

Secondary Waste Form Testing: Ceramicrete Phosphate Bonded Ceramics

Nuclear Engineering Division

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Secondary Waste Form Testing: Ceramicrete Phosphate Bonded Ceramics

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June 2011

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ACKNOWLEDGMENTS

The work at Argonne National Laboratory was supported by Inter-laboratory Work Order# 118877 from Pacific Northwest National Laboratory (PNNL) as part of the American Recovery and Reinvestment Act funding. Valuable insights and discussions with Mr. Joseph Westsik of PNNL are appreciated.

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ABBREVIATIONS

ANS	American Nuclear Society
B	Baseline
C1	Cluster 1
C2	Cluster 2
DOE	Department of Energy
ETF	Effluent Treatment Facility
HLW	High-Level Waste
ICP-MS	Induction Coupled Plasma – Mass Spectroscopy
IDF	Integrated Disposal Facility
IHLW	Immobilized High-Level Waste
ILAW	Immobilized Low Activity Waste
LAW	Low-Activity Waste
LI	Leachability Index
M	Mixed
ORP	Office of River Protection
PNNL	Pacific Northwest National Laboratory
RCRA	Resource Conservation and Recovery Act
SEM	Scanning Electron Microscopy
STU	Solidification Treatment Unit
TCLP	Toxicity Characteristic Leaching Procedure
UTS	Universal Treatment Standard
WRPS	Washington River Protection Solutions
WS	Waste Simulant
WTP	Waste Treatment and Immobilization Plant

EXECUTIVE SUMMARY

The cleanup activities of the Hanford tank wastes require stabilization and solidification of the secondary waste streams generated from the processing of the tank wastes. The treatment of these tank wastes to produce glass waste forms will generate secondary wastes, including routine solid wastes and liquid process effluents. Liquid wastes may include process condensates and scrubber/off-gas treatment liquids from the thermal waste treatment. The current baseline for solidification of the secondary wastes is a cement-based waste form. However, alternative secondary waste forms are being considered. In this regard, Ceramicrete technology, developed at Argonne National Laboratory, is being explored as an option to solidify and stabilize the secondary wastes.

The Ceramicrete process has been demonstrated on four secondary waste formulations: baseline, cluster 1, cluster 2, and mixed waste streams. Based on the recipes provided by Pacific Northwest National Laboratory, the four waste simulants were prepared in-house. Waste forms were fabricated with three filler materials: Class C fly ash, CaSiO_3 , and Class C fly ash + slag. Optimum waste loadings were as high as 20 wt.% for the fly ash and CaSiO_3 , and 15 wt.% for fly ash + slag filler.

Waste forms for physical characterizations were fabricated with no additives, hazardous contaminants, and radionuclide surrogates. Physical property characterizations (density, compressive strength, and 90-day water immersion test) showed that the waste forms were stable and durable. Compressive strengths were $>2,500$ psi, and the strengths remained high after the 90-day water immersion test. Fly ash and CaSiO_3 filler waste forms appeared to be superior to the waste forms with fly ash + slag as a filler. Waste form weight loss was ~ 5 -14 wt.% over the 90-day immersion test. The majority of the weight loss occurred during the initial phase of the immersion test, indicative of washing off of residual unreacted binder components from the waste form surface.

Waste forms for ANS 16.1 leach testing contained appropriate amounts of rhenium and iodine as radionuclide surrogates, along with the additives silver-loaded zeolite and tin chloride. The leachability index for Re was found to range from 7.9 to 9.0 for all the samples evaluated. Iodine was below detection limit (5 ppb) for all the leachate samples. Further, leaching of sodium was low, as indicated by the leachability index ranging from 7.6-10.4, indicative of chemical binding of the various chemical species. Target leachability indices for Re, I, and Na were 9, 11, and 6, respectively. Degradation was observed in some of the samples post 90-day ANS 16.1 tests.

Toxicity characteristic leaching procedure (TCLP) results showed that all the hazardous contaminants were contained in the waste, and the hazardous metal concentrations were below the Universal Treatment Standard limits.

Preliminary scale-up (2-gal waste forms) was conducted to demonstrate the scalability of the Ceramicrete process. Use of minimal amounts of boric acid as a set retarder was used to control the working time for the slurry.

Flexibility in treating waste streams with wide ranging compositional make-ups and ease of process scale-up are attractive attributes of Ceramicrete technology.

1 INTRODUCTION

The Hanford Site in southeast Washington State has 54 million gallons of radioactive and chemically hazardous wastes stored in 177 underground tanks. The U.S. Department of Energy, Office of River Protection (ORP), through its contractors, is constructing the Waste Treatment and Immobilization Plant (WTP) to convert the radioactive and hazardous wastes into stable glass waste forms for disposal. Within the WTP, the pretreatment facility will receive the retrieved waste from the tank farms and separate it into two treated process streams. The low-activity waste (LAW) stream is characterized as a high-volume, low-activity liquid process stream stripped of most solids and high-activity radioisotopes. The high-level waste (HLW) stream will be a much smaller volume slurry containing most of the solids, which have the high-activity isotopes, including ^{137}Cs and long-lived radioisotopes. The pretreated HLW mixture will route to the HLW Vitrification Facility, and the pretreated LAW stream will route to the LAW Vitrification Facility and to a supplemental treatment facility. The two WTP vitrification facilities will convert these process streams into glass, which is poured directly into stainless steel canisters. The immobilized HLW (IHLW) canisters will ultimately be disposed of off-site at a federal repository. The immobilized LAW (ILAW) canisters will be disposed of on-site in the Integrated Disposal Facility (IDF).

In addition to the primary IHLW and ILAW glass waste forms, the processing of the tank wastes will generate secondary wastes, including routine solid wastes and liquid process effluents. Liquid wastes may include process condensates and scrubber/off-gas treatment liquids from the thermal waste treatment processes. The liquid-effluent secondary wastes will be sent to the Effluent Treatment Facility (ETF) for further treatment and disposal, either as treated liquid effluent under the ETF State Wastewater Discharge Permit or as solidified liquid effluents under the Dangerous Waste Permit for disposal at the IDF. An additional solidification treatment unit has been proposed for the ETF. This new treatment unit would solidify the ETF evaporator brines into a cement-based waste form for disposal.

The ETF is multi-waste treatment and storage unit that can accept dangerous, low-level, and mixed wastewaters for treatment and is permitted by the Resource Conservation and Recovery Act (RCRA). The ETF receives, treats, and disposes of liquid effluents from cleanup projects on the Hanford Site. Plans are to increase the capacity of ETF to process the increased volume of secondary wastes when the WTP begins waste treatment and immobilization operations. A Solidification Treatment Unit (STU) will be added to the ETF to provide the needed additional capacity.

Washington River Protection Solutions (WRPS) has been chartered to move forward with the design and construction of the STU for ETF. The STU needs to be operational by 2018 to receive secondary liquid wastes from the WTP.

The current baseline calls for solidification of the ETF evaporator concentrate in a cement-based waste form. However, alternative secondary waste forms are being considered. In 2006, Pacific Northwest National Laboratory (PNNL) completed for DOE an evaluation of three low-temperature technologies for the immobilization of mixed radioactive and hazardous waste [1]. That testing program showed that Ceramcrete phosphate bonded ceramic has potential as a

waste form for the liquid secondary waste stream from WTP based on TCLP, compressive strength, and sodium leachability index requirements. However, the Ceramicrete's effectiveness in immobilizing ^{99}Tc and ^{129}I required additional testing. Immobilization of ^{99}Tc is of principal concern, whereas, containment of ^{129}I is a secondary issue. Further, immersion of the waste form in water over a period of time showed formation of white precipitate on the waste form surface.

Therefore, the focus of the current work was to develop additional information regarding the Ceramicrete phosphate bonded ceramic as a waste form for liquid secondary waste to support selection of a final waste form for the WTP secondary liquid wastes to be disposed in the IDF. Testing focused on resolving the formulation issues identified in the 2006 study and optimizing waste loading and evaluating the robustness of the Ceramicrete waste form to waste variability.

The specific objectives of this laboratory-scale work were three-fold:

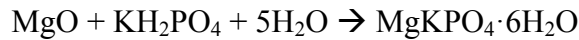
- (a) develop and optimize Ceramicrete binder formulations to stabilize and encapsulate four secondary waste stream formulations,
- (b) conduct physical characterizations on the waste forms, and
- (c) evaluate the waste forms for leaching of the contaminants

As a result of this study, waste form compositions, loadings, and performances have been optimized. Results from this study will form the basis of an engineering-scale demonstration on stabilization of secondary waste streams using Ceramicrete technology.

2 CERAMICRETE PHOSPHATE BONDED CERAMIC

Ceramicrete, a low-temperature forming phosphate ceramic, was developed at Argonne National Laboratory as part of DOE's Environmental Management program to stabilize and contain radioactive and hazardous contaminants such as Tc, Cs, and Hg that can volatilize during a high temperature immobilization process. In previous work, Ceramicrete technology has been demonstrated on various waste streams (liquids, fly ashes, and debris) and has been shown to successfully contain both radioactive (U, Tc, Pu) and hazardous contaminants (Hg, Pb, Cr, etc.) [2-6].

Ceramicrete is fabricated by acid/base reaction of magnesium oxide and mono-potassium phosphate, which when mixed with water forms a slurry that sets into a hard ceramic in a few hours. The process is simple and quite similar to the Portland cement process and easily scalable, as shown schematically in Fig. 2-1. No additional equipment requirements are needed. Ceramicrete is a strong (as high as 10,000 psi compressive strength), but dense matrix, and has superior ability to bind contaminants, making it an excellent candidate for microencapsulation. The chemical reaction for Ceramicrete formation can be represented as [2-4]:



The resulting $\text{MgKPO}_4 \cdot 6\text{H}_2\text{O}$ phase is extremely stable and has a solubility product of 2×10^{-11} under ambient conditions. There is no residual water in the system; it is bound as the water of hydration. Ceramicrete has a unique property of binding to itself, unlike cement concrete. Thus, Ceramicrete is easy to repair or patch.

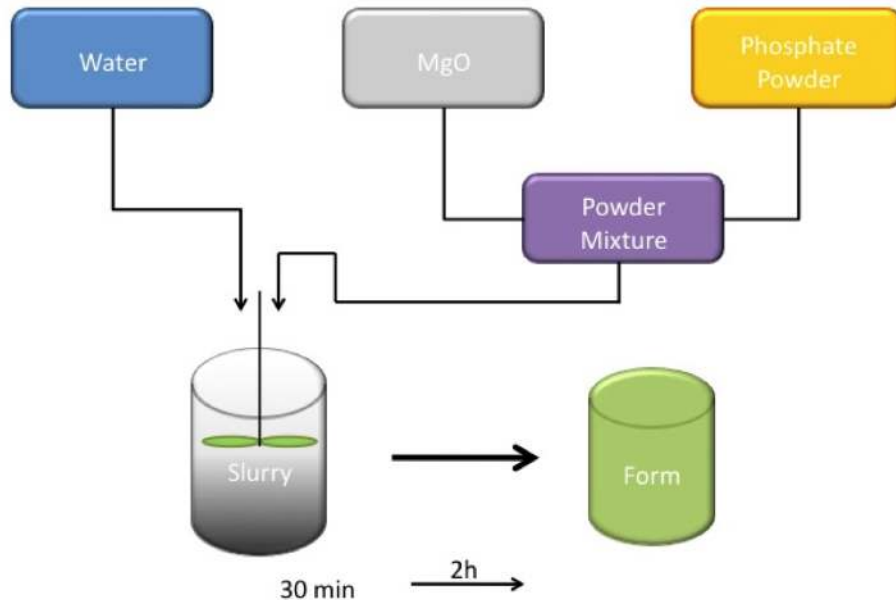


Figure 2-1. Schematic of Ceramicrete process

Because of the flexibility of this process, various second phase materials may be added for higher strength, fracture toughness, and reduced porosity. For example, one can add as much as 80 wt.% fly ash to Ceramicrete dry powder mix. There is no specific requirement of the particle size of the fly ash. In addition to the phosphate bonding, silico-phosphate bonding that provides enhanced structural properties may also occur in Ceramicrete system [3].

3 SAMPLE PREPARATION

3.1 Waste Simulant Formulations

Four waste stream formulations with typical WTP secondary waste effluents were provided by PNNL. The compositional makeups of the four streams are listed in Table 3-1. These streams are the baseline caustic scrubber, low salt, high salt, and mixed simulant (caustic scrubber stream blended with 10% of submerged bed scrubber waste stream). The baseline, cluster 1, cluster 2, and mixed simulant are referred to as “B”, “C1”, “C2”, and “M”, respectively. Since Argonne’s effort was on nonradioactive waste streams, surrogates to simulate radioactive ^{99}Tc and ^{129}I were used. To simulate ^{99}Tc , rhenium surrogate was used in ReO_4^- form. For ^{129}I , NaI compound was used. It should be noted that use of “water stream” and “waste simulant” are used interchangeably throughout the text.

The four waste streams were prepared in-house by using analytical grade chemicals procured from commercial suppliers. Formulations were prepared for the four waste streams with and without RCRA metals. Waste streams without RCRA metals were used in conducting various scoping tests such as density, compressive strength, and water immersion test, etc. The pH values for the waste simulants for scoping studies were 12.20, 12.45, 12.33, and 8.62 for baseline, cluster 1, cluster 2, and mixed simulants, respectively. Densities of the waste streams were measured to be about 1.04 g/cc. Waste streams with RCRA metals were used in fabricating waste forms that underwent the leach tests such as the TCLP [7]. Waste forms tested for leaching per the American Nuclear Society (ANS) 16.1 procedure [8] contained iodine and rhenium to simulate ^{129}I and ^{99}Tc , respectively.

Table 3-2 lists the weights of each chemical ingredient added to 3.5 L of water to produce the four waste streams. All waste streams were at 1 M sodium. Also, Table 3-2 lists the sequence of the additions of the various chemical species followed to produce the simulants. As indicated above, the waste stream formulations in Table 3-2 do not include the surrogates for radionuclides (Re and I) or any hazardous (RCRA) metals. Those additions were made to the waste streams for samples prepared for leaching tests only, as also discussed above.

Figure 3-1 shows the typical baseline simulant made in the laboratory. Other simulants also appeared quite similar. The fluid looks turbid. However, if the simulant is not disturbed, with time there is some settling of the suspended particles, as shown in the Figure 3-2 for the cluster 1 simulant. Thus, thorough mixing of the simulant was essential before using it to prepare the waste forms.

Table 3-1. Secondary waste simulant compositions (provided by PNNL)

	Concentration (moles/liter)			
	Baseline Caustic Scrubber, Medians	Cluster 1 (low NO ₃ ⁻ & Cl ⁻) model operating month 3/16/2038	Cluster 2 (high NO ₃ ⁻ & Cl ⁻) model operating month 05/28/2024	Mixed Caustic Scrubber /10% of SBS Blend
Na	1	1	1	1
Al(OH) ₃	9.39E-02	1.14E-01	9.22E-02	4.24E-02
Si	1.88E-03	2.04E-03	7.74E-04	1.39E-02
K	5.82E-04	6.51E-04	2.18E-03	2.87E-02
NH ⁴⁺	---	---	---	4.41E-01
OH ⁻	3.98E-01	4.35E-01	2.45E-01	1.02E-08
NO ₃ ⁻	3.28E-01	1.90E-01	3.97E-01	1.13E+00
CO ₃ ²⁻	2.28E-02	4.66E-02	3.94E-02	1.04E-02
Cl ⁻	2.25E-02	2.17E-02	2.91E-02	1.04E-02
NO ₂ ⁻	1.20E-02	1.05E-02	3.83E-02	4.31E-02
PO ₄ ³⁻	6.87E-03	4.85E-03	6.03E-03	5.10E-03
SO ₄ ²⁻	4.41E-03	5.81E-03	5.14E-03	4.36E-02
F ⁻	5.57E-04	3.75E-04	4.42E-04	1.02E-08
Cr	2.03E-04	2.03E-04	2.03E-04	1.09E-03
Ag	6.27E-06	6.27E-06	6.27E-06	2.35E-05
As	3.48E-05	3.48E-05	3.48E-05	1.61E-05
Cd	1.57E-06	1.57E-06	1.57E-06	2.16E-05
Hg	1.13E-05	1.13E-05	1.13E-05	5.30E-06
Pb	8.99E-06	8.99E-06	8.99E-06	8.28E-06
Tc	1.81E-05	1.81E-05	1.81E-05	5.59E-04
⁹⁹ Tc ^(a)	3.05E-05	3.05E-05	3.05E-05	9.41E-04
I	4.62E-06	4.62E-06	4.62E-06	6.29E-05
¹²⁹ I ^(a)	9.53E-08	9.53E-08	9.53E-08	1.30E-06
Total organic carbon ^(b)	9.39E-02	1.14E-01	9.22E-02	4.24E-02

^{a.} Ci/liter.

^{b.} As oxalate.

3.2 Ceramicrete Raw Materials

Base ingredients to produce Ceramicrete are magnesium oxide, potassium phosphate, and water. In addition to these, a filler material is added. Three types of fillers were used in the study: Class C fly ash, calcium silicate (wollastonite), and a mixture of blast furnace slag (slag) and Class C fly ash. The blast furnace slag was attempted since it has been shown in literature that because of its reducing characteristic it can possibly help in keeping Tc-99 (or Re) in its reduced relatively insoluble form and also helps modifying the pore structure of the cement matrix [9]. Similarly, silver-loaded zeolite has been demonstrated to capture and contain iodine and was therefore used as one of the additives in the present study [1]. Further, small amounts of reducing agents, such as tin chloride and potassium sulfide, were added as needed for stabilization of ⁹⁹Tc (or Re) and mercury, respectively. The raw ingredients and their procured sources are listed in Table 3-3.

Table 3-2. Waste simulant formulations for ANS 16.1 and waste form characterization tests (added to 3.5 L of water)

	Baseline (B) (g)	B Add Order ^a	Cluster 1 (C1) (g)	C1 Add Order	Cluster 2 (C2) (g)	C2 Add Order	Mixed (M) (g)	M Add Order
Al(OH) ₃	25.6359	12	31.1235	12	25.1718	13	11.5757	11
Na ₂ SiO ₃	0.8032	1	0.8715	1	0.3307	1	5.9384	1
KNO ₃	0.2059	2	0.2304	2	0.7714	2	10.1558	2
NH ₄ NO ₃	0		0		0		93.0105	3
NaOH	55.716	10	55.716	10	55.716	10	0	
NaNO ₃	97.574	3	56.2241	3	117.5053	3	228.7635	4
Na ₂ CO ₃	1.8548	4	17.2867	4	0		0	
NaHCO ₃	0		0		11.5845	4	0	
(NH ₄) ₂ CO ₃	0		0		0		3.4975	5
NaCl	4.6024	5	4.4387	5	5.9524	5	2.1273	6
NaNO ₂	2.8978	6	2.5356	6	9.2488	6	10.4079	7
NaH ₂ PO ₄ ·2H ₂ O	3.7512	7	0		3.2925	7	0	
Na ₃ PO ₄ ·12H ₂ O	0		6.4526	7	0		6.7852	8
Na ₂ SO ₄	2.1924	8	2.8884	8	2.5553	8	0	
(NH ₄) ₂ SO ₄	0		0		0		20.1646	9
NaF	0.0819	9	0.0551	9	0.065	9	0	
H ₂ C ₂ O ₄	0		0		16.1343	11	0	
Na ₂ C ₂ O ₄	44.0387	11	53.4655	11	19.2288	12	19.8854	10
Water	3500		3500		3500		3500	
Total Mass	3739.3542		3731.2881		3767.5568		3912.3118	

^a Add order indicates the sequence the chemicals were added.

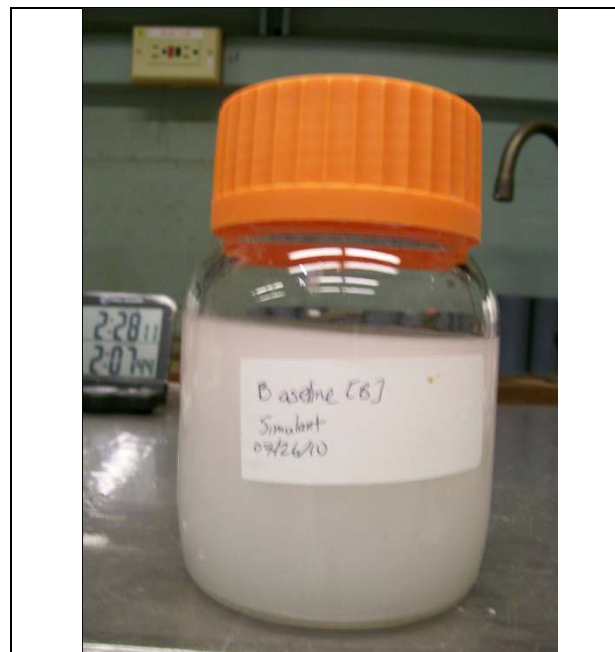


Figure 3-1. As-prepared baseline waste simulant



Figure 3-2. Settling of suspended particulates with time in cluster 1 waste simulant

Table 3-3. Materials and suppliers used in laboratory testing

Material	Supplier
MgO (magnesium oxide)	
MgO P98	Martin Marietta Magnesia Specialties, Baltimore, MD
MKP (mono potassium phosphate)	
MKP-771	Bindan Corp., Oakbrook, IL ICL Performance Products, Saint Louis, MO
Powder Filler	
Class C fly ash	La Farge, Chicago, IL
Calcium silicate (wollastonite)	NYCO, Willsboro, NY
Blast furnace slag	La Farge, Chicago, IL
Additives	
Boric acid, technical grade, H ₃ BO ₃	Fisher Scientific, Pittsburgh, PA
Tin chloride, SnCl ₂ , 98% Reagent grade	Sigma Aldrich, Saint Louis, MO
Ag-loaded zeolite (Ionex Type Ag 400)	Molecular Products, Golden, CO
Potassium sulfide (I-5130)	Chem Service, Westchester, PA
Water	ANL laboratory tap water

3.3 Waste Form Fabrication

3.3.1 Waste Forms for Physical Characterizations and ANS 16.1 Leaching Test

Waste forms were fabricated by first placing the requisite amount of waste stream in a mixing bowl. It should be noted that for ANS 16.1 leach test samples, waste stream compositions listed in Table 3-2 were spiked with appropriate amounts of NaReO_4 and NaI such that concentrations of Re and I in the waste streams were 1.81×10^{-5} g/L and 4.62×10^{-6} g/L, respectively. A pre-weighed amount of tin chloride was added and mixed for 3-4 minutes. Then, silver-loaded zeolite beads were added to the mix and stirred for approximately 3 minutes. Additional water was added to the mix, followed by the premixed and weighed powder of MgO , MKP-771, and filler. In some instances (for waste loadings > 20 wt.%), a small amount of set retarder such as boric acid (1 wt.% on dry powder basis) was also added. The mixture was typically mixed for 15-20 minutes to produce a slurry (Fig. 3-3). This slurry was poured into plastic syringe molds (0.5-in. diameter x 4 in. long) for curing (Fig. 3-4). Samples were left for curing for at least 2-3 weeks before extracting them out of the molds for evaluation. Optimum time for full cure of the waste form was not determined as part of this study. Laboratory-scale waste forms were fabricated at batch size of ~200-250 cc. For the waste forms fabricated for physical characterizations, tin chloride and zeolite additions were skipped because no radionuclide simulants were used in the waste streams.

Tables 3-4 to 3-7 show the composition of the various waste forms fabricated for the four waste streams. Waste loading is presented in weight percent (wt. %) and is defined as on the basis of additions made in preparing the slurry:

$$\frac{(\text{weight of waste simulant})}{(\text{weight of waste simulant} + \text{weight of all other ingredients})} \times 100$$

For fly ash as a filler, waste loadings were 5-25 wt.%; for CaSiO_3 filler, waste loading were 15 and 20 wt.%; and for slag + fly ash filler, waste loadings were 10 and 15 wt.%. In addition, samples evaluated per ANS 16.1 leaching were fabricated with silver zeolite and tin chloride additions, whereas samples for physical property characterizations did not have these additives. For high waste loading (25 wt.%) with fly ash filler, 1 wt.% (on dry powder basis) of boric acid was added as a set retarder. The total amount of waste simulant and water added was approximately constant. Any additional water added was qualitative and based on achieving the desired consistency of the slurry.

The nomenclature followed in naming the various formulations in Tables 3-4 to 3-7 is in terms of the filler (fly ash:FA, CaSiO_3 : CaSi, and slag: Slg) weight percent in dry powders and waste loading (WS) on wet basis. For example, "25FA + 25WS + 1BA" translates to 25% fly ash by dry weight basis, 25% waste simulant by wet basis, and 1% boric acid on dry basis.

