determining Chemical Durability of Nuclear Waste Glasses: The Product Consistency Test (PCT)*. American Society for Testing and Materials, Easton, Maryland.
- ² SW-846, *Test Methods for Evaluating Solid Waste, Physical/Chemical Methods*. U.S. Environmental Protection Agency, Office of Solid Waste and Emergency Response, Washington, D.C.
- ³ R. Puigh, "Appendix I, Disposal Facility Data for the Hanford Immobilized Low-Activity Tank Waste Performance Assessment," HNF-5636,

Rev. 0, HNF-4950, Rev. 1, Fluor Federal Services, Richland, Washington, (December 1999).

⁴ River Protection Project – Waste Treatment Plant (RPP-WTP) Contract, Bechtel National, Inc.-- Design, Construction, and Commissioning of the Hanford Tank Waste Treatment and Immobilization Plant: Contract No. DE-AC27-01RV14136, <http://www.hanford.gov/orp/>, (October 2002).

⁵ 40CFR268. *Land Disposal Restrictions*. Code of Federal Regulations. U.S. Environmental Protection Agency, Washington, D.C.

⁶ G. L. Smith et al., “Vitrification and Product Testing of AW-101 and AN-107 Pretreated Wastes,” PNNL-13372, WTP-RPT-003, Rev. 0 (October 2000); C. L. Crawford et al., “Crucible-Scale Active Vitrification Testing Envelope A, Tank 241-AN-103,” WSRC-TR-2000-00322, SRT-RPP-2000-00021, Rev. 1, (June 2001); C. L. Crawford et al., “Crucible-Scale Active Vitrification Testing Envelope C, Tank 241-AN-102,” WSRC-TR-2000-00371, SRT-RPP-2000-00022, Rev. 0 (June 2001).

⁷ W. L. Ebert and S. L. Wolf, “Dissolution Test for Low-Activity Waste Product Acceptance,” Argonne National Laboratory, Proceedings of Spectrum '98, Denver, CO, Sept 13-18, 1998, pp. 724-731.

⁸ I. S. Muller, A. C. Buechele and I. L. Pegg, “Glass Formulation and Testing with RPP-WTP LAW Simulants,” VSL-01R3560-2, Rev. 0, (February 23, 2001).