ANL-00/12

Development of a Method for Monitoring the Consistency of Glass-Bonded Sodalite Waste Forms

by M. A. Lewis, M. L. Stanley, and W. L. Ebert

Argonne National Laboratory, Argonne, Illinois 60439
operated by The University of Chicago
for the United States Department of Energy under Contract W-31-109-Eng-38

Argonne National Laboratory, with facilities in the states of Illinois and Idaho, is owned by the United States Government and operated by The University of Chicago under the provisions of a contract with the Department of Energy.

Available electronically at http://www.doe.gov/bridge

Available for a processing fee to U.S. Department of Energy and its contractors, in paper, from:

U.S. Department of Energy
Office of Scientific and Technical Information
P.O. Box 62
Oak Ridge, TN 37831-0062
phone: (865) 576-8401
fax: (865) 576-5728
email: reports@adonis.osti.gov

ANL-00/12

ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois 60439-4837

DEVELOPMENT OF A METHOD FOR MONITORING THE CONSISTENCY OF GLASS-BONDED SODALITE WASTE FORMS

by

M. A. Lewis, M. L. Stanley, and W. L. Ebert

Chemical Technology Division

May 2000

TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

Development of a Method for Monitoring the Consistency of Glass-Bonded Sodalite Waste Forms

W. L. Ebert, M. L. Lewis, and M. L. Stanley

ABSTRACT

The results of tests conducted to support development of a product consistency test for a ceramic waste form of glass-bonded sodalite are presented and discussed in this report. The focus of the study is to distinguish between the dissolution of different phases present in the waste form, primarily sodalite, glass binder, and halite by using simple test methods that can be conducted remotely in a hot cell with highly radioactive material. Information from the tests is needed to support process control and qualification of the waste form for disposal in a federal high-level waste repository. It is recommended that the same product consistency test used for vitrified high-level waste forms, namely, Product Consistency Test Method A, be used for the ceramic waste form. Analysis of the water wash solution generated during sample preparation should be included.

I. INTRODUCTION

A glass-bonded sodalite waste form is being developed to immobilize waste salt generated during the conditioning of spent sodium-bonded nuclear fuel from the Experimental Breeder Reactor (EBR-II) at Argonne National Laboratory-West. Because fission products and transuranic elements accumulate in the salt during the electrometallurigical treatment process, the waste salt must be disposed of as high-level radioactive waste. A testing program is in progress to support qualification of the glass-bonded sodalite waste form for disposal in a federal high-level waste facility. The requirements for qualification of a high-level radioactive waste form for disposal are summarized in the Waste Acceptance Specification Requirements Document (WASRD) [DOE-1996]. One of the requirements of the WASRD is identification and validation of a method for monitoring the chemical and physical consistency of product waste forms in order to demonstrate control of waste form production. The method must be sufficiently sensitive to indicate when processing limits have been exceeded and should also be sensitive to the chemical durability of the waste form. In this report, we discuss the results of scoping tests conducted with reference glass-bonded sodalite materials and describe how the results are being used to develop a product consistency test method for that waste form.

Our approach in the development of a consistency test is based on gaining an understanding of the chemical and physical attributes of the waste form and its corrosion behavior. This ensures that the waste form attribute that is monitored in the consistency test is sensitive to both possible processing upsets and the performance of the waste form under disposal conditions. It is instructive to first consider the approach used for monitoring the consistency of glass waste forms of defense high-level waste (DHLW); borosilicate glass is the reference waste form described in the WASRD. For DHLW glass, the consistency of its composition is used as evidence that the vitrification process was adequately controlled. The consistency is monitored with the Product Consistency Test Method A (PCT-A), which has been standardized by the American Society for Testing and Materials (ASTM-1998A). The PCT-A is a partial dissolution test wherein the amounts of boron, silicon, and alkali metals released over a seven-day period are measured. This method is adequate because the DHLW glass is essentially a homogeneous material, and the release of radionuclides is presumed to be bounded by the dissolution rate of the glass. Therefore, process control can be adequately demonstrated by the consistency of the glass composition, as measured by the release of soluble components over a short test duration.

The compositions of waste sludge in the different tanks at the Savannah River Site (SRS) are sufficiently diverse that the compositions of the glasses produced in different campaigns at the Defense Waste Processing Facility (DWPF) will be different. A range of acceptable compositions has thus been defined for DWPF waste based on various processing constraints. The consistency requirement for these glasses is that their response in the PCT must be within two standard deviations of the response of the benchmark reference glass identified in the Environmental Assessment (EA) for the DWPF [DOE-1990] after being normalized to the glass composition. The DWPF approach provides a measure of whether the waste forms dissolve more slowly than the EA glass under the PCT-A conditions.

Monitoring the consistency of the ceramic waste form is more complicated than it is for DHLW glass because the ceramic waste form is heterogeneous and radionuclides are present in several phases. Per the WASRD requirements, both the chemical composition and relative abundance of all phases present in excess of 10 vol% must be monitored to demonstrate that waste form production was adequately controlled. The development of a suitable consistency test method requires an understanding of the dissolution behavior of the major phases present in the waste form. This can be done by utilizing the known compositions of the three major phases present in the ceramic waste form: sodalite, glass binder, and halite (although halite comprises less than 10% of the waste form volume, the amount of halite present in the waste form is important for monitoring waste form production). Most of the major components of the waste form are present in more than one phase: silicon and aluminum are present in both the glass and the sodalite; sodium is present in the glass, sodalite, and halite; and chlorine is present in the sodalite and halite. Lithium, one of the components of the waste salt, is not present initially in the glass binder, but some lithium may diffuse into the glass during processing. Boron is present only in the glass binder, and so the release of boron provides a unique measure of the glass dissolution. Because halite is much more soluble than sodalite or glass binder, it can be distinguished in the water-wash solution generated during sample preparation. A marker for the dissolution of the sodalite phase has not yet been determined.

The method that we recommend to monitor the consistency of ceramic waste forms is a slightly modified version of PCT-A. In PCT-A used for vitrified waste forms, the material to be tested is crushed and sieved to isolate the fraction that passes through a 100 mesh sieve but is retained by a 200 mesh sieve (i.e., the −100 +200 mesh fraction). The crushed and sieved material is washed several times with demineralized water and ethanol to remove the fines. Because the ceramic waste form contains soluble components, the washing procedure is less aggressive. The −100 +200 mesh fraction of the ceramic waste form is washed once with ethanol to remove fine material and once with demineralized water. The recovered water-wash solution is analyzed to quantify the amounts of salt components (Na^+, Cs^+, Cl^-, and I^-) that dissolved. After drying, the recovered material is reacted with demineralized water at 90°C for seven days. The test solution is analyzed for pH, boron, alkali metals, and silicon. The procedure advises the user to either analyze the wash solutions if phases that may dissolve during the wash procedure are present or omit the wash steps. Our scoping tests have shown that salt crystals that were not incorporated into the sodalite structure and which became exposed at the surface due to crushing or cutting the ceramic waste form dissolve readily when exposed to water at room temperature. Since these salts contain small amounts of Cs and I, the amount of exposed salt should be tracked in the product consistency procedure for the ceramic waste form. This is readily accomplished by analyzing the wash solutions during sample preparation. The dissolution of the sodalite and glass binder phases can then be monitored with the measured concentrations of boron, alkali metals, chloride, and silicon in the test solution.

II. DESCRIPTION OF CERAMIC WASTE FORM

The ceramic waste form is produced by first mixing and heating contaminated salt with dried zeolite 4A to occlude most of the salt within the zeolite cages. The zeolite used to occlude the salt is in the form of <10-μm-sized grains of polycrystalline zeolite 4A that have been aggregated with a clay binder into particles with a size range of 74-230 μm (the –60 +200 mesh fraction). The aggregated material is referred to as "granular zeolite." The granular zeolite is dried to a residual moisture level of about 0.1 mass % water. When contacted by the residual moisture in the granular zeolite, rare earth and actinide elements that are present in the waste salt as chlorides form oxide phases. These are distributed primarily on the outer surface of the granular zeolite particles, although a small fraction is present between zeolite grains within the granular zeolite aggregates. The salt-loaded granular zeolite is then mixed with glass frit at a ratio of about 75 mass % salt-loaded granular zeolite and 25 mass % glass. The mixture is placed in a stainless steel can, compacted, then hot isostatically pressed (HIPed) to densify the mixture. During HIPing, the salt-loaded zeolite 4A transforms *in situ* to the mineral sodalite, which becomes fixed in the glass binder. The glass becomes sufficiently soft during HIPing that it flows to form an intimate interface with the sodalite. Small crystals of mixed rare earth and actinide oxides form inclusions within the glass binder and, to a lesser extent, within the sodalite domains. Halite crystals are also formed during processing. These are generally observed as small inclusions within the glass binder.

Most of our scoping tests were conducted with materials made with nonradioactive isotopes of fission products expected to be in the waste salts generated during fuel conditioning. These are referred to as reference ceramic waste forms (reference CWFs). Tests with reference CWFs that had been doped with natural uranium and plutonium-239 indicate that the addition of these actinides did not measurably affect the microstructure or dissolution behavior of the waste form. Examination of the microstructure of the CWF shows the sodalite and glass phases to be fairly evenly distributed with typical domain sizes of about one millimeter. The photomicrograph in Fig. 1 shows the microstructure of a reference CWF made with uranium. Crystallites of various rare earth element and uranium oxides and oxychlorides are seen to have formed inclusions within the glass and sodalite phases. Most of the crystallites are near the sodalite/glass phase boundaries or the boundaries between adjacent sodalite grains. Several small pores are observed within the glass phase near phase boundaries. Salt crystals (e.g., halite) have been observed in some of these pores. It is likely that salt was lost from many pores when they became exposed to cutting fluid during preparation of the polished cross-section samples.

The transformation of zeolite 4A to sodalite when the waste form is HIPed changes the cage structure. The cage structure of sodalite is slightly different from that of zeolite A. Zeolite 4A contains alpha cages, which have an aperture of 0.4 nm, and beta cages, which have an aperture of 0.22 nm [BRECK-1974]. Sodalite contains only beta cages. Although the mechanism by which zeolite 4A transforms to sodalite is not known, the transformation slightly densifies the structure, which may result in the expulsion of material from the alpha cage as it transforms to a beta cage. Tests were conducted to measure the leachability of salts that (1) are occluded in the cage structure of the zeolite during the salt loading process and (2) remain occluded in the sodalite cages after transformation from zeolite. Tests were conducted following a modified ANS 16.1 leach test procedure [ANS-1992] wherein finely divided samples were

leached in demineralized water for various durations [SIMPSON-1998]. Test results, plotted in Fig. 2, are normalized to the measured concentration of silicon. The release of Li, Na, and Cl from salt-loaded zeolite is greater than the release of Si at all test intervals, but the release of these elements from salt-loaded sodalite is less than the release of Si at all but the first two intervals. Salts that are not occluded by the sodalite dissolve rapidly when contacted by water (within the first two exchanges during the first day). Salt that is occluded in the sodalite cages can be released to solution only after the cages themselves dissolve.

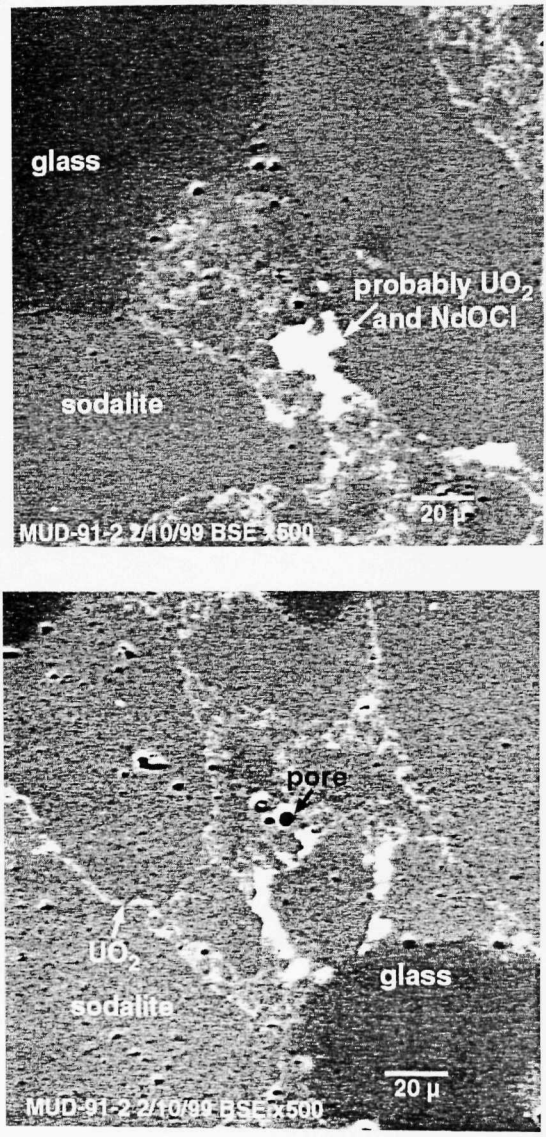
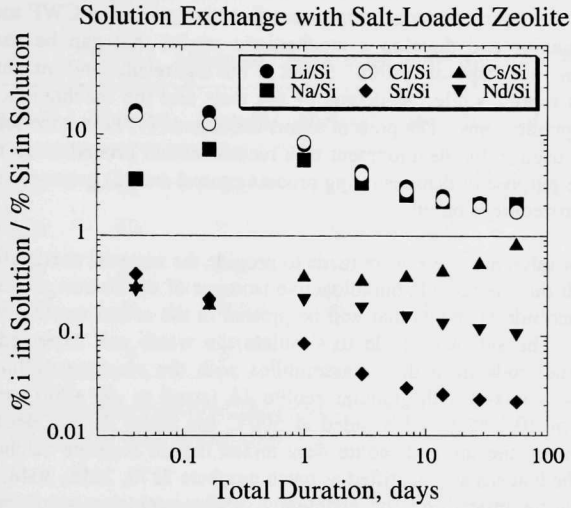


Fig. 1. Photomicrographs of U-Doped Reference Ceramic Waste Form



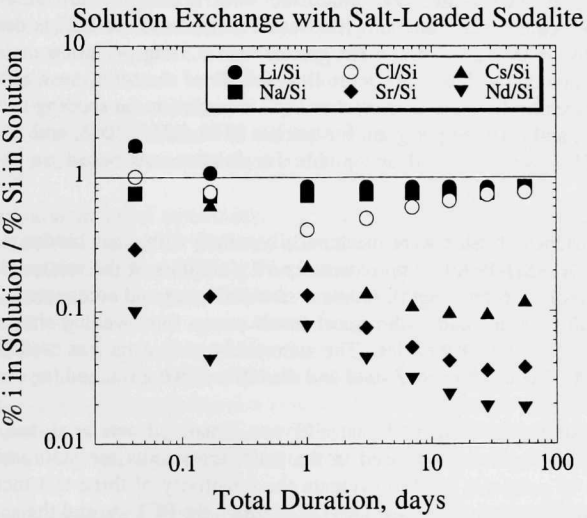


Fig. 2. Release of Components (i) Normalized to Release of Silicon in Solution Replacement Tests (a) with Salt-Loaded Zeolite and (b) with Salt-Loaded Sodalite

III. SAMPLE PREPARATION

Many tests have been conducted with nonradioactive reference CWF materials to study their corrosion behavior and develop a mechanistic model that can be used to calculate radionuclide releases in a disposal system. Some of the test results and insight gained will be summarized in this report, while the details of the tests and the mechanistic model will be presented in future publications. The present report documents (1) how knowledge gained from the scoping tests is used in the development of a recommended procedure to monitor product consistency with the purpose of demonstrating process control and (2) presents the data on which the recommended procedure is based.

Forty cans of reference CWF were made to provide the material needed for scoping tests. A large batch of salt that contained nonradioactive isotopes of the fission product elements and surrogates for the actinide elements that will be present in the actual waste salts was prepared and homogenized. The salt was made to simulate the waste salt expected to result after conditioning 300 fuel rods from driver assemblies with the electrometallurgical treatment process. The salt was mixed with granular zeolite 4A (sized to -60 +200 mesh) at a salt-to-zeolite mass ratio of 10.7/89.3 and blended at 500°C for about 20 h. Because of volume limitations in the mixer, the salt and zeolite were mixed in four separate batches following the same procedure. The batches are identified as batch numbers 2178, 2258, 3048, and 3118. As a part of the preparation procedure, the efficiency of the occlusion step was estimated by determining the amount of "free salt" that dissolved when 1 g of the salt-loaded zeolite (SLZ) was washed for about one minute with 10 mL of demineralized water. The resulting chloride concentration in the wash water was measured with a chloride ion selective electrode. According to the procedure for processing the waste form, a batch of SLZ is deemed acceptable if the mass of chloride that dissolves is not greater than 0.5 mg per gram material used in the analysis (i.e., dissolved chloride is not more than 0.05% of the total mass of SLZ used in the analysis). The free chloride in the four batches of SLZ made for the scoping tests was measured as 0.08, 0.04, 0.28, and 0.10 mg per gram for batches 2178, 2258, 3048, and 3118, respectively. All batches of SLZ were deemed acceptable for further use based on the free chloride measurement.

The four batches of SLZ were mechanically mixed with glass binder at a SLZ-to- glass mass ratios of 3:1 [HASH-1996]. Approximately 50 g aliquots of the mixture were loaded into cylindrical (2.5-cm dia, 7.6-cm length), stainless steel HIP cans and compacted. The loaded cans were heated to 500°C, evacuated with a mechanical pump, then welded shut. Five cans were processed during each of ten HIP cycles. The material from 40 cans was used in testing, and ten cans were archived. The batch of SLZ used and the HIP run were tracked for each can.

A sample of the consolidated material was dissolved and analyzed to measure the composition of the reference CWF used in the tests; the results are summarized in Table 1. Material from all 40 cans was used to evaluate the sensitivity of three test methods to product consistency: the MCC-1 static leach test [ASTM-1998B], the PCT-A, and the accessible free salt measurement (AFSM), described in Sec. IV. Monolithic samples that were used in MCC-1 static leach tests were prepared from materials recovered from individual cans. Samples for most PCTs were taken from a consolidated mixture of crushed material recovered from 28 cans,

although a few PCTs were conducted with crushed material recovered from a single can. AFSMs were made with crushed material from individual cans.

Table 1. Measured Composition of Reference CWF

Element	Mass %	Element	Mass %	Element	Mass %	Element	Mass %
Al	13.3	Cl[1]	4.8	La	0.04	Rb	0.02
B	1.26	Cs	0.15	Li	0.42	Si	19.7
Ba	0.08	Eu	<0.002	Na	12.0	Sm	0.03
Ca	0.24	I[1]	0.1	Nd	0.16	Sr	0.04
Ce	0.10	K	1.71	Pr	0.06	Y	0.01

[1]Calculated from batch concentration. The balance is primarily oxygen, although there are also small amounts of other elements such as Mg, Zr, and Zn.

The MCC-1 tests were conducted for 3 days with samples from each can to measure the can-to-can variation in the MCC-1 response. Monolithic samples were prepared by core drilling the HIPed material from the center of the can, then cutting disk-shaped wafers from the core. Disks taken from two locations from some cores were subjected to 3-day MCC-1 tests to measure the within-can variation in the test response. Disks of material from near the top of the core from each can were tested; disks from the middle of cores from four cans and from the bottom of cores from four cans were also used in MCC-1 tests. The variation in the release of elements from the discs taken from the top, middle, and bottom samples was compared to the total variations of materials made with the same SLZ, variations of materials made in the same HIP run, and variation of all material used in the tests.

The annular material remaining in 28 cans after the cores were taken and the cored material that remained after all disk samples had been prepared were crushed and sieved to isolate the –100 +200 and the –200 +325 mesh fractions. Most of the separated –100 +200 mesh fractions was consolidated and mechanically mixed for use in PCTs. Five replicate PCTs were conducted for 7 days with the consolidated material to measure the reproducibility of PCT execution. Ten PCTs were conducted for 7 days with material taken from ten cans to compare the can-to-can variation in the PCT response with the overall reproducibility of the PCT method. Material in the –200 +325 mesh fraction from each of 37 cans was used in AFSM conducted to measure the amount of chloride that dissolved in water at room temperature. The AFSM results were also used to help interpret the MCC-1 and PCT results.

The tests conducted with reference CWF material from each can are summarized in Table 2. The table also includes the identification number of each can of HIPed material and the batch number of the SLZ used in each can. The can numbers are grouped according to the five cans that were processed together in the same HIP run. The field of the test matrix identifies the

cans from which material was taken for ASFM, MCC-1, and PCT. Under the "3-d MCC-1" column, the location of the sample in the core taken from the can for MCC-1 tests is identified as top, middle, or bottom. The makeup of the material consolidated for PCTs was 13.3% from HIP run 207, 13.5% from HIP run 208, 12.5% from HIP run 209, 13.7% from HIP run 210, 13.9% from HIP run 211, 16.8% from HIP run 213, and 16.2% from HIP run 214. The consolidated material was used in PCTs conducted for various durations. Cans from which some material was tested individually in 7-day PCT are identified in the "7-day PCT" column.

Table 2. Matrix of Materials and Test Samples

Can Number	SLZ Batch	HIP Run	AFSM	3-d MCC-1	PCT Mixture	7-day PCT
2071	2178	207	Yes	Top	Yes	Yes
2072	2178	207	Yes	Top, Middle	Yes	
2073	2178	207	Yes	Top	Yes	
2074 archived	2178	207				
2075	2178	207	Yes	Top	Yes	
2081	2178	208	Yes	Top	Yes	
2082 archived	2178	208				
2083	2178	208	Yes	Top	Yes	
2084	2178	208	Yes	Top, Bottom	Yes	
2085	2178	208	Yes	Top	Yes	Yes
2091	2178	209	Yes	Top	Yes	
2092	2178	209	Yes	Top	Yes	Yes
2093	2178	209	Yes	Top	Yes	
2094	2258	209	Yes	Top, Bottom	Yes	
2095 archived	2258	209				
2101	2258	210	Yes	Top	Yes	
2102	2258	210	Yes	Top	Yes	
2103	2258	210	Yes	Top	Yes	
2104	2258	210	Yes	Top	Yes	Yes
2105 archived	2258	210				
2111	2258	211	Yes	Top	Yes	
2112	2258	211	Yes	Top, Bottom	Yes	
2113	2258	211	Yes	Top	Yes	
2114 archived	2258	211				
2115	2258	211	Yes	Top	Yes	
2121	2258	212	No	Top, Bottom		
2122	2258	212	No	Top		
2123 archived		212				
2124	3048	212	Yes	Top		
2125	3048	212	Yes	Top		
2131 archived	3048	213	Yes			
2132	3048	213	Yes	Top	Yes	
2133	3048	213	Yes	Top	Yes	Yes
2134	3048	213	Yes	Top	Yes	Yes
2135	3048	213	Yes	Top	Yes	
2141	3048	214	Yes	Top	Yes	
2142 archived	3048	214				
2143	3048	214	Yes	Top	Yes	Yes
2144	3048	214	Yes	Top, Middle	Yes	
2145	3048	214	Yes	Top	Yes	Yes
2151 archived		215				
2152	3118	215	Yes	Top		
2153	3118	215	Yes	Top		
2154	3118	215	Yes	Top		
2155	3118	215	Yes	Top, Middle		Yes
2161	3118	216	Yes	Top, Middle		Yes
2162	3118	216	Yes	Top		
2163	3118	216	Yes	Top		
2164 archived	3118	216				
2165	3118	216	Yes	Top		

IV. TEST METHODS

A. Accessible Free Salt Measurement

The AFSM was performed with the –200 +325 mesh fraction of the crushed material. This size fraction was used instead of the –100 +200 mesh fraction to allow most of the –100 +200 mesh fraction to be used for long-term testing. The crushed material was washed with absolute ethanol to remove fines. A 1-g sample of material was then washed with a 10-g aliquot of demineralized water in an ultrasonic bath for two minutes, then allowed to settle for eight minutes. The water was decanted and passed through a 0.1-μm-pore filter (0.02-μm-pore filter was used for some samples). The material was washed again with another 10-g aliquot of demineralized water following the same procedure. The two wash solutions were combined and analyzed with a chloride ion selective electrode. The percent chloride loss was calculated by dividing the mass of chloride measured in the wash solutions by the mass of chloride calculated to be in the reference CWF sample.

B. MCC-1 Static Leach Tests

The MCC-1 tests to measure the between-can and within-can variations were conducted with monolithic samples and demineralized water at 90°C for three days. Samples were prepared by core-drilling the HIPed material using ethanol as a cutting fluid. The cores were then dry-cut with a diamond wafering blade into disks about 11-mm in diameter and 2-mm thick. The faces of the cores were dry-ground to a 240-grit finish. Samples were utrasonically cleaned with absolute ethanol for about two minutes to remove fines, then they were dried in a 40°C oven. The dimensions of each sample were measured with calipers, and the geometric surface area was calculated. The mass of demineralized water, in grams, that was used in each test was 10 times the surface area of the sample, in cm^2, so that the glass surface area/solution volume (S/V) ratio was about 10 m^{-1} for each test. Tests were conducted in Teflon vessels. After three days (72±2h), the vessels were removed from the oven. Aliquots of the solution were taken to measure the pH (with a combination electrode) and chloride concentration (with a chloride ion selective electrode). The remaining solution was passed through a 0.45-μm-pore cellulose acetate filter and stabilized by adding a few drops of ultrapure concentrated nitric acid. The sample was removed from the vessel, which was rinsed three times with demineralized water, then filled with demineralized water. A few drops of ultrapure concentrated nitric acid were added, and the vessel was resealed and placed back into the 90°C oven for about 16 h. This was done to acid-strip the vessel and dissolve any material that had become fixed to the vessel walls during the test. The acid-strip solution was removed from the vessel and passed through a 0.45-μm-pore filter. The test solutions and the acid strip solutions were analyzed with inductively coupled plasma-mass spectrometry (ICP-MS). Test blanks were conducted by adding demineralized water to Teflon vessels. The blank test solutions were treated identically to tests with the ceramic waste form. The mass of material released from the sample during the test was calculated by adding the masses in the test solution and acid strip and subtracting the mass in the blank. The normalized elemental mass loss was calculated by dividing the mass of an element released from the sample by the surface area of the sample and by the mass fraction of the element in the reference CWF.

C. Product Consistency Tests

Product consistency tests were conducted with demineralized water and the –100 +200 mesh fraction. The crushed material was washed with absolute ethanol to remove fines. Some PCTs were conducted with a mixture of material from 28 cans, and other PCTs were conducted with material from a single can. Most PCTs were conducted in Teflon vessels, but a few were conducted in Type 304L stainless steel or titanium vessels. Tests were conducted by placing about 1 g of material in the vessel and adding a mass of water equal to ten times the mass of reference CWF that had been added. The vessels were sealed and placed in a convection oven set at 90°C. Tests were conducted for seven days (168±3 h). Vessels were then removed from the oven and opened. Aliquots of the solution were taken to measure the pH and the chloride ion concentration. The remaining solution was passed through a filter with a 0.45-μm-pore and stabilized with a few drops of ultrapure concentrated nitric acid. The reacted solids were removed from the vessel, and the vessel was rinsed and acid-stripped. The acid strip and test solutions were analyzed with ICP-MS. Blank tests were conducted, and the solutions were analyzed with ICP-MS.

V. TEST RESULTS

The solution concentrations of several components were measured to determine the amount of material dissolved during a test. The mass of material released from the sample was calculated based on the sum of the amounts measured in the test solution and acid strip solution minus the mass measured in the blank test solution. In practice, the concentrations in the acid strip and blank test solutions were negligible compared to the concentrations in the test solutions. The measured concentration of an element was normalized to the mass fraction of that element in the waste form to compare the release of different elements as percent loss in AFSM and MCC-1 tests. For some MCC-1 tests and the PCTs, the normalized elemental mass loss was calculated by dividing the mass of an element released from the sample by the surface area of the sample and by the mass fraction of that element in the reference CWF. The expression for the normalized mass loss based on element i is

$$NL(i) = (m_{i,ts} + m_{i,as} - m_{i,b}) / (S \bullet f_i) \qquad (1)$$

where $m_{i,ts}$ is the mass in the test solution, $m_{i,as}$ is the mass in the acid strip solution, $m_{i,b}$ is the mass in the blank solution, S is the surface area, and f_i is the mass fraction of element i in the reference CWF. The mass of element i in each solution was calculated from the measured concentration and solution volume.

A. Measurement of Accessible Free Salt

1. AFSM Results

The –200 +325 mesh crushed materials recovered from the cans was subjected to AFSMs. The materials were washed once with absolute ethanol before the measurement. The mass of sample used in the AFSM and the measured chloride concentrations are given in Table 3. Replicate measurements were made with samples from some cans. The per cent of total chloride in the sample that was dissolved in the AFSM is plotted in Fig. 3. The results for samples prepared from the same SLZ batch are grouped together, and the symbols identify the batch of SLZ used in each can (see Table 2 for correspondence of can with HIP run and SLZ batch). The variation in the amount of released free salt between cans is greatest for cans made with SLZ batches 3048 and 3118. The batch of SLZ used to make the reference CWF had a greater effect on the test response than the particular HIP run. This is shown most clearly in the results for cans made in HIP runs 214 (cans 2141, 2143, 2144, and 2145) and 216 (cans 2161, 2162, 2163, and 2165). The variation in the response of cans made in each HIP run is the same as the variation in the response of materials made with the different SLZ batches (SLZ batches 3048 and 3118, respectively). The overall variation in the chloride release from material (i.e., dissolution of halite) in all the cans that were tested is about a factor of two.

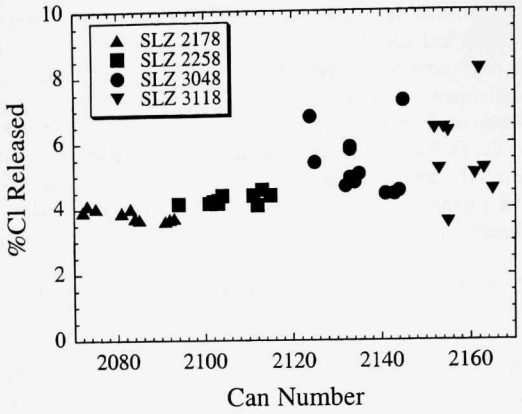


Fig. 3.
Mass Percent of Chloride Released in AFSM Conducted with Reference CWF from Individual Cans

2. Effects of Different Procedures for Fines Removal

A study of the effects of the procedure to remove fine material on the subsequent measurement of accessible free salt was conducted with the –100 +200 mesh fractions of five samples. Samples of crushed material from five cans were washed following one of two procedures before the AFSM was run. In Procedure I, the material was first washed three times with demineralized water, then washed four times with 100% ethanol. In Procedure II, the material was washed seven times with 100% ethanol. The subsequent release of chloride from the washed material was measured following the AFSM procedure. The results are summarized in Table 4 as the percent of total chloride in the sample that dissolved. This was calculated by dividing the mass of chloride released to the wash solution by the mass of chloride in the test sample. (The reference CWF contains 0.048 g chloride per g reference CWF.) It is evident in the AFSM results for all five samples that washing with water results in the dissolution of a much greater fraction of available salt than washing with 100% ethanol.

Procedure III used a consolidated mixture of material from the five samples, which was not washed before the AFSM. The fraction of chloride released in this AFSM was 2.08%. The average fraction of chloride released in the five AFSMs with material that had been washed seven times with 100% ethanol (Procedure II) is 1.77%. This implies that only about 15% of the chloride at the surface of the samples that was accessible was actually dissolved during the ethanol washes.

Table 3. Results of Accessible Free Salt Measurements

Samples Made with SLZ Batch 2178					Samples Made with SLZ Batch 3048				
Can No.	Sample Mass, g	Water Mass, g	[Cl], mg/L	% Cl Released,	Can No.	Sample Mass, g	Water Mass, g	[Cl], mg/L	% Cl Released
2072	1.00	20	94.7	3.94	2124	1.00	20	154	6.38
2073	1.03	10	204	4.11	2124	1.00	18	192	7.17
2075	1.01	20	98.0	4.03	2125	1.00	20	161	6.01
2081	0.99	10	188	3.96	2125	1.00	18	115	4.77
2081	0.99	20	87.3	3.67	2132	1.00	20	112	4.66
2081	1.00	20	97.6	4.06	2133	0.99	20	117	4.92
2083	1.01	20	97.6	4.02	2133	1.00	17	163	5.77
2084	1.03	10	185	3.73	2133	1.00	16	176	5.86
2085	1.00	20	88.7	3.69	2134	1.00	20	115	4.77
2091	1.01	10	184	3.78	2135	1.00	20	121	5.04
2091	1.00	17	101	3.57	2141	1.00	20	107	4.43
2091	1.00	20	84.8	3.53	2143	1.00	20	107	4.43
2092	0.99	10	176	3.69	2144	1.00	20	109	4.53
2093	1.00	20	89.5	3.72	2145	1.00	20	174	7.25
Samples Made with SLZ Batch 2258					Samples Made with SLZ Batch 3118				
Can No.	Sample Mass, g	Water Mass, g	[Cl], mg/L	% Cl Released,	Can No.	Sample Mass, g	Water Mass, g	[Cl], mg/L	% Cl Released
2094	1.00	20	99.5	4.14	2152	1.01	20	143	5.88
2101	1.00	20	100	4.16	2152	1.00	18	183	6.86
2102	0.99	10	201	4.21	2153	1.00	20	124	5.14
2103	1.00	10	200	4.15	2154	1.00	20	154	6.39
2103	1.00	20	102	4.24	2155	1.01	20	86.0	3.54
2104	0.99	10	209	4.39	2155	1.00	18	168	6.28
2111	1.00	20	106	4.40	2161	0.99	20	119	5.00
2112	1.00	20	98.8	4.11	2162	1.04	10	359	7.17
2113	1.02	10	196	4.00	2162	1.00	20	188	7.81
2113	1.00	18	137	5.13	2162	1.00	18	254	9.51
2115	1.00	20	106	4.40	2163	1.00	20	124	5.15
					2165	1.01	20	110	4.51

Table 4. Percent Chloride Released in AFSM and PCT after Different Wash Procedures

Procedure	Sample Number				
	2085	2113	2133	2143	2145
I. AFSM with –100+200 mesh fraction after 3 water washes and 4 ethanol washes	0.03	0.02	0.04	0.03	0.03
II. AFSM with –100+200 mesh fraction after 7 ethanol washes	1.07	1.59	1.35	1.86	2.68
III. AFSM with –100+200 mesh fraction of consolidated samples	2.08	2.08	2.08	2.08	2.08
IV. PCT with –100+200 mesh fraction after 7 ethanol washes	1.56	N/A	2.19	2.18	3.39
V. AFSM with –200 +325 mesh material after 1 ethanol wash	3.70	5.20	5.60	4.45	7.28

The chloride losses measured in 7-day PCTs conducted with the –100 +200 mesh fraction after seven washes in 100% ethanol are included in Table 4 as Procedure IV. There is good agreement between the chloride loss from material that was washed seven times with ethanol in the AFSM (Procedure II) and in the 7-day PCTs. This indicates that most of the chloride loss in the PCTs occurs due to dissolution of accessible surface salt when water first contacts the material; longer exposure in the PCTs results in the dissolution of only a small amount of additional halite.

Finally, the results of AFSM conducted with the –200 +325 mesh fraction of the same materials that were washed seven times with 100% ethanol are listed as Procedure V. Those results are higher than tests conducted with –100 +200 mesh fraction of the same material because of the higher specific surface area of the smaller particle size (about two times greater) and the greater total amount of salt that is exposed. Note that the relative values of the chloride release from the different samples are the same in the ASFM with the –200 +325 mesh fraction (Procedure V) and in the PCT (Procedure IV).

B. MCC-1 Tests

Three-day MCC-1 tests were conducted with samples taken from near the top of each of the forty cans. Samples from near the center or bottoms of some cans were also tested. The complete set of results of solution analyses for these tests are presented in Appendix A. The results for key elements are summarized in Table 5.

Table 5. Elemental Release in 3-day MCC-1 Tests

Can No. (position)	Solution Concentrations, mg/L						
	Al	B	Cl	Cs	I	Na	Si
2071 (top)	6.59	0.060	8.40	0.115	0.012	9.0	7.19
2072 (top)	5.73	0.048	6.01	0.106	0.009	9.2	8.39
2072 (middle)	5.91	0.082	8.98	0.096	0.019	10.9	7.26
2073 (top)	6.73	0.048	7.81	0.090	0.013	8.1	7.60
2075 (top)	6.18	0.062	8.38	0.097	0.012	8.5	6.84
2081 (top)	4.53	0.060	6.53	0.100	0.010	9.5	5.27
2083 (top)	6.32	0.043	6.45	0.084	0.014	8.4	6.39
2084 (top)	6.43	0.078	6.40	0.124	0.019	10.0	7.11
2084 (bottom)	**8.53**	0.063	6.40	0.110	0.030	11.8	7.67
2085 (top)	7.20	0.071	6.07	0.116	0.015	9.7	6.78
2091 (top)	6.74	0.074	6.17	0.113	0.013	9.8	6.65
2092 (top)	5.84	0.092	5.49	0.081	0.012	9.9	5.93
2093 (top)	6.94	0.071	5.54	0.095	0.012	9.0	5.87
2094 (top)	6.53	0.084	6.07	0.103	0.013	11.2	6.14
2094 (bottom)	**8.32**	0.078	6.07	0.099	0.025	12.9	7.59
2101 (top)	**8.47**	0.104	7.08	0.136	0.017	11.9	7.40
2102 (top)	5.55	0.096	5.63	0.111	0.019	11.3	6.00
2103 (top)	7.02	0.069	5.60	0.113	0.026	10.4	7.29
2104 (top)	5.38	0.089	6.32	0.126	0.014	11.7	5.84
2111 (top)	7.41	0.107	5.88	0.120	0.016	11.1	7.28
2112 (top)	6.51	0.125	9.34	0.109	0.018	12.7	6.86
2112 (bottom)	7.02	0.071	9.34	0.128	0.021	13.2	**7.93**
2113 (top)	5.12	0.104	10.73	0.228	0.014	13.4	5.90
2115 (top)	4.95	0.096	5.86	0.104	0.012	9.7	5.97
2121 (top)	**8.04**	0.095	6.75	0.107	0.017	11.2	6.87
2121 (bottom)	**8.27**	0.064	6.88	0.107	0.015	11.4	**10.21**
2122 (top)	**8.50**	0.099	7.77	0.172	0.017	12.0	6.87
2124 (top)	6.11	0.096	11.60	0.267	**0.041**	**16.0**	6.48
2125 (top)	4.75	0.073	10.25	0.146	0.015	10.3	5.75
2132 (top)	7.21	0.066	7.62	0.160	0.019	12.4	7.18
2133 (top)	6.02	0.105	**32.0**	**0.476**	**0.082**	**24.0**	5.97
2134 (top)	6.05	0.082	6.81	0.112	0.027	12.9	6.52
2135 (top)	7.33	0.097	7.56	0.158	**0.049**	12.5	6.47
2141 (top)	4.93	0.104	7.20	0.144	0.027	11.9	5.57
2143 (top)	5.83	0.075	7.29	0.121	0.024	13.4	6.27
2144 (top)	6.04	0.103	7.15	0.147	0.024	12.4	6.39
2144 (middle)	5.59	0.099	7.27	0.228	0.017	11.7	6.85
2145 (top)	4.52	**0.393**	**34.0**	**1.25**	**0.063**	**23.7**	5.20
2152 (top)	**8.15**	0.062	8.16	0.147	0.032	12.9	6.99
2153 (top)	6.40	0.070	6.47	0.120	0.015	9.9	6.29
2154 (top)	6.62	0.107	13.8	0.346	0.030	**18.9**	6.69
2155 (top)	7.05	0.114	8.59	0.150	0.020	13.4	7.36
2155 (middle)	7.05	0.123	**25.0**	**0.956**	0.025	**19.9**	7.37
2161 (top)	6.89	0.120	9.87	0.288	0.016	13.4	7.76
2161 (middle)	7.32	0.109	9.87	0.142	**0.042**	13.5	7.28
2162 (top)	**8.16**	0.092	7.44	0.180	0.015	12.7	**8.48**
2163 (top)	6.66	0.095	7.10	0.135	0.015	12.7	7.52
2165 (top)	6.18	0.095	9.20	0.186	0.014	15.0	7.15
mean± std dev[1]	6.58±1.07	0.092±0.049	9.00±5.91	0.19±0.21	0.022±0.014	12.3±3.4	6.85±0.91
% rsd[2]	16	53	66	112	63	27	13

[1]std dev = standard deviation

[2]rsd = relative standard deviation or 100 times the standard deviation divided by the mean.

The concentrations of several key components in the waste form were used to calculate the percent of the mass available in the sample that was released during the test. The percent releases of Cl, I, Cs, Al, Si, and B are plotted in Fig. 4a to show the relative can-to-can variations in these elements. The percent releases of Cl, I, and Cs are higher than those of Al, Si, and B for all cans. The releases of Al, Si, and B are similar for all cans, but the releases of Cl, I, and Cs from the different cans vary significantly. The vertical lines in the plot distinguish between materials made with SLZ batches 2178, 2258, 3084, and 3118. The greatest variations occur for cans made with SLZ batches 3048 and 3118.

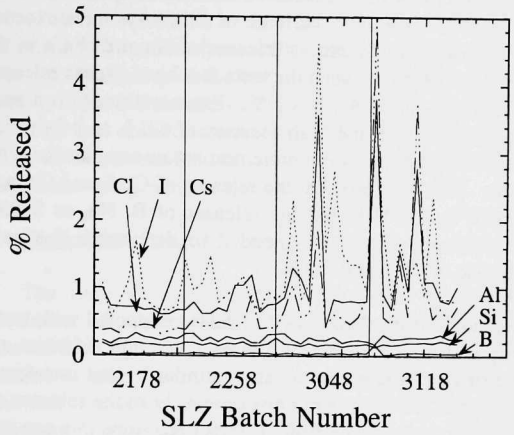
The differences in the release results for the elements in Fig. 4a reflect the difference in the solubilities of the phases that contain them: Al and Si are present in the sodalite and glass binder; B is present in the glass binder; and Cl, Cs, and I are probably present in the sodalite, glass, and salt phases. The releases of Cl, I, and Cs are higher than those of Al, B, and Si for all samples because the salt is more soluble than the sodalite or glass binder phases in demineralized water at room temperature. It is likely that the can-to-can variation in the release of Cl, I, and Cs reflects small differences in the amount of halite formed during preparation of the reference CWF, that is, how well the salt is incorporated into the zeolite and is retained in the sodalite during HIPing. The releases of Na and Cl shown in Fig. 4b indicate that the fraction of the total amount of Cl that is in the halite is greater than the fraction of the total Na present in the halite.

We emphasize that only a very small fraction of the total amounts of any of these elements is released during the test and that the fractions that dissolve are similar to the fraction of material at the surface. The amount of silicon dissolved in each MCC-1 test corresponds to dissolution of a layer of material about 1 μm thick. For the monoliths used in the tests, the outer 1 μm of the sample comprises less than 0.2 % of the sample volume.

These results indicate that (1) the amounts of salt on the surface varies from sample to sample, and (2) the 3-day MCC-1 test is sufficiently sensitive to distinguish between small variations in the amounts of exposed salt. They also indicate that the release of chloride (and the amount of exposed salt) is not correlated with the dissolution of the sodalite or glass binder phases. This is because the dissolution of those phases is reflected by the releases of silicon and boron, which are essentially the same for all cans.

We emphasize that only a very small fraction of the total amounts of any of these elements is released during the test and that the fractions that dissolve are similar to the fraction of material at the surface. The amount of silicon dissolved in each MCC-1 test corresponds to dissolution of a layer of material about 1 μm thick. For the monoliths used in the tests, the outer 1 μm of the sample comprises less than 0.2 % of the sample volume.

These results indicate that (1) the amounts of salt on the surface varies from sample to sample, and (2) the 3-day MCC-1 test is sufficiently sensitive to distinguish between small variations in the amounts of exposed salt. They also indicate that the release of chloride (and the amount of exposed salt) is not correlated with the dissolution of the sodalite or glass binder phases. This is because the dissolution of those phases is reflected by the releases of silicon and boron, which are essentially the same for all cans.



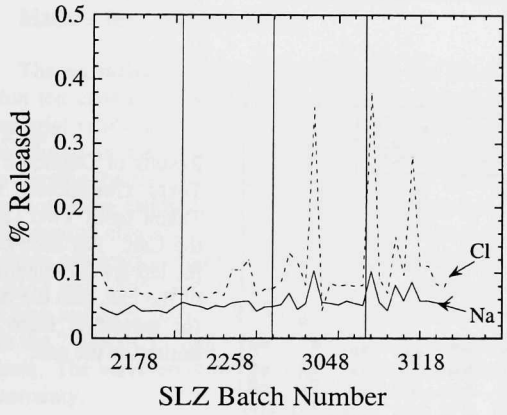


Fig. 4. Percent of Components Released in 3-day MCC-1 Test with Material from Each Can: (a) Al, B, Cl, Cs, I, and Si and (b) Na and Cl.

The mean and standard deviation for the measured concentrations in all tests are included in Table 5. The values for individual tests that are greater than the mean plus one standard deviation are shown in bold font. The values in bold are termed "excessive." Note that there is some correlation between the tests that have excessive releases of alkali metals and chloride or iodide. For example, the tests that have excess release of Cl always have excess releases of Cs and Na. Tests with excess release of I had excess releases of Na and/or Cs in three of the five cases. However, there is no correlation between the tests that have excess release of Cl, Cs, or I and the tests that have excess release of Al, B, or Si. Examination with a scanning electron microscope of some of the samples that had high releases of Cl, I, and Cs revealed that those samples were not as well consolidated (i.e., were more porous) as samples for which the releases of those elements were lower. This suggests that the releases of Cl, I, and Cs may indicate how well the CWF is consolidated by the HIP, but the releases of B, Na, or Si do not. A more thorough examination of additional samples is needed to determine if a correlation exists between porosity and the release of Cl, I, and/or Cs.

Figure 5 shows the results of the 3-day MCC-1 tests conducted with two samples taken from different locations (top and middle or bottom) in each of eight different cans of material. The variation in the release of elements within a can is similar to that measured between cans (compare Fig. 5 with Figs. 3 and 4); this is seen most obviously in the releases of Cl, Cs, and I. The variation in NL(i) between material taken from different positions in a can and between cans is about 2 times except for material taken from cans 2155 and 2161. The materials in those cans were made with SLZ from batch 3118. The releases of Cl, Cs, and K from the middle of can 2155 are more than 2 times higher than those from the top of can 2155. The release of iodide is higher from the sample taken from the middle of can 2161 than from the sample taken from the top of the can, but the releases of Cs and K are lower.

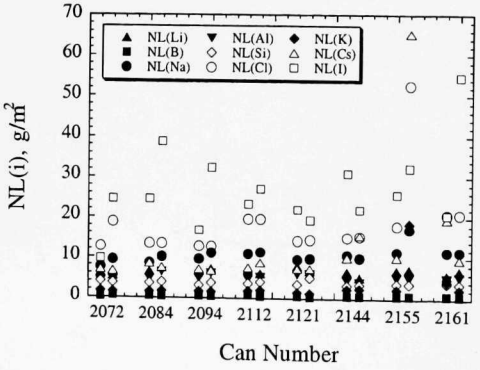


Fig. 5.
Results of Duplicate 3-Day MCC-1 Tests Conducted with Samples Taken from Two Locations within the Can. For each can, the data on the left are for materials from the top of the can, and the data on the right for materials from the middle or bottom of the can.

C. Product Consistency Tests

We conducted five 7-day PCTs using a consolidated mixture of material from 28 cans and ten 7-day PCTs using material recovered from individual cans. All PCTs were done at 90°C with the –100 +200 mesh fraction of crushed material. The material was washed seven times with absolute ethanol to remove fines. The tests conducted with the mixed material provide a measure of the reproducibility of the test method, while the tests conducted with material from individual cans provide a measure of the sensitivity of the PCT to variations in the material composition. The test solutions were analyzed with ICP-MS and a chloride ion selective electrode. The solution results are compiled in Appendix B. The calculated normalized elemental mass losses are presented in Table 6.

1. Consolidated Mixed Material

The normalized elemental mass losses for 7-day PCTs conducted with the consolidated mixture of reference CWF material are plotted in Fig. 6. The uncertainty bars are drawn to show 15% analytical uncertainty. The results of replicate tests conducted in Teflon, Type 304L stainless steel, and titanium vessels indicate no measurable effect of the vessel material. The test-to-test variations in the normalized elemental mass losses are within the analytical uncertainty for the plotted elements. The variation for each element measured in these tests is used to represent the precision of the data generated in PCTs. The noticeably higher normalized mass losses for Cl, Cs, and I are due to the dissolution of accessible free salt. The samples used in these PCTs were rinsed only with ethanol to remove fines; they were not water washed.

2. Material from Individual Cans

The normalized elemental mass losses for the PCTs conducted with material recovered from ten cans are plotted in Fig. 7. The mean of the tests with the consolidated mixture of material is included on the left hand side. In this figure, two times the standard deviations measured in the tests with the consolidated samples is used to represent the uncertainty in running the test and analyzing the solutions. Differences in the PCT responses of materials recovered from different cans exceed the testing and analytical uncertainty for Cl, Cs, I, and K. The results also show that, like the AFSM and the 3-day MCC-1 tests, the PCT response for these elements is higher for materials made with SLZ batches 3048 (can numbers 2133, 2134, 2143 and 2145) and 3118 (can numbers 2155 and 2161) than those made with batches 2178 (can numbers 2071, 2085, and 2092) and 2258 (can number 2104). The NL(Cl) and NL(Cs) in the 7-day PCTs vary by factors of 1.5 to 3, compared to factors of 6-15 in the 3-day MCC-1 tests. The variation in the normalized mass losses based on Al and Si are within the analytical uncertainty.

Table 6. Normalized Elemental Mass Losses for 7-day PCT with Reference CWF

	Normalized Mass Losses, g/m^2								
	Al	B	Cl	Cs	I	K	Li	Na	Si
Test Vessel Type				Consolidated Material					
Teflon-1	0.024	0.11	1.1	1.0	0.52	0.18	0.25	0.29	0.031
Teflon-2	0.028	0.11	1.2	1.2	0.57	0.19	0.26	0.29	0.037
Teflon-3	0.037	0.093	1.3	1.1	0.66	0.18	0.39	0.28	0.036
Teflon-4	0.032	0.11	1.3	1.1	0.57	0.18	0.32	0.28	0.034
Teflon-5	0.035	0.097	1.2	1.3	0.71	0.18	0.38	0.28	0.040
Steel-1	0.032	0.14	1.2	1.1	0.61	0.23	0.33	0.27	0.040
Steel-2	0.033	0.11	1.2	1.3	0.65	0.20	0.29	0.27	0.039
Steel-3	0.026	0.19	1.3	1.0	1.05	0.18	0.21	0.12	0.038
Steel-4	0.024	0.22	1.3	1.0	1.00	0.19	0.20	0.14	0.038
Titanium-1	0.043	0.14	1.4	1.0	0.68	0.22	0.39	0.28	0.046
Titanium-2	0.043	0.14	1.4	1.1	0.72	0.22	0.42	0.27	0.045
Overall mean ± 1 std dev	0.032 ±0.007	0.13 ±0.04	1.3 ±0.09	1.1 ±0.1	0.70 ±0.17	0.20 ±0.02	0.31 ±0.08	0.25 ±0.06	0.039 ±0.004
% rsd	18	17	8	9	11	10	18	4	13
Can No.				Material from Individual Cans					
2071	0.039	0.096	0.61	0.55	0.44	0.11	0.31	0.13	0.039
2085	0.042	0.098	0.68	0.88	0.48	0.11	0.38	0.15	0.045
2092	0.041	0.10	0.59	0.88	0.29	0.20	0.31	0.15	0.047
2104	0.041	0.12	0.78	0.72	0.40	0.16	0.34	0.20	0.045
2133	0.043	0.12	0.95	1.0	0.39	0.16	0.41	0.21	0.049
2134	0.035	0.15	0.90	1.1	0.28	0.21	0.32	0.21	0.045
2143	0.043	0.14	0.95	1.3	0.39	0.21	0.40	0.22	0.054
2145	0.037	0.19	1.47	1.3	0.50	0.68	0.45	0.31	0.045
2155	0.027	0.20	1.33	1.5	0.89	0.22	0.21	0.15	0.039
2161	0.026	0.21	1.23	1.1	1.02	0.16	0.21	0.14	0.039
Overall mean ± 1 std dev	0.037 ±0.006	0.14 ±0.04	0.95 ±0.31	1.0 ±0.3	0.51 ±0.25	0.22 ±0.17	0.33 ±0.08	0.19 ±0.05	0.045 ±0.00
% rsd	17	31	32	28	49	75	24	29	11

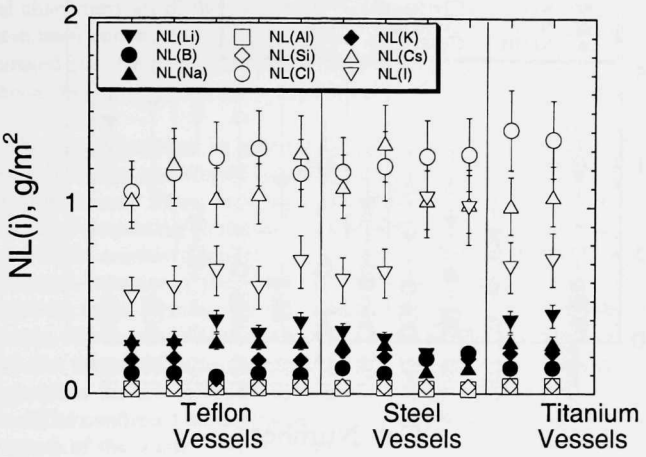


Fig. 6. Results of Replicate 7-Day PCTs Conducted with a Mixture of Material from 28 Cans. Tests were conducted in Teflon, Type 304L stainless steel, or titanium vessels.

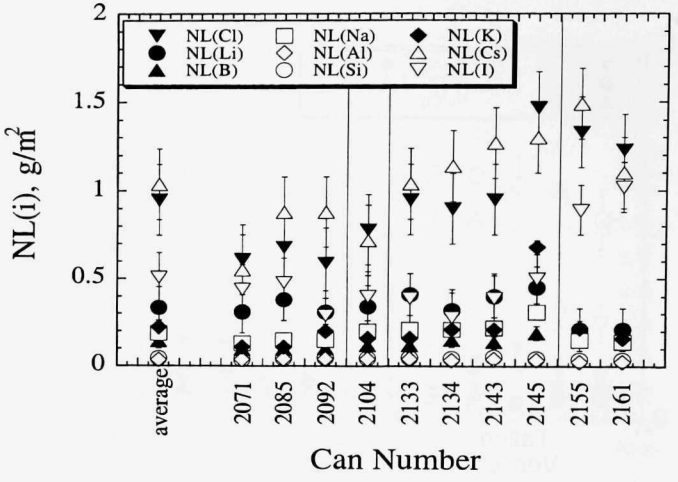


Fig 7. Results of PCTs Conducted with Reference CWF from Individual Cans

VI. DISCUSSION

A test method is needed to monitor the consistency of waste forms destined for disposal at a high-level waste repository. This test must be sensitive to variations in materials and processing conditions. Development of such a test requires insight into the chemical and physical characteristics of the material of interest, as well as its dissolution behavior. Scoping tests have been conducted with the ceramic waste form that is being developed to immobilize contaminated salts from the electrometallurgical conditioning process, and the results have led to an understanding of the waste form microstructure and key corrosion modes.

The tests described in this report were conducted to measure the variations in the dissolution behavior of different batches of reference CWF material that were made following the same procedure. These tests provide a measure of the reproducibility of the waste form under identical processing conditions with regard to its dissolution behavior. They also provide a measure of the sensitivities of three test methods (AFSM, MCC-1, and PCT) to differences in the dissolution behavior of the materials. These tests established the limits of the consistency with which the waste form can be made and the sensitivity of the test methods to variations in the waste forms. Both must be known to evaluate the effects of changes in processing conditions. (The fact that the materials used for testing were not made with the same equipment that will be used to produce the actual waste forms is not expected to affect the conclusions of this study. This should be confirmed during the demonstration phase of the project.) The reproducibility of other aspects of the waste form that may be important to its long-term performance (e.g., the consistency of the microstructure) was not evaluated.

The reference CWF material contains two predominant phases: sodalite and glass binder. Small amounts of other phases are distributed within the sodalite and glass binder phases. Radionuclides are probably present to some degrees in all phases, but the majority of radionuclides are present as separate oxide phases that form as inclusions within the glass binder and, to a lesser extent, between sodalite grains. The release of radionuclides can occur when the phases containing them dissolve, whether they be separate oxide phases, the glass, or sodalite. Thus, the effectiveness of the CWF in immobilizing radionuclides is determined primarily by the solubilities of the component phases and the surface area of each phase that is exposed.

The property monitored for vitrified HLW forms is the glass composition. Key properties for process control at the DWPF, such as melt viscosity, are correlated with the composition. Variation in the composition of the waste stream will alter the composition of the waste glass. This will result in different concentrations of key glass matrix components (Al, B, Fe, Si, etc.), which control the glass dissolution behavior. Thus, the predominant role of product consistency testing of waste glasses is to verify that the glass composition is within the acceptable composition space.

The composition of the ceramic waste form and its durability are not as sensitive to variations in the composition of the waste salt. This is because the matrix is composed primarily of sodalite and the glass binder, the compositions of which are not expected to vary significantly between waste forms. The variations in the composition of the waste stream will be the amount of fission products and actinides. These radionuclides do not contribute significantly to the

structure or chemical durability of the waste form. Hence, the primary role of product consistency testing will be to verify that salt has been occluded, the waste form contains the proper ratio of sodalite and glass binder phases, and that those phases have been consolidated.

Our understanding of the dissolution mechanism of the ceramic waste form indicates that the test used to monitor the product consistency should be sensitive to the dissolution of the free salt (halite), sodalite, and glass binder phases. This can be accomplished by using the same method that is used for high-level waste glasses, namely, PCT-A. The PCT-A includes the option to analyze the water wash solution generated during sample preparation if it is suspected that soluble phases are present in the waste form. By including the water wash option, the free salt can be measured. In fact, the water wash step of the PCT-A is similar to the AFSM discussed earlier in this report; only the size fractions of crushed material and the material/water mass ratios are different. To reduce the contribution of fines to the measurement, the crushed material should be washed with ethanol prior to the water wash. Measurement of the free salt exposed at the surface of the –100 +200 mesh sample serves two purposes. First, the amount of exposed free salt indicates the efficiency with which the salt was retained in the sodalite during processing. This information feeds back to process control and could be used to adjust the salt loading in the zeolite. Second, it provides an estimate of the amounts of Cs and I in the halite phase.

After water-washing, the material would then be subjected to the usual PCT-A procedure. Tests conducted following the PCT-A presented in this report used crushed material that was washed repeatedly with absolute ethanol to remove fines. A direct comparison of material subjected to PCT-A with and without water wash cannot be made at this time; such a data base of test results is needed to support the proposed PCT procedure. Available test results do show halite to be removed from ceramic waste form samples prepared for PCT-A tests by washing with 95% ethanol (i.e., higher releases of Cl, Cs, and I), but not from samples washed only with absolute alcohol.

VII. ACKNOWLEDGMENTS

Waste form materials were prepared by Mark Hash. Solution analyses were conducted by Activation Laboratories, Ltd. and by the Argonne National Laboratory Analytical Chemistry Laboratory. Funding provided by the U.S. Department of Energy, Nuclear Energy Research and Development Program, under Contract No. W-31-109-Eng-38.

VIII. REFERENCES

ANS-1992
"*American National Standard for Measurement of the Leachability of Solidified Low-Level Radioactive Waste*," American Nuclear Society, LaGrange, IL, ANSI/ANS-16.1 (1992).

ASTM-1998A
American Society for Testing and Materials, *Annual Book of ASTM Standards*, **12.01**, "Standard Test Method for Determining Chemical Durability of Nuclear Waste Glasses: The Product Consistency Test (PCT)," C1285-94, pp. 796-813 (1998).

ASTM-1998B
American Society for Testing and Materials, *Annual Book of ASTM Standards*, **12.01**, "Standard Test Method for Static Leaching of Monolithic Waste Forms for Disposal of Radioactive Waste," C1220-98, pp. 1-16 (1998).

BRECK-1974
D. W. Breck, "Zeolite Molecular Sieves," Krieger Publishing Co., Malabar, FL (1974).

DOE-1996
Waste Acceptance System Requirements Document (WASRD), Rev. 2, DCN 02, U.S. Department of Energy, Office of Civilian Radioactive Waste Management Report E00000000-0081101708-00001 (December 1996).

DOE-1990
Waste Form Compliance Plan for the Defense Waste Processing Facility, Rev. 0, U. S. Department of Energy report WSRC-SW4-6, Westinghouse Savannah River Company, Aiken, SC (March 1990).

HASH-1996
M. C. Hash, C. Pereira, M. A. Lewis, R. J. Blaskovitz, V. N. Zyryanov, J. P. Ackerman, "Hot Isostatic Pressing of Glass-Zeolite Composites," *in Environmental Issues and Waste Management Technologies in the Ceramic and Nuclear Industries II*, eds. V. Jain and D. K. Peeler, American Ceramic Society, pp. 135-143 (1996).

SIMPSON-1999
L. J. Simpson and C. W. VanderKooi, "Solution Exchange Corrosion Testing with a Glass-Zeolite Ceramic Waste Form in Demineralized Water at 90°C," *in Environmental Issues and Waste Management Technologies in the Ceramic and Nuclear Industries IV*, eds. J. C. Marra and G. T. Chandler, American Ceramic Society, pp. 305-312 (1999).

MORSS-1999
Private Communication, Argonne National Laboratory, 1999.

IX. APPENDICES

Column Headings Defined

The following is a listing of all column headings used with a comment describing what each one references.

Column Heading	Comments
Surface Area	Geometric surface of the disc or calculated surface area of the 100-200 mesh powder
Sample Mass	The weight of the sample after washing and drying, before testing
Water	The final volume of the demineralized water used in the test (initial volume less any lost during the test).
Dilution Factor	This adjustment accounts for the additional volume due to the addition of acid
AS Volume	The volume of acid and water used in the acid strip
Measured Concentrations	The raw data of the elemental analysis as reported
Corrected Concentrations	The measured concentrations adjusted with the dilution factor
Normalized Elemental Mass Loss (NL)	The calculated normalized release per surface area

APPENDIX A

Solution Results for MCC-1

Table A-1. Composition of CWF and the Component Phases

	Mass %								
	Li	B	Na	Al	Si	Cl	K	I	Cs
Binder Glass	<0.002	5.99	4.83	3.95	29.5	--	0.37	--	--
Salt	5.49	--	5.23	0.0	0.0	59.5	21.1	0.12	1.82
Zeolite 4A	0.00	--	14.9	18.8	18.7	--	0.04	--	--
Reference CWF	0.44	1.50	11.6	13.6	19.9	4.77	1.81	0.01	0.15

Table A-2. MCC-1 Test Results for Reference CWF from 40 HIP Cans
Test Data

Sample ID:	Surface Area, cm^2	Sample Mass, g	Water Volume, mL	Dilution Factor	Acid Strip, mL	pH
MRD-3-2071	2.6863	0.47758	26.84	1.002	26.96	9.2
MRD-3-2072	2.9529	0.61629	28.18	1.002	26.84	9.0
MRD-3-2073	2.7044	0.44639	27.01	1.002	26.66	8.9
MRD-3-2075	2.8548	0.55765	27.92	1.002	26.82	8.7
MRD-3-2081	2.7372	0.47414	23.90	1.003	28.07	9.0
MRD-3-2083	2.7334	0.49879	27.35	1.004	27.75	8.4
MRD-3-2084	2.6720	0.47539	26.78	1.002	27.88	8.7
MRD-3-2085	2.8541	0.52915	28.55	1.004	28.20	8.8
MRD-3-2091	2.7213	0.48545	27.26	1.003	27.65	8.8
MRD-3-2092	2.7156	0.48260	27.19	1.004	27.69	8.8
MRD-3-2093	2.7448	0.51499	27.50	1.003	27.91	8.7
MRD-3-2094	2.7872	0.53128	27.93	1.003	27.44	8.8
MRD-3-2101	2.7326	0.50222	27.35	1.005	27.79	8.6
MRD-3-2102	2.7150	0.48848	27.15	1.003	27.95	8.6
MRD-3-2103	2.7125	0.48069	27.14	1.003	27.78	8.7
MRD-3-2104	2.7308	0.50384	27.31	1.004	27.90	8.7
MRD-3-2111	2.6742	0.49205	26.80	1.002	27.90	8.7
MRD-3-2112	2.6770	0.49147	26.78	1.004	27.83	8.7
MRD-3-2113	2.6984	0.49347	27.03	1.005	27.56	8.5
MRD-3-2115	2.7131	0.50937	27.18	1.003	27.90	8.7
MRD-3-2121	2.6673	0.49584	26.73	1.005	27.90	8.6
MRD-3-2122	2.6830	0.49357	26.87	1.003	28.19	8.4
MRD-3-2124	2.7389	0.51895	27.41	1.004	27.73	8.6
MRD-3-2125	2.6289	0.45623	26.40	1.003	27.39	8.3
MRD-3-2132	2.6890	0.50119	26.93	1.004	27.58	8.6
MRD-3-2133	2.7917	0.56470	27.86	1.004	27.58	8.3
MRD-3-2134	2.6955	0.48776	26.98	1.003	27.73	8.5
MRD-3-2135	2.7074	0.49689	27.14	1.003	27.99	8.6
MRD-3-2141	2.6993	0.49577	27.00	1.004	27.48	8.5
MRD-3-2143	2.6842	0.47766	26.87	1.004	27.71	8.5
MRD-3-2144	2.6716	0.48329	26.79	1.002	27.49	8.1
MRD-3-2145	2.7547	0.52658	27.51	1.004	27.77	8.4
MRD-3-2152	2.6769	0.48274	26.85	1.005	28.01	8.3
MRD-3-2153	2.6735	0.48366	26.75	1.004	27.65	8.4
MRD-3-2154	2.6968	0.48527	27.00	1.004	27.68	8.5
MRD-3-2155	2.7345	0.50372	27.41	1.004	27.84	8.5
MRD-3-2161	2.7307	0.51091	27.34	1.004	27.55	8.4
MRD-3-2162	2.7142	0.50583	27.16	1.004	27.72	8.7
MRD-3-2163	2.6809	0.49363	26.82	1.004	27.68	8.7
MRD-3-2165	2.7109	0.50167	27.20	1.003	27.56	8.6

[1]NM = not measured

Table A-2. Contd.
Solution Concentrations, Corrected for Dilution

	Concentration, µg/L								
Sample ID:	Li	B	Na	Al	Si	Cl	K	I	Cs
MRD-3-2071	368	60	8964	6589	7193	8395	341	12	115
MRD-3-2072	288	48	9175	5734	8386	6010	308	9	106
MRD-3-2073	331	48	8060	6735	7602	7807	348	13	90
MRD-3-2075	330	62	8530	6183	6844	8379	316	12	97
MRD-3-2081	208	60	9490	4530	5271	6530	305	10	100
MRD-3-2083	281	43	8350	6322	6393	6450	226	14	84
MRD-3-2084	288	78	10016	6432	7114	6400	264	19	124
MRD-3-2085	271	71	9680	7200	6783	6070	283	15	116
MRD-3-2091	267	74	9762	6742	6646	6172	243	13	113
MRD-3-2092	249	92	9880	5843	5932	5492	218	12	81
MRD-3-2093	293	71	9005	6942	5868	5536	216	12	95
MRD-3-2094	293	84	11156	6535	6141	6074	284	13	103
MRD-3-2101	383	104	11942	8467	7398	7078	321	17	136
MRD-3-2102	265	96	11349	5548	5996	5626	260	19	111
MRD-3-2103	297	69	10383	7024	7294	5603	234	26	113
MRD-3-2104	254	89	11718	5384	5839	6323	313	14	126
MRD-3-2111	336	107	11148	7415	7281	5881	273	16	120
MRD-3-2112	307	125	12662	6512	6859	9340	324	18	109
MRD-3-2113	234	104	13355	5118	5898	10730	772	14	228
MRD-3-2115	237	96	9677	4953	5973	5860	243	12	104
MRD-3-2121	353	95	11172	8041	6867	6753	264	17	107
MRD-3-2122	362	99	12002	8500	6870	7768	482	17	172
MRD-3-2124	289	96	15976	6112	6480	11601	699	41	267
MRD-3-2125	217	73	10297	4750	5754	10245	331	15	146
MRD-3-2132	343	66	12385	7206	7175	7623	361	19	160
MRD-3-2133	379	105	24011	6015	5967	32000	1324	82	476
MRD-3-2134	267	82	12857	6046	6525	6807	293	27	112
MRD-3-2135	341	97	12503	7330	6475	7563	386	49	158
MRD-3-2141	244	104	11891	4929	5567	7203	361	27	144
MRD-3-2143	247	75	13400	5828	6267	7294	284	24	121
MRD-3-2144	291	103	12397	6041	6393	7147	424	24	147
MRD-3-2145	470	393	23651	4522	5202	34000	5541	63	1250
MRD-3-2152	358	62	12897	8146	6992	8156	459	32	147
MRD-3-2153	262	70	9853	6403	6291	6473	342	15	120
MRD-3-2154	325	107	18916	6623	6694	13836	849	30	346
MRD-3-2155	310	114	13399	7051	7359	8591	416	20	150
MRD-3-2161	259	120	13354	6893	7761	9875	781	16	288
MRD-3-2162	313	92	12721	8156	8480	7440	471	15	180
MRD-3-2163	263	95	12689	6657	7523	7095	395	15	135
MRD-3-2165	263	95	14952	6177	7153	9196	628	14	186
Blank1	0	0	0	0	0	0	0	0	0

Table A-2. Contd.
Concentrations in the Acid Strip Solutions

	Concentration, µg/L								
Sample ID:	Li	B	Na	Al	Si	Cl	K	I	Cs
AS-MRD-3-2071	0	0	27	14	0	NM	0	0.02	0.00
AS-MRD-3-2072	0	0	26	22	0	NM	0	0.02	0.00
AS-MRD-3-2073	0	0	32	45	0	NM	0	0.03	0.00
AS-MRD-3-2075	0	0	24	40	0	NM	0	0.03	0.00
AS-MRD-3-2081	1	6	0	20	43	NM	0	4.10	0.42
AS-MRD-3-2083	0	5	0	70	48	NM	0	2.50	0.11
AS-MRD-3-2084	0	3	24	47	51	NM	0	1.50	0.14
AS-MRD-3-2085	0	3	0	31	24	NM	0	0.50	0.07
AS-MRD-3-2091	0	3	0	51	66	NM	0	1.80	0.13
AS-MRD-3-2092	0	3	0	98	61	NM	0	0.50	0.13
AS-MRD-3-2093	0	3	0	62	75	NM	0	1.10	0.14
AS-MRD-3-2094	1	3	0	41	39	NM	13	1.10	0.46
AS-MRD-3-2101	0	2	0	60	27	NM	0	0.00	0.14
AS-MRD-3-2102	0	1	0	217	79	NM	0	0.70	0.20
AS-MRD-3-2103	0	2	0	119	43	NM	0	0.40	0.10
AS-MRD-3-2104	0	1	0	142	71	NM	0	0.00	0.13
AS-MRD-3-2111	0	2	0	100	81	NM	0	0.00	0.31
AS-MRD-3-2112	0	2	0	46	339	NM	0	0.00	0.14
AS-MRD-3-2113	0	1	0	72	60	NM	0	0.00	0.08
AS-MRD-3-2115	0	2	0	186	70	NM	0	0.00	0.10
AS-MRD-3-2121	0	2	0	101	86	NM	11	0.00	0.11
AS-MRD-3-2122	0	1	28	58	81	NM	29	0.00	0.14
AS-MRD-3-2124	0	0	0	60	68	NM	0	0.00	0.07
AS-MRD-3-2125	0	2	35	72	75	NM	14	0.00	0.37
AS-MRD-3-2132	0	2	0	80	54	NM	0	0.00	0.14
AS-MRD-3-2133	0	2	0	61	44	NM	0	0.00	0.20
AS-MRD-3-2134	0	1	0	46	43	NM	0	0.00	0.06
AS-MRD-3-2135	0	0	0	146	47	NM	0	0.00	0.11
AS-MRD-3-2141	0	0	51	125	98	NM	29	0.00	0.09
AS-MRD-3-2143	0	1	0	51	108	NM	0	0.00	0.07
AS-MRD-3-2144	0	0	0	58	0	NM	0	0.00	0.07
AS-MRD-3-2145	2	1	35	15	29	NM	14	0.00	0.09
AS-MRD-3-2152	0	0	0	40	34	NM	0	0.00	0.05
AS-MRD-3-2153	0	0	0	33	23	NM	0	0.00	0.05
AS-MRD-3-2154	0	1	0	26	36	NM	0	0.00	0.09
AS-MRD-3-2155	0	1	0	123	43	NM	83	0.00	0.07
AS-MRD-3-2161	0	1	24	79	51	NM	26	0.00	0.38
AS-MRD-3-2162	0	0	0	99	31	NM	0	0.00	0.09
AS-MRD-3-2163	0	0	0	191	93	NM	0	0.00	0.11
AS-MRD-3-2165	0	0	0	66	31	NM	0	0.00	0.07