During the fabrication of the waste form samples, the goal was to mix the slurry for approximately 20 minutes. During the mixing sequence, the temperature of the slurry was also monitored. The typical temperature rise of the slurry over 20 minutes of mixing was 40°C for the 200-250 cc batches. For the CaSiO_3 filler, the temperature rise was faster than that of fly ash



Figure 3-3. Slurry of Ceramicrete mix with Class C fly ash and 20 wt.% baseline waste simulant

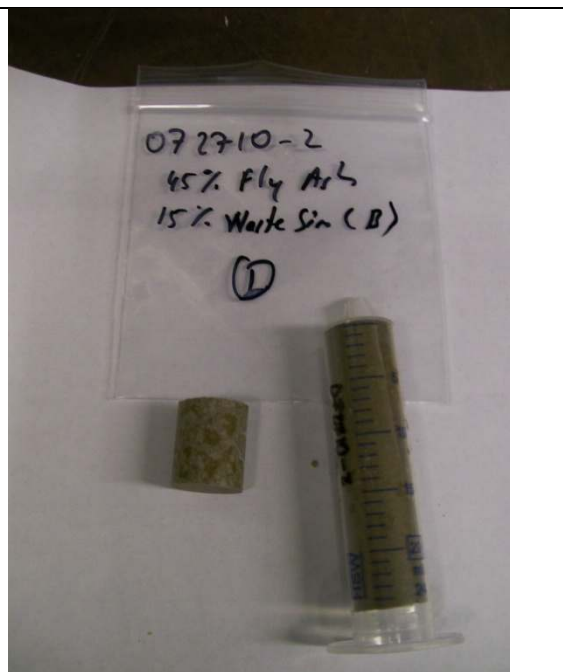


Figure 3-4. Ceramicrete waste form for Class C fly ash and 15 wt.% baseline waste simulant

Table 3-4. Waste form compositions (wt.%) with baseline waste simulant

	MgO	MKP	Waste Stream	Fly Ash	CaSiO ₃	Slag	Boric Acid	Silver Zeolite ^a	SnCl ₂ ^a	Water
FA Blank	9.5	32.4		34.3				1.1	1.3	21.4
45FA+20WS	9.5	32.4	19.6	34.3				1.1	1.3	1.8
45FA+15WS	9.5	32.4	14.7	34.3				1.1	1.3	6.8
45FA+10WS	9.8	33.2	10.0	35.1						11.9
45FA+5WS	9.8	33.2	5.0	35.1						16.9
25FA+25WS+1BA	12.3	41.6	24.8	18.0			0.73			2
CaSi Blank	9.5	32.4			34.3			1.1	1.3	21.4
45CaSi+20WS	9.5	32.4	19.6		34.3			1.1	1.3	1.8
45CaSi+15WS	9.5	32.4	14.7		34.3			1.1	1.3	6.8
Slag Blank	9.5	32.4		26.7		7.6		1.1	1.3	21.4
35FA+10Slg+10WS	9.8	33.3	10.1	27.4		7.9				11.9
35FA+10Slag+15WS	9.5	32.4	14.7	26.7		7.6		1.1	1.3	6.8

^a Not added for samples made for physical property evaluations.

Table 3-5. Waste form compositions (wt.%) with cluster 1 waste simulant

	MgO	MKP	Waste Stream	Fly Ash	CaSiO ₃	Slag	Boric Acid	Silver Zeolite ^a	SnCl ₂ ^a	Water
FA Blank	9.5	32.4		34.3				1.1	1.3	21.4
45FA+20WS	9.5	32.4	19.6	34.3				1.1	1.3	1.8
45FA+15WS	9.5	32.4	14.7	34.3				1.1	1.3	6.8
45FA+10WS	9.8	33.2	10.0	35.1						11.9
45FA+5WS	9.8	33.2	5.0	35.1						16.9
25FA+25WS+1BA	12.3	41.6	24.8	18.0			0.73			2
CaSi Blank	9.5	32.4			34.3			1.1	1.3	21.4
45CaSi+20WS	9.5	32.4	19.59		34.3			1.1	1.3	1.8
45CaSi+15WS	9.5	32.4	14.66		34.3			1.1	1.3	6.8
Slag Blank	9.5	32.4		26.7		7.6		1.1	1.3	21.4
35FA+10Slg+10WS	9.8	33.3	10.1	27.4		7.9				11.9
35FA+10Slag+15WS	9.5	32.4	14.66	26.7		7.6		1.1	1.3	6.8

^a. Not added for samples made for physical property evaluations.

Table 3-6. Waste form compositions (wt.%) with cluster 2 waste simulant

	MgO	MKP	Waste Stream	Fly Ash	CaSiO ₃	Slag	Boric Acid	Silver Zeolite ^a	SnCl ₂ ^a	Water
FA Blank	9.5	32.4		34.3				1.1	1.3	21.4
45FA+20WS	9.5	32.4	19.6	34.3				1.1	1.3	1.8
45FA+15WS	9.5	32.4	14.7	34.3				1.1	1.3	6.8
45FA+10WS	9.8	33.2	10.0	35.1						11.9
45FA+5WS	9.8	33.2	5.0	35.1						16.9
25FA+25WS+1BA	12.3	41.6	24.8	18.0			0.73			2
CaSi Blank	9.5	32.4			34.3			1.1	1.3	21.4
45CaSi+20WS	9.5	32.4	19.59		34.3			1.1	1.3	1.8
45CaSi+15WS	9.5	32.4	14.66		34.3			1.1	1.3	6.8
Slag Blank	9.5	32.4		26.7		7.6		1.1	1.3	21.4
35FA+10Slg+10WS	9.8	33.3	10.1	27.4		7.9				11.9
35FA+10Slag+15WS	9.5	32.4	14.66	26.7		7.6		1.1	1.3	6.8

^a. Not added for samples made for physical property evaluations.

Table 3-7. Waste form compositions (wt.%) with mixed waste simulant

	MgO	MKP	Waste Stream	Fly Ash	CaSiO ₃	Slag	Boric Acid	Silver Zeolite ^a	SnCl ₂ ^a	Water
FA Blank	9.5	32.4		34.3				1.1	1.3	21.4
45FA+20WS	9.5	32.4	19.6	34.3				1.1	1.3	1.8
45FA+15WS	9.5	32.4	14.7	34.3				1.1	1.3	6.8
45FA+10WS	9.8	33.2	10.0	35.1						11.9
45FA+5WS	9.8	33.2	5.0	35.1						16.9
25FA+25WS+1BA	12.3	41.6	24.8	18.0			0.73			2
CaSi Blank	9.5	32.4			34.3			1.1	1.3	21.4
45CaSi+20WS	9.5	32.4	19.59		34.3			1.1	1.3	1.8
45CaSi+15WS	9.5	32.4	14.66		34.3			1.1	1.3	6.8
Slag Blank	9.5	32.4		26.7		7.6		1.1	1.3	21.4
35FA+10Slg+10WS	9.8	33.3	10.1	27.4		7.9				11.9
35FA+10Slag+15WS	9.5	32.4	14.66	26.7		7.6		1.1	1.3	6.8

^a Not added for samples made for physical property evaluations.

filler. This resulted in the faster thickening of the slurry; hence, the mixing was done for 10-15 minutes for these samples.

3.3.2 Waste Forms for Toxicity Characteristic Leaching Procedure

The TCLP was conducted on waste forms fabricated with hazardous elements in the waste streams, as per the instructions provided in the waste stream formulations [email from J. Westsik, July 2010]. Waste streams were fabricated with the requisite amounts of hazardous contaminants added as per the sequence identified in Table 3-8. After the waste forms were cured for 2-3 weeks, samples were extracted from plastic molds and shipped to GEL Laboratories (Charleston, South Carolina) for testing. GEL labs crushed the samples as per the standard to perform the TCLP tests. For these samples, no Ag-zeolite was added. Further, the hazardous contaminants were only Ag, Cr, As, Cr, Hg, and Pb. However, for mercury stabilization, 0.2 wt.% (of the dry powder mix) of potassium sulfide was added to the waste simulant and mixed until it dissolved.

Table 3-8. Waste stimulant formulations for TCLP tests (added to 3.5 L of water)

Chemical	Baseline (B) (g)	B Add Order	Cluster 1 (C1) (g)	C1 Add Order	Cluster 2 (C2) (g)	C2 Add Order	Mixed (M) (g)	M Add Order
Al(OH) ₃	25.6359	20	31.1235	20	25.1718	21	11.5757	19
Na ₂ SiO ₃	0.8032	1	0.8715	1	0.3307	1	5.9384	1
KNO ₃	0.2059	2	0.2304	2	0.7714	2	10.1558	2
NH ₄ NO ₃	0		0		0		93.0105	3
NaOH	55.716	18	55.716	17	55.716	18	0	
NaNO ₃	97.574	3	56.2241	3	117.5053	3	228.7635	4
Na ₂ CO ₃	1.8548	9	17.2867	9	0		0	
NaHCO ₃	0		0		11.5845	9	0	
(NH ₄) ₂ CO ₃	0		0		0		3.4975	10
NaCl	4.6024	10	4.4387	10	5.9524	10	2.1273	11
NaNO ₂	2.8978	11	2.5356	11	9.2488	11	10.4079	12
NaH ₂ PO ₄ ·2H ₂ O	3.7512	12	0		3.2925	12	0	
Na ₃ PO ₄ ·12H ₂ O	0		6.4526	12	0		6.7852	13
Na ₂ SO ₄	2.1924	13	2.8884	13	2.5553	13	0	
(NH ₄) ₂ SO ₄	0		0		0		20.1646	14
NaF	0.0819	14	0.0551	14	0.065	14	0	
Na ₂ Cr ₂ O ₇ ·2H ₂ O	0.1059	15	0.1059	15	0.1059	15	0.5684	15
AgNO ₃	0.0037	4	0.0037	4	0.0037	4	0.014	5
Na ₂ HAsO ₄ ·7H ₂ O	0.038	5	0.038	5	0.038	5	0.0176	6
Cd(NO ₃) ₂ ·4H ₂ O	0.0017	6	0.0017	6	0.0017	6	0.0233	7
Hg(NO ₃) ₂ ·H ₂ O	0.0136	7	0.0136	7	0.0136	7	0.0064	8
Pb(NO ₃) ₂	0.0104	8	0.0104	8	0.0104	8	0.0096	9
NaI	0.0024	16	0.0024	16	0.0063	16	0.033	16
H ₂ C ₂ O ₄	0		0		16.1343	19	0	
Na ₂ C ₂ O ₄	44.0387	19	53.4655	19	19.2288	20	19.8854	18
NaReO ₄	0.0173	17	0.0173	18	0.0173	17	0	

4 PHYSICAL PROPERTY CHARACTERIZATIONS

4.1 Expansion Evaluation

After curing of the blank and the waste form samples, they were removed from the plastic syringe molds and sliced into appropriate lengths for various evaluations. Figures 4-1 and 4-2 show a typical sample with Class C fly ash and CaSiO_3 fillers with cluster 1 and baseline waste streams. After the slurry was poured into the syringe, a mark was made to keep track of any expansion that occurs during the two-week curing. After at least two weeks of curing, the samples were removed from the plastic syringes and cut in appropriate sizes (approximately 1 inch in length) for subsequent characterizations.

Figures 4-1 and 4-2 indicate no discernible expansion in the waste forms after setting. The black line on the syringe indicates the level of the slurry after pouring. The set product was at the same length, indicating no expansion. This comparison was performed for all the waste forms fabricated.

Most of the other samples exhibited no expansion or shrinkage. There was no evidence of any segregation. However, there was expansion (1-10%) for some formulations, particularly the cluster 2 waste simulant at high waste loading (25 wt.%) fabricated with 25 wt.% Class C fly ash filler and the slag + ash filler at both 10 and 15 wt.% loadings. Interestingly, the 25 wt.% CaSiO_3 filler with 25 wt.% cluster 2 waste simulant did not expand. Also, cluster 2 waste simulant fabricated with ≤ 20 wt.% loading and 45 wt.% Class C fly ash showed no discernible expansion. It appears that high waste loadings (>20 wt.%) and addition of 1 wt.% boric acid lead to slight expansion. It should be noted that unlike other waste simulants, cluster 2 has sodium

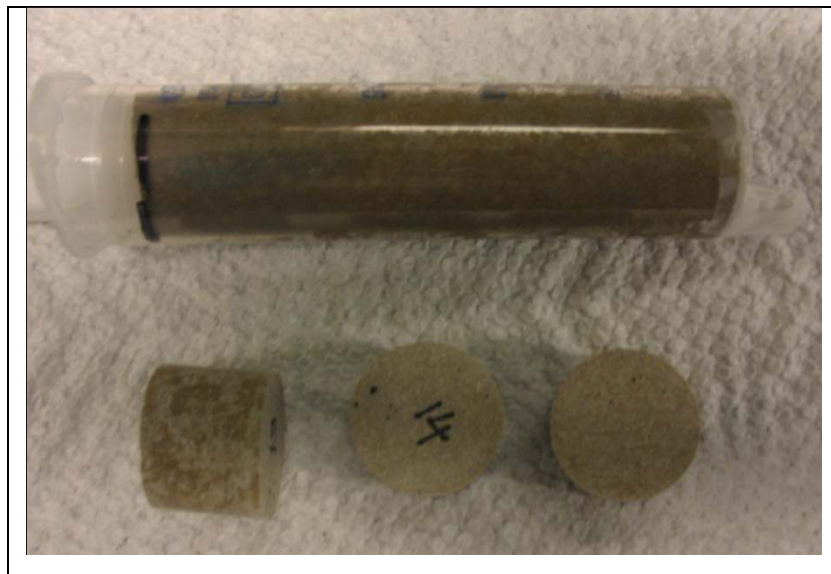


Figure 4-1. As-fabricated waste form for 45 wt.% Class C fly ash and 15 wt.% cluster 1 waste simulant after setting



Figure 4-2. As-fabricated waste form for 45 wt.% CaSiO_3 and 20 wt.% baseline waste simulant

bicarbonate that probably breaks down in the presence of an acid (phosphate or boric acid) and releases CO_2 that leads to the expansion. For future work and scale-up activity, it may be prudent to breakdown the bicarbonate/carbonates in the waste stream prior to making the waste form.

4.2 Density

Densities of the fabricated samples were calculated from the ratio of the sample weight and its volume. The volume of each sample was calculated from measurements of sample diameter and length. Typically, 4-5 samples of each formulation were used to determine the average density. Densities were determined for waste forms fabricated using plain water (Table 4-1), the baseline waste simulant (Table 4-2), cluster 1 simulant (Table 4-3), cluster 2 simulant (Table 4-4), and mixed waste simulant (Table 4-5). Samples at 25 wt.% loading and 25 wt.% fly ash filler and boric acid additions had lower density because of the expansion in the waste form, as discussed in the previous section.

Table 4-1. Density of waste forms fabricated using plain water

Sample Composition	Average Density (kg/m ³)
45FA Blank	1951 ± 94
45CaSi Blank	2032 ± 25
25CaSi Blank	1897 ± 16
25FA Blank	1816 ± 82
35FA+10Slg Blank	1995 ± 20

Table 4-2. Density of waste forms fabricated using baseline waste simulant

Sample Composition	Average Density (kg/m ³)
45FA+5W	1951 ± 24
45FA+10W	1959 ± 21
45FA+15W	1973 ± 14
45FA+20W	1978 ± 19
45CaSi+15W	2052 ± 16
45CaSi+20W	2067 ± 14
25CaSi+25W	1951 ± 95
25FA+25W	1875 ± 23
25FA+25W+1BA	1838 ± 57
35FA+10Slg+10W	1978 ± 49
35FA+10Slg+15W	1920 ± 86

Table 4-3. Density of waste forms fabricated using cluster 1 waste simulant

Sample Composition	Average Density (kg/m ³)
45FA+5W	1950 ± 23
45FA+10W	1994 ± 16
45FA+15W	1974 ± 19
45FA+20W	1978 ± 13
45CaSi+15W	2067 ± 17
45CaSi+20W	2075 ± 13
25CaSi+25W	1941 ± 11
25FA+25W	1874 ± 30
25FA+25W+1BA	1827 ± 46
35FA+10Slg+10W	2007 ± 17
35FA+10Slg+15W	1995 ± 18

Table 4-4. Density of waste forms fabricated using cluster 2 waste simulant

Sample Composition	Average Density (kg/m ³)
45FA+5W	1956 ± 24
45FA+10W	1975 ± 17
45FA+15W	1982 ± 19
45FA+20W	1997 ± 34
45CaSi+15W	2075 ± 10
45CaSi+20W	2079 ± 10
25CaSi+25W	1828 ± 28
25FA+25W ^a	1829 ± 20
25FA+25W+1BA ^a	1722 ± 107
35FA+10Slg+10W ^a	1853 ± 163
35FA+10Slg+15W	1902 ± 54

^a. Compositions showed expansions and lower densities.

Table 4-5. Density of waste forms fabricated using mixed waste simulant

Sample Composition	Average Density (kg/m ³)
45FA+5W	1973 ± 12
45FA+10W	1983 ± 12
45FA+15W	1991 ± 15
45FA+20W	1994 ± 12
45CaSi+15W	2061 ± 17
45CaSi+20W	2076 ± 22
25CaSi+25W	1899 ± 27
25FA+25W	1885 ± 25
25FA+25W+1BA ^a	1639 ± 104
35FA+10Slg+10W	1989 ± 12
35FA+10Slg+15W	1995 ± 19

^a. Compositions showed expansions and lower densities.