Table A-2. Contd.
Normalized Mass Loss

	Normalized Mass Loss, g/m^2								
Sample ID:	Li	B	Na	Al	Si	Cl	K	I	Cs
MRD-3-2071	8.31	0.38	7.71	4.83	3.59	17.62	1.88	14.03	7.83
MRD-3-2072	6.20	0.29	7.54	4.02	4.00	12.62	1.62	9.58	6.87
MRD-3-2073	7.47	0.30	6.94	4.96	3.80	16.39	1.92	15.32	6.16
MRD-3-2075	7.29	0.39	7.18	4.45	3.34	17.59	1.70	13.74	6.47
MRD-3-2081	4.70	0.40	8.19	3.35	2.67	13.72	1.68	12.53	6.86
MRD-3-2083	6.36	0.29	7.21	4.70	3.24	13.56	1.25	18.10	5.75
MRD-3-2084	6.51	0.52	8.65	4.76	3.60	13.43	1.45	24.53	8.49
MRD-3-2085	6.14	0.48	8.36	5.32	3.42	12.77	1.56	19.40	7.95
MRD-3-2091	6.04	0.50	8.43	5.00	3.37	12.96	1.34	16.79	7.70
MRD-3-2092	5.64	0.62	8.53	4.37	3.01	11.55	1.20	15.52	5.56
MRD-3-2093	6.63	0.48	7.77	5.15	2.99	11.63	1.19	15.51	6.47
MRD-3-2094	6.63	0.56	9.63	4.84	3.11	12.76	1.57	16.80	7.08
MRD-3-2101	8.67	0.69	10.31	6.27	3.73	14.91	1.77	22.02	9.33
MRD-3-2102	5.99	0.64	9.80	4.24	3.05	11.82	1.43	24.55	7.62
MRD-3-2103	6.72	0.46	8.96	5.25	3.69	11.77	1.29	33.60	7.71
MRD-3-2104	5.75	0.60	10.12	4.06	2.97	13.30	1.73	18.10	8.62
MRD-3-2111	7.60	0.72	9.62	5.53	3.70	12.35	1.50	20.66	8.24
MRD-3-2112	6.95	0.84	10.93	4.82	3.62	19.64	1.79	23.28	7.48
MRD-3-2113	5.30	0.69	11.53	3.82	2.99	22.59	4.26	18.13	15.56
MRD-3-2115	5.36	0.64	8.35	3.78	3.04	12.31	1.34	15.50	7.14
MRD-3-2121	7.99	0.64	9.64	5.99	3.49	14.21	1.46	22.01	7.29
MRD-3-2122	8.20	0.66	10.36	6.29	3.49	16.32	2.66	21.97	11.79
MRD-3-2124	6.55	0.64	13.79	4.54	3.29	24.40	3.86	53.04	18.25
MRD-3-2125	4.90	0.49	8.89	3.55	2.93	21.52	1.83	19.37	10.00
MRD-3-2132	7.77	0.44	10.69	5.36	3.63	16.02	1.99	24.56	10.93
MRD-3-2133	8.57	0.70	20.73	4.47	3.02	67.32	7.30	106.10	32.53
MRD-3-2134	6.04	0.55	11.10	4.48	3.30	14.30	1.62	34.89	7.68
MRD-3-2135	7.72	0.65	10.79	5.50	3.28	15.89	2.13	63.30	10.79
MRD-3-2141	5.52	0.70	10.27	3.72	2.85	15.14	1.99	34.91	9.86
MRD-3-2143	5.59	0.50	11.57	4.32	3.20	15.34	1.57	31.04	8.25
MRD-3-2144	6.58	0.69	10.70	4.48	3.21	15.00	2.34	30.97	10.08
MRD-3-2145	10.64	2.62	20.42	3.34	2.63	71.51	30.57	81.49	85.38
MRD-3-2152	8.10	0.42	11.13	6.02	3.53	17.17	2.53	41.43	10.02
MRD-3-2153	5.93	0.47	8.51	4.73	3.17	13.61	1.89	19.40	8.20
MRD-3-2154	7.37	0.72	16.33	4.89	3.38	29.11	4.68	38.82	23.63
MRD-3-2155	7.02	0.76	11.57	5.27	3.72	18.06	2.30	25.86	10.25
MRD-3-2161	5.87	0.80	11.53	5.13	3.93	20.78	4.31	20.70	19.71
MRD-3-2162	7.09	0.62	10.98	6.07	4.28	15.64	2.60	19.39	12.27
MRD-3-2163	5.96	0.64	10.95	5.03	3.83	14.92	2.18	19.40	9.21
MRD-3-2165	5.95	0.64	12.91	4.59	3.61	19.32	3.46	18.09	12.68
Mean	**6.74**	**0.62**	**10.47**	**4.78**	**3.37**	**18.52**	**2.92**	**27.36**	**12.09**
Standard Deviation	**1.20**	**0.35**	**3.02**	**0.76**	**0.37**	**12.42**	**4.64**	**19.05**	**12.99**
RSD%[1]	17.8%	56.9%	28.9%	16.0%	11.0%	67.1%	159.1%	69.6%	1.07

[1]RSD% = Relative standard deviation (standard deviation/mean), in percent.

Table A-3. MCC-1 Test Results for Discs from Bottom and Middle Positions in 8 HIP Cans Test Data

Sample ID:	Surface Area, cm^2	Sample Mass, g	Water Volume, mL	Dilution Factor	Acid Strip Volume, mL	pH
MRD-3-2072[1]	2.9529	0.61629	28.18	1.002	26.84	9.02
MRD-3-2072-m	2.7511	0.51024	27.49	1.003	28.44	
MRD-3-2084	2.6720	0.47539	26.78	1.002	27.88	8.74
MRD-3-2084-b	2.7650	0.52169	27.63	1.004	28.39	
MRD-3-2094	2.7872	0.53128	27.93	1.003	27.44	8.75
MRD-3-2094-b	2.7152	0.50157	27.15	1.004	28.50	
MRD-3-2112	2.6770	0.49147	26.78	1.004	27.83	8.68
MRD-3-2112-b	2.6835	0.50267	26.96	1.003	28.36	
MRD-3-2121	2.6673	0.49584	26.73	1.005	27.90	8.60
MRD-3-2121-b	2.0645	0.46021	20.62	1.005	28.36	
MRD-3-2144	2.6716	0.48329	26.79	1.002	27.49	8.14
MRD-3-2144-m	2.7250	0.51014	27.36	1.004	28.54	
MRD-3-2155	2.7345	0.50372	27.41	1.004	27.84	8.48
MRD-3-2155-m	2.7052	0.49768	27.17	1.003	28.18	
MRD-3-2161	2.7307	0.51091	27.34	1.004	27.55	8.44
MRD-3-2161-m	2.7155	0.50237	27.23	1.008	28.34	

[1]MRD-3-xxxx specifies a 3-d MCC-1 test with a disc from the top of can xxxx. MRD-3-xxxx-m or -b specifies that the disc is from either the middle (-m) or the bottom (-b) of the can.

TABLE A-3. Contd.
Solution Concentrations, Corrected for Dilution

	Concentration, µg/L								
Sample ID:	Li	B	Na	Al	Si	Cl	K	I	Cs
MRD-3-2072	288	48	9175	5734	8386	6010	308	8.9	105.5
MRD-3-2072-m	234	82	10882	5911	7260	8980	259	19.1	96.0
MRD-3-2084	288	78	10016	6432	7114	6400	264	19.0	124.1
MRD-3-2084-b	322	63	11780	8527	7669	6400	296	30.1	109.8
MRD-3-2094	293	84	11156	6535	6141	6074	284	13.0	103.2
MRD-3-2094-b	318	78	12869	8318	7590	6070	279	25.1	99.1
MRD-3-2112	307	125	12662	6512	6859	9340	324	18.1	109.3
MRD-3-2112-b	270	71	13167	7016	7930	9340	358	21.1	127.8
MRD-3-2121	353	95	11172	8041	6867	6753	264	17.1	106.6
MRD-3-2121-b	300	64	11400	8271	10212	6880	222	15.1	106.6
MRD-3-2144	291	103	12397	6041	6393	7147	424	24.0	147.5
MRD-3-2144-m	229	99	11654	5587	6853	7270	445	17.1	228.4
MRD-3-2155	310	114	13399	7051	7359	8591	416	20.1	149.9
MRD-3-2155-m	321	123	19943	7051	7374	25000	3371	25.1	956.1
MRD-3-2161	259	120	13354	6893	7761	9875	781	16.1	288.2
MRD-3-2161-m	306	109	13454	7319	7284	9870	419	42.4	142.5

Table A-3. Contd.
Concentrations in Acid Strip Solutions

	Concentration, µg/L								
Sample ID:	Li	B	Na	Al	Si	Cl	K	I	Cs
AS-MRD-3-2072	0	0	26	22	0	NM	0	0.0	0.00
AS-MRD-3-2072-m	0	1	0	18	0	NM	0	0.0	0.04
AS-MRD-3-2084	0	3	24	47	51	NM	0	1.5	0.14
AS-MRD-3-2084-b	0	0	0	140	50	NM	0	0.0	0.03
AS-MRD-3-2094	1	3	0	41	39	NM	13	1.1	0.46
AS-MRD-3-2094-b	0	0	0	16	0	NM	0	0.0	0.02
AS-MRD-3-2112	0	2	0	46	339	NM	0	0.0	0.14
AS-MRD-3-2112-b	0	0	0	134	62	NM	0	0.0	0.04
AS-MRD-3-2121	0	2	0	101	86	NM	11	0.0	0.11
AS-MRD-3-2121-b	0	0	0	105	44	NM	0	0.0	0.04
AS-MRD-3-2144	0	0	0	58	0	NM	0	0.0	0.07
AS-MRD-3-2144-m	0	0	0	25	90	NM	0	0.0	0.09
AS-MRD-3-2155	0	1	0	123	43	NM	83	0.0	0.07
AS-MRD-3-2155-m	0	0	0	18	30	NM	0	0.0	0.13
AS-MRD-3-2161	0	1	24	79	51	NM	26	0.0	0.38
AS-MRD-3-2161-m	0	0	0	70	54	NM	0	0.0	0.06

Table A-3. Contd.
Normalized Mass Loss

	Normalized Mass Loss, g/m^2								
Sample ID:	Li	B	Na	Al	Si	Cl	K	I	Cs
MRD-3-2072	6.20	0.29	7.54	4.02	4.00	12.62	1.62	9.58	6.868
MRD-3-2072-m	5.29	0.55	9.39	4.36	3.65	18.86	1.43	24.55	6.56
MRD-3-2084	6.51	0.52	8.65	4.76	3.60	13.43	1.45	24.53	8.49
MRD-3-2084-b	7.30	0.42	10.17	6.37	3.88	13.46	1.63	38.80	7.50
MRD-3-2094	6.63	0.56	9.63	4.84	3.11	12.76	1.57	16.80	7.08
MRD-3-2094-b	7.21	0.52	11.11	6.13	3.81	12.77	1.54	32.35	6.77
MRD-3-2112	6.95	0.84	10.93	4.82	3.62	19.64	1.79	23.28	7.48
MRD-3-2112-b	6.11	0.48	11.37	5.26	4.02	19.62	1.98	27.13	8.73
MRD-3-2121	7.99	0.64	9.64	5.99	3.49	14.21	1.46	22.01	7.29
MRD-3-2121-b	6.78	0.43	9.84	6.16	5.15	14.49	1.23	19.43	7.28
MRD-3-2144	6.58	0.69	10.70	4.48	3.21	15.00	2.34	30.97	10.08
MRD-3-2144-m	5.18	0.66	10.06	4.13	3.49	15.29	2.45	21.99	15.60
MRD-3-2155	7.02	0.76	11.57	5.27	3.72	18.06	2.30	25.86	10.25
MRD-3-2155-m	7.27	0.82	17.22	5.20	3.72	52.55	18.60	32.32	65.31
MRD-3-2161	5.87	0.80	11.53	5.13	3.93	20.78	4.31	20.70	19.71
MRD-3-2161-m	6.92	0.73	11.61	5.43	3.69	20.85	2.31	54.57	9.74