4.3 Compressive Strength of As-fabricated Samples

Compressive strength tests were conducted with cylindrical samples of 0.5-in. diameter and 1.0-in. length. Tests were conducted under a standard laboratory atmosphere on a Model 4505 Instron Universal Testing System, shown in Figure 4-3. The loading rate was 1 mm/min. Loads versus cross-head displacements were recorded. Compressive strength was calculated by measuring the peak load at failure and dividing by the initial sample cross-sectional area. For each formulation, at least three samples were tested, and an average value was determined. The minimum compressive strength requirement for the waste forms is 500 psi [11].

Figures 4-4 through 4-6 show the compressive strengths measured for the baseline samples with varying waste stream loadings and filler proportions. Since strength of the waste forms is expected to increase with time as the curing continues, for a specific waste form composition, strengths were measured at time intervals of approximately 14, 30, and 60 days. Figures 4-7 through 4-9 report similar data for cluster 1 waste forms; Figures 4-10 through 4-12 for cluster 2 waste forms; and Figures 4-13 through 4-15 for mixed waste forms. Raw data for the plots are presented in Appendix A.



Figure 4-3. Test setup on Instron for measurement of waste form compressive strength

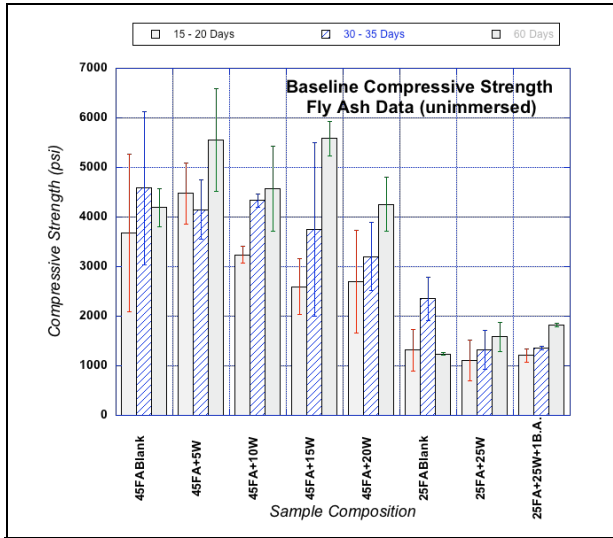


Figure 4-4. Compressive strength of waste forms with baseline waste simulant and fly ash filler

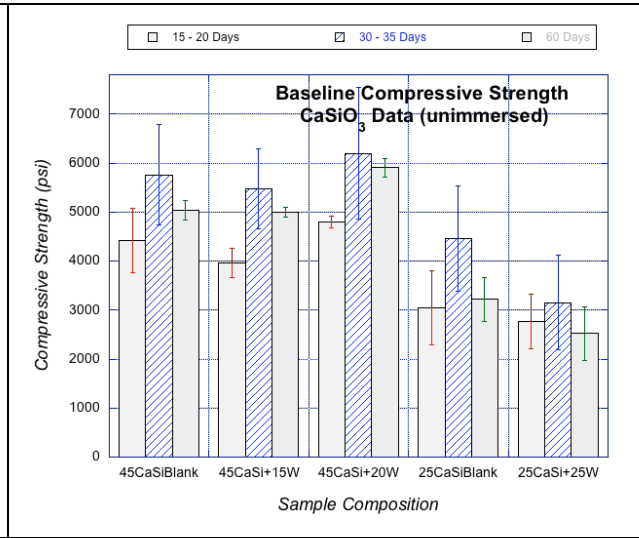


Figure 4-5. Compressive strength of waste forms with baseline waste simulant and CaSiO₃ filler

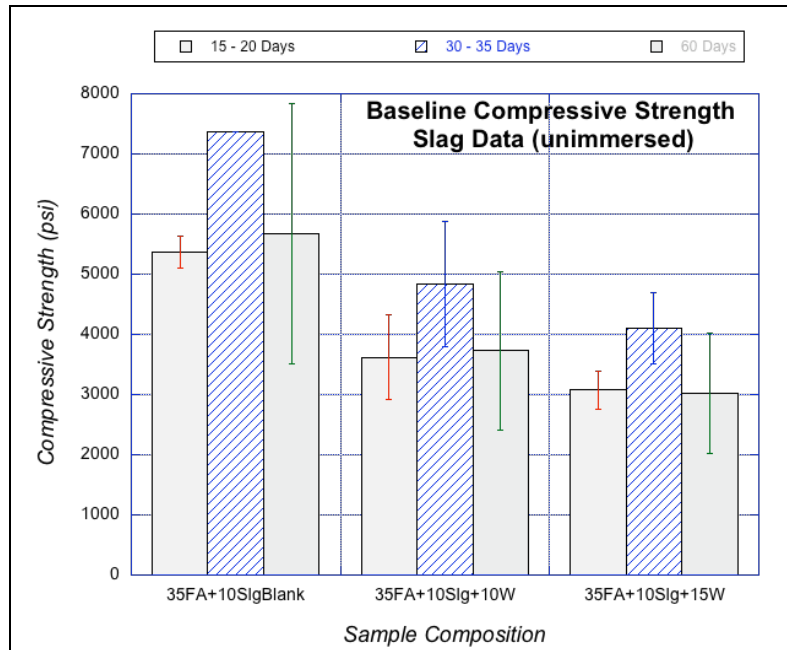


Figure 4-6. Compressive strength of waste forms with baseline waste simulant and slag + fly ash filler

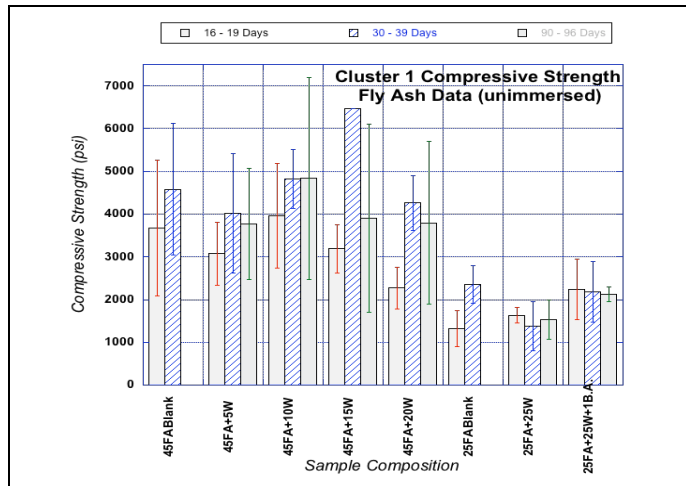


Figure 4-7. Compressive strength of waste forms with cluster 1 waste simulant and fly ash filler

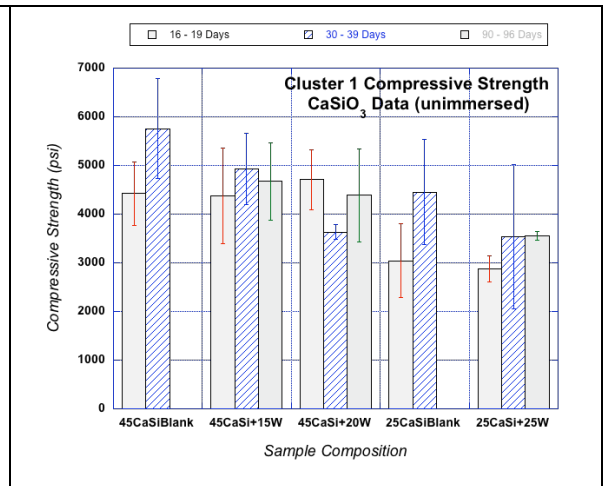


Figure 4-8. Compressive strength of waste forms with cluster 1 waste simulant and CaSiO₃ filler

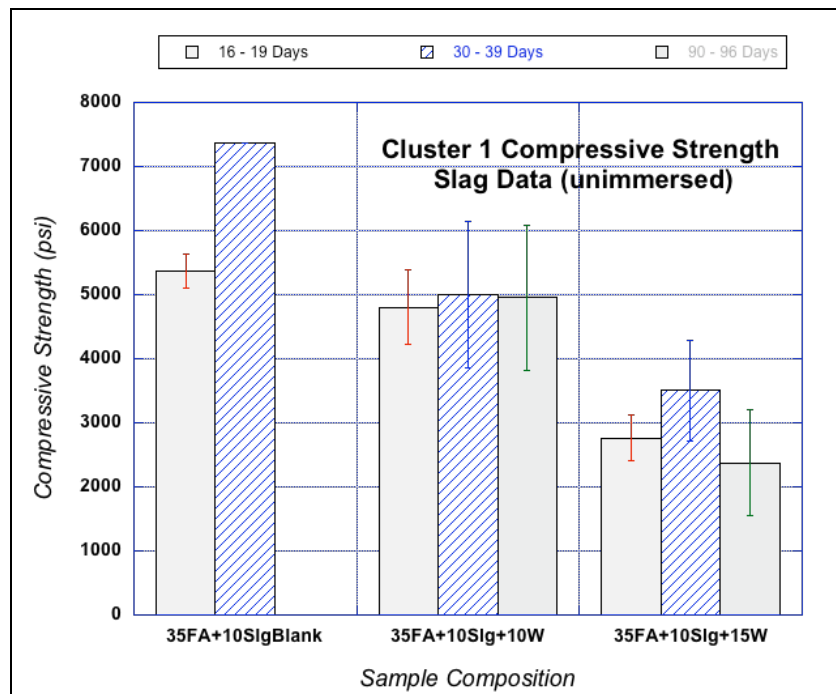


Figure 4-9. Compressive strength of waste forms with cluster 1 waste simulant and slag + fly ash filler

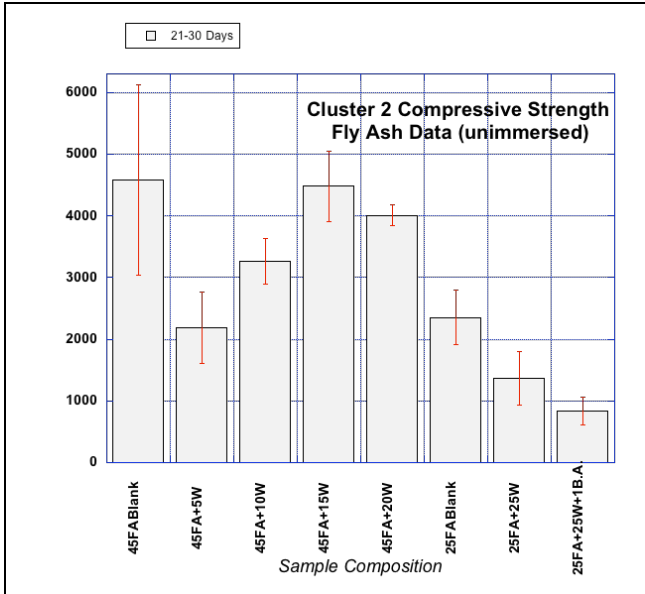


Figure 4-10. Compressive strength of waste forms with cluster 2 waste simulant and fly ash filler

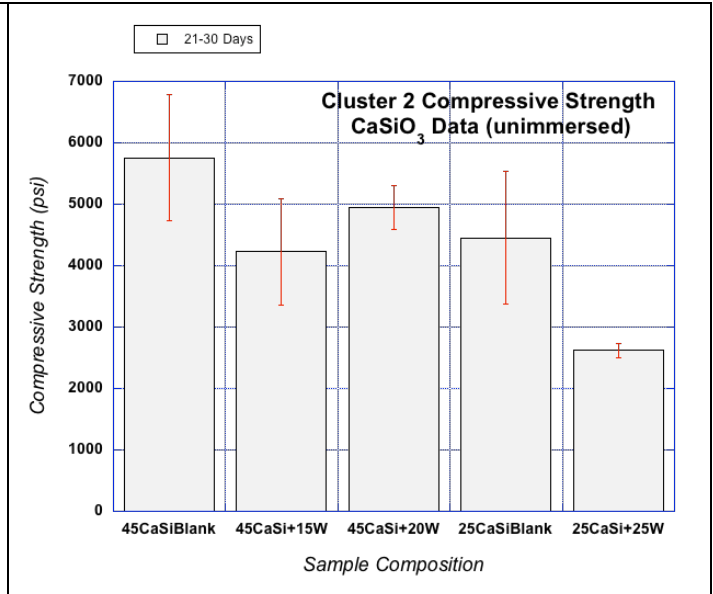


Figure 4-11. Compressive strength of waste forms with cluster 2 waste simulant and CaSiO₃ filler

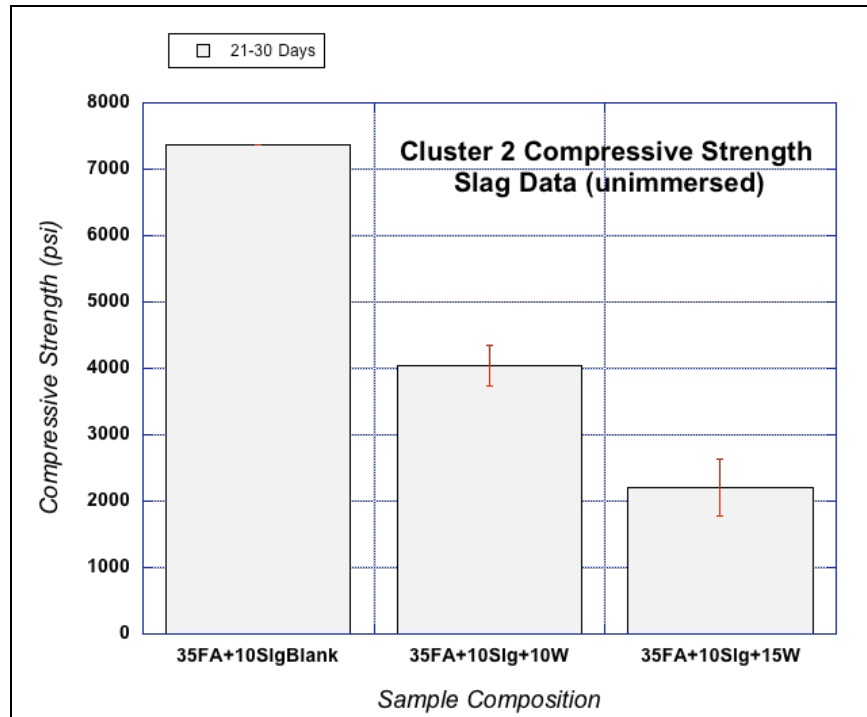


Figure 4-12. Compressive strength of waste forms with cluster 2 waste simulant and slag + fly ash filler

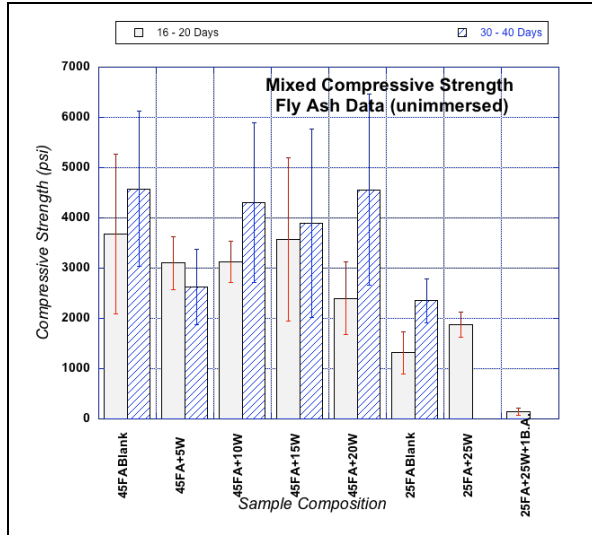


Figure 4-13. Compressive strength of waste forms with mixed simulant stream and fly ash filler

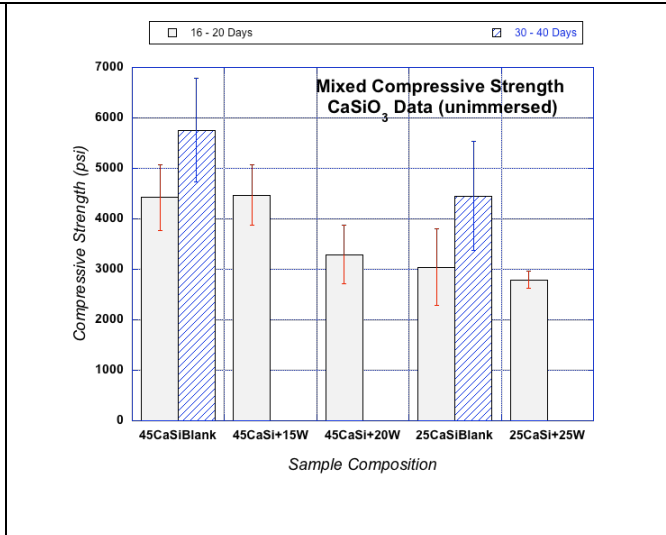


Figure 4-14. Compressive strength of waste forms with mixed waste simulant and CaSiO₃ filler

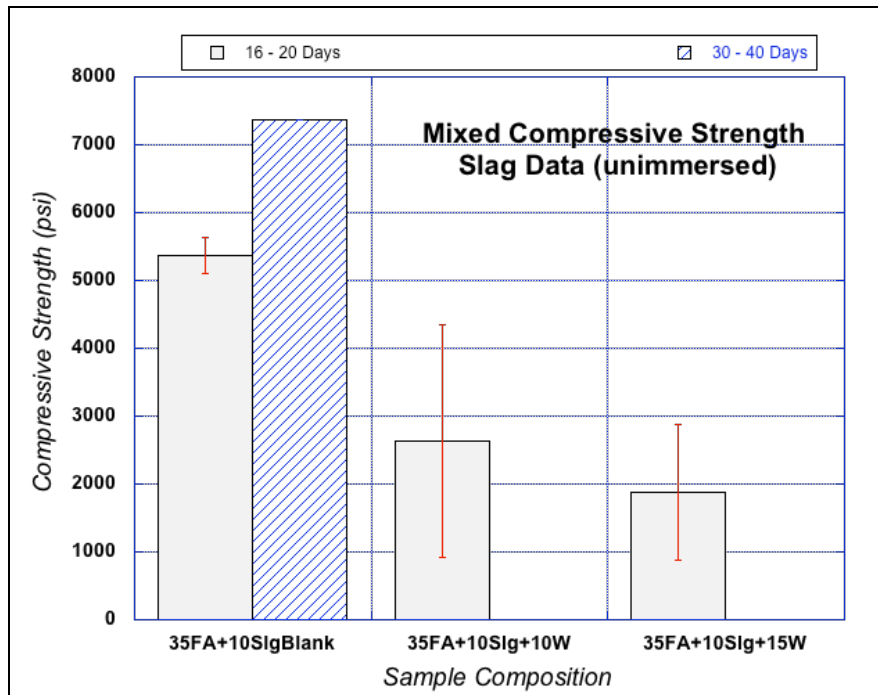


Figure 4-15. Compressive strength of waste forms with mixed waste simulant and slag + fly ash filler

4.4 Summary

1. Compressive strengths of the waste forms fabricated with baseline simulants (up to 20 wt.% waste loading) showed strengths > 2500 psi, independent of the filler material. The compressive strength appears to increase with waste loading and is optimum at about 20 wt.%.
2. Compressive strengths for baseline waste forms increased with time from 2500 psi to 5000 psi over a 60-90 day period.
3. Baseline waste forms with 25 wt.% filler (fly ash or CaSiO_3) and waste loadings of 25 wt.% (with and without boric acid) showed significantly lower strengths as compared to the 45 wt.% filler forms. This effect is probably due to insufficient binder phase in the waste form.
4. On the average, waste forms with CaSiO_3 filler performed slightly better in compressive strengths as compared to those with the fly ash and fly ash + slag.
5. Cluster 1 waste forms showed trends in compressive strength that were quite similar to those of the baseline waste forms.
6. For cluster 2 waste forms, compressive strengths were measured only for 20-30 day curing period. Trends were also similar to those of the baseline waste forms.
7. Mixed waste forms showed somewhat larger scatter in the data. Again, optimized strengths were at 15-20 wt.% loading for fly ash and CaSiO_3 fillers and 10 wt.% waste loading for slag + fly ash filler.

5 WATER IMMERSION TEST

Water immersion tests were conducted on samples from the same batch that were used for the strength evaluations, for which the samples were not immersed in water. For each composition, nine samples were placed in water. At the end of approximately 1, 2, and 3 months, the samples were retrieved, dried, weighed, and tested for compression strength. In addition to the compressive strengths, weight loss of the waste forms as a function of water exposure time was monitored. This test provides insight into the stability of the waste forms.

5.1 Waste Form Appearance

Once the waste forms were exposed for the requisite interval of time in water, they were wiped dried in a hood and inspected for white residue formation or cracking in the sample. Figures 5-1 through 5-4 are representative photographs of the waste forms fabricated using baseline, cluster 1, cluster 2, and mixed waste simulants, respectively, after their exposure for three months in water. These waste forms did not have any RCRA metals or additives such as zeolite, SnCl_2 , or K_2S . In each set, samples are for the three filler materials: class C fly ash, CaSiO_3 , and mixture of class C fly ash and slag. The waste loadings were 10-20 wt.%.

As can be seen in the figures, no residue was observed on the waste form surface. In addition, there are no cracks or sign of degradation of the waste forms after 3 months of the water immersion test.



Figure 5-1. Waste form samples with baseline waste simulant loading after 3 months water immersion: (top) 45 % class C fly ash filler +20 % waste, (middle) 45 % CaSiO_3 filler + 20 % waste, and (bottom) 35% class C fly ash + 10 % slag + 10 % waste

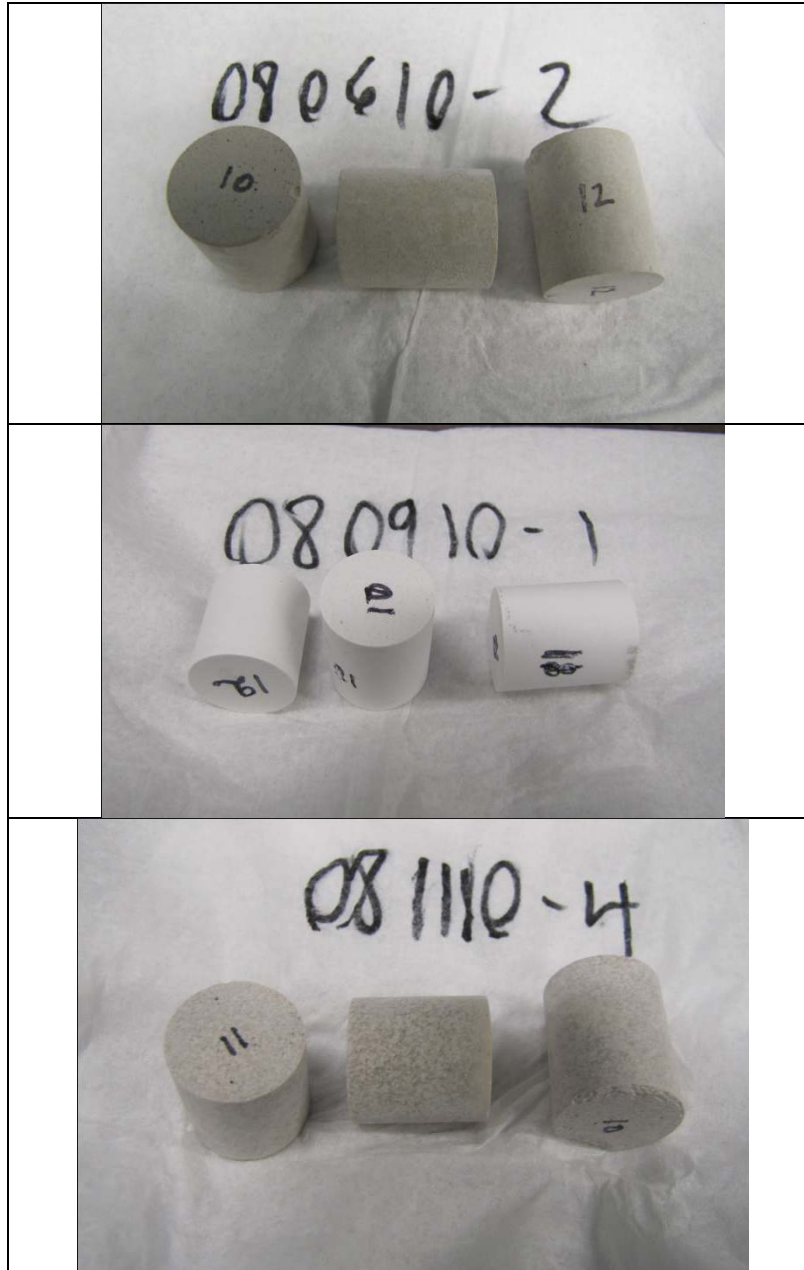


Figure 5-2. Waste form samples with cluster 1 waste simulant loading after 3 months water immersion: (top) 45 % class C fly ash filler + 20 % waste, (middle) 45 % CaSiO_3 filler + 20 % waste, and (bottom) 35% class C fly ash + 10 % slag + 10 % waste