APPENDIX B

Solution Results for PCT-A

Table B. PCT with Consolidated CWF and with CWF from Individual Cans
Test Data

Sample ID:	S/V, m^{-1}	Surface Area, cm^2	Sample Mass, g	Water Volume, mL	S/V, m^{-1}	Dilution Factor	Acid Strip Volume, mL	pH
Tests with consolidated CWF from 28 cans								
PRD-7-1	2300	462	2.01	20.10	2300	1.003	25.18	9.19
PRD-7-2	2299	460	2.00	20.01	2299	1.004	25.32	9.13
PRD-7-3	2302	230	1.00	9.99	2302	1.006	15.24	8.91
PRD-7-4	2300	230	1.00	10.00	2300	1.005	15.11	9.07
PRD-7-5	2302	460	2.00	19.98	2302	1.002	25.20	9.13
Tests in Stainless Steel 304L vessels with consolidated CWF from 28 cans								
PRD-A-mix-1-SS	2302	230	1.00	9.99	2302	1.004	14.74	9.10
PRD-A-mix-2-SS	2298	230	1.00	10.01	2298	1.003	15.23	8.91
PRD-A-mix-3-SS	2319	230	1.00	9.92	2319	1.005	9.42	8.39
PRD-A-mix-4-SS	2305	230	1.00	9.98	2305	1.006	10.02	8.88
Tests in Titanium vessels with consolidated CWF from 28 cans								
PRD-A-mix-1-Ti	2302	230	1.00	9.99	2302	1.004	14.96	9.13
PRD-A-mix-2-Ti	2298	230	1.00	10.01	2298	1.004	14.73	9.06
Tests with CWF from individual cans, represented by last 4 digits in Sample ID.								
PRD-A-2071	2307	231	1.00	10.00	2307	1.005	15.05	9.29
PRD-A-2085	2312	231	1.01	10.00	2312	1.005	14.40	9.10
PRD-A-2092	2302	230	1.00	10.01	2302	1.004	14.58	9.23
PRD-A-2104	2309	230	1.00	9.98	2309	1.007	12.97	9.14
PRD-A-2133	2298	230	1.00	10.00	2298	1.006	15.30	9.02
PRD-A-2134	2305	231	1.00	10.01	2305	1.006	14.74	9.06
PRD-A-2143	2302	230	1.00	9.99	2302	1.005	14.91	8.93
PRD-A-2145	2305	230	1.00	10.00	2305	1.002	14.45	9.02
PRD-A-2155	2302	230	1.00	9.99	2302	1.006	10.02	8.82
PRD-A-2161	2291	230	1.00	10.04	2291	1.005	10.03	8.53

Table B. Contd.
Solution Concentrations, Corrected for Dilution

	Concentration, µg/L								
Sample ID:	Li	B	Na	Al	Si	Cl	K	I	Cs
PRD-7-1	2513	3802	76358	7322	14051	119120	7553	116	3486
PRD-7-2	2617	3674	75122	7356	15150	129300	7707	125	4118
PRD-7-3	3949	3211	73213	10973	15689	138388	7634	147	3540
PRD-7-4	3182	3736	74253	9077	14494	144000	7318	127	3602
PRD-7-5	3813	3336	74675	10394	17725	129830	7446	158	4335
PRD-A-mix-1-SS	3302	4817	71884	8675	16741	131198	9209	134	3715
PRD-A-mix-2-SS	2880	3841	71749	8059	15244	133849	8289	142	4470
PRD-A-mix-3-SS	2136	6483	31276	6248	14950	NM	7413	235	3505
PRD-A-mix-4-SS	2071	7450	38229	6582	16209	NM	7737	223	3430
PRD-A-mix-1-Ti	3810	4888	71879	9722	16881	155020	8806	150	3388
PRD-A-mix-2-Ti	4218	4837	72429	11186	18180	148983	8909	159	3529
PRD-A-2071	3149	3316	35201	10988	16079	67230	4623	98	1829
PRD-A-2085	3840	3406	40681	12372	19268	74730	4727	108	2963
PRD-A-2092	3130	3561	37032	10764	19153	65340	4661	64	2939
PRD-A-2104	3421	4311	50195	10500	17375	86130	5785	90	2401
PRD-A-2133	4158	4228	56303	12269	20992	104330	6657	87	3471
PRD-A-2134	3244	5029	53997	9095	18359	99150	8306	62	3835
PRD-A-2143	3979	4656	56897	10975	20998	104330	8515	85	4252
PRD-A-2145	4530	6572	81819	8583	17142	162050	27995	110	4369
PRD-A-2155	2166	7011	40486	7397	16329	NM	9124	198	5005
PRD-A-2161	2113	7369	36219	7229	16267	NM	6546	226	3657

Table B. Contd.
Concentrations in the Acid Strip Solutions

	Concentration, µg/L								
Sample ID:	Li	B	Na	Al	Si	Cl	K	I	Cs
AS-PRD-7-1	0	NM	79	108	121	NM	0	0.2	0.6
AS-PRD-7-2	36	NM	881	1150	1291	NM	100	1.2	8.2
AS-PRD-7-3	9	NM	291	341	433	NM	28	0.7	3.8
AS-PRD-7-4	19	NM	644	645	858	NM	77	0.8	6.3
AS-PRD-7-5	9	NM	231	373	446	NM	28	0.9	3.0
AS-PRD-A-mix-1-SS	24	NM	609	1002	1093	NM	226	1.9	11.8
AS-PRD-A-mix-2-SS	41	NM	864	1476	1802	NM	143	2.0	18.9
AS-PRD-A-mix-3-SS	39	NM	1192	2003	2769	NM	138	1.8	26.6
AS-PRD-A-mix-4-SS	8	NM	291	1026	1463	NM	39	1.2	15.3
AS-PRD-A-mix-1-Ti	78	NM	1810	2612	2944	NM	283	2.6	19.6
AS-PRD-A-mix-2-Ti	36	NM	808	1580	1942	NM	138	2.4	16.1
AS-PRD-A-2071	20	NM	648	860	1202	NM	93	0.9	8.8
AS-PRD-A-2085	10	NM	227	669	927	NM	51	0.9	8.1
AS-PRD-A-2092	40	NM	2591	1346	1577	NM	2413	1.4	10.4
AS-PRD-A-2104	63	NM	1831	1830	2576	NM	762	1.3	18.2
AS-PRD-A-2133	15	NM	406	772	1089	NM	61	0.6	11.0
AS-PRD-A-2134	33	NM	1098	1219	1712	NM	328	1.3	13.9
AS-PRD-A-2143	53	NM	1549	1683	2424	NM	205	1.7	18.1
AS-PRD-A-2145	61	NM	1393	2045	2365	NM	351	2.0	21.4
AS-PRD-A-2155	9	NM	496	978	1468	NM	58	1.3	19.0
AS-PRD-A-2161	11	NM	330	982	1357	NM	35	1.3	17.0

Table B. Contd.
Normalized Mass Loss

	NL(i), g/m^2								
Sample ID:	Li	B	Na	Al	Si	Cl	K	I	Cs
PRD-7-1	0.25	0.11	0.29	0.024	0.031	1.08	0.18	0.52	1.04
PRD-7-2	0.26	0.11	0.29	0.028	0.037	1.18	0.19	0.57	1.23
PRD-7-3	0.39	0.09	0.28	0.037	0.036	1.26	0.18	0.66	1.05
PRD-7-4	0.32	0.11	0.28	0.032	0.034	1.31	0.18	0.57	1.07
PRD-7-5	0.38	0.10	0.28	0.035	0.040	1.18	0.18	0.71	1.29
Mean	0.32	0.10	0.28	0.031	0.035	1.20	0.18	0.61	1.13
Standard Deviation	0.06	0.01	0.00	0.005	0.003	0.09	0.00	0.08	0.11
RSD%	20.2%	7.4%	1.5%	16.6%	9.1%	7.3%	2.1%	12.9%	10.1%
PRD-A-mix-1-SS	0.33	0.14	0.27	0.032	0.040	1.19	0.23	0.61	1.11
PRD-A-mix-2-SS	0.29	0.11	0.27	0.033	0.039	1.22	0.20	0.65	1.34
PRD-A-mix-3-SS	0.21	0.19	0.12	0.026	0.038	1.27	0.18	1.05	1.04
PRD-A-mix-4-SS	0.20	0.22	0.14	0.024	0.038	1.28	0.19	1.00	1.02
Mean	0.26	0.16	0.20	0.029	0.039	1.24	0.20	0.83	1.13
Standard Deviation	0.06	0.05	0.08	0.004	0.001	0.04	0.02	0.23	0.15
RSD%	23.3%	28.6%	40.5%	15.5%	2.2%	3.3%	11.0%	28.1%	12.9%
PRD-A-mix-1-Ti	0.39	0.14	0.28	0.043	0.046	1.41	0.22	0.68	1.01
PRD-A-mix-2-Ti	0.42	0.14	0.27	0.043	0.045	1.36	0.22	0.72	1.06
Mean	0.40	0.14	0.28	0.043	0.046	1.39	0.22	0.70	1.03
Standard Deviation	0.02	0.00	0.00	0.000	0.000	0.04	0.00	0.03	0.03
RSD%	6.1%	0.6%	0.8%	0.5%	0.7%	2.6%	0.8%	4.1%	2.9%
PRD-A-2071	0.31	0.096	0.13	0.039	0.039	0.61	0.11	0.44	0.55
PRD-A-2085	0.38	0.098	0.15	0.042	0.045	0.68	0.11	0.48	0.88
PRD-A-2092	0.31	0.10	0.15	0.041	0.047	0.59	0.20	0.29	0.88
PRD-A-2104	0.34	0.12	0.20	0.041	0.045	0.78	0.16	0.40	0.72
PRD-A-2133	0.41	0.12	0.21	0.043	0.049	0.95	0.16	0.39	1.04
PRD-A-2134	0.32	0.15	0.21	0.035	0.045	0.90	0.21	0.28	1.14
PRD-A-2143	0.40	0.14	0.22	0.043	0.054	0.95	0.21	0.39	1.27
PRD-A-2145	0.45	0.19	0.31	0.037	0.045	1.47	0.68	0.50	1.30
PRD-A-2155	0.21	0.20	0.15	0.027	0.039	1.33	0.22	0.89	1.49
PRD-A-2161	0.21	0.21	0.14	0.026	0.039	1.23	0.16	1.02	1.10

[1]RSD = relative standard deviation (standard deviation/mean), in percent.

Distribution for ANL-00/12

Internal:

C. H. Adams
R. K. Ahluwalia
S. E. Aumeier
K. J. Bateman
T. J. Battisti
R. W. Benedict
A. R. Brunsvold
D. B. Chamberlain
J. C. Cunnane
S. S. Cunningham
K. B. Davies
W. L. Ebert (25)
R. E. Einziger
R. N. Elliot
R. A. Evans
T. A. Fanning
E. K. Fujita
H. E. Garcia
E. C. Gay
K. M. Goff
M. M. Goldberg
M. L. Gougar
C. Grandy
D. D. Hagan
J. E. Harmon
M. C. Hash
J. E. Herceg
K. L. Hirsche
S. G. Johnson
J. R. Kennedy
C. J. Knight
J. R. Krsul
C. E. Lahm
R. M. Lell
G. L. Lentz
D. Lewis
M. A. Lewis (25)
S. X. Li
R. P. Lind
J. S. Luo
R. D. Mariani
K. C. Marsden
S. D. McBride
S. M. McDeavitt
H. F. McFarlane
L. R. Morss
T. L. Moschetti
T. P. O'Holleran
R. L. Parks
D. R. Pedersen
R. D. Phipps
C. L. Pope
M. K. Richmann
R. H. Rigg
S. R. Sherman
M. F. Simpson
G. M. Teske
D. Vaden
D. L. Wahlquist
B. R. Westphal
R. A. Wigeland
J. L. Willit
T. L. Wright
V. N. Zyryanov
TIS Files

External:

DOE-OSTI (2)
ANL-E Library
ANL-W Library
A. Bindokas, DOE-CH
P. M. Ferrigan, DOE-CH
Chemical Technology Division Review Committee Members:
H. U. Anderson, University of Missouri-Rolla, Rolla, MO
A. L. Bement, Purdue University, West Lafayette, IN
C. Hussey, University of Mississippi, University, MS
M. V. Koch, University of Washington, Seattle, WA
V. P. Roan, University of Florida, Palm Beach Gardens, FL
J. R. Selman, Illinois Institute of Technology, Chicago, IL
J. S. Tulenko, University of Florida, Gainsville, FL
M. L. Stanley, El Cerrito, CA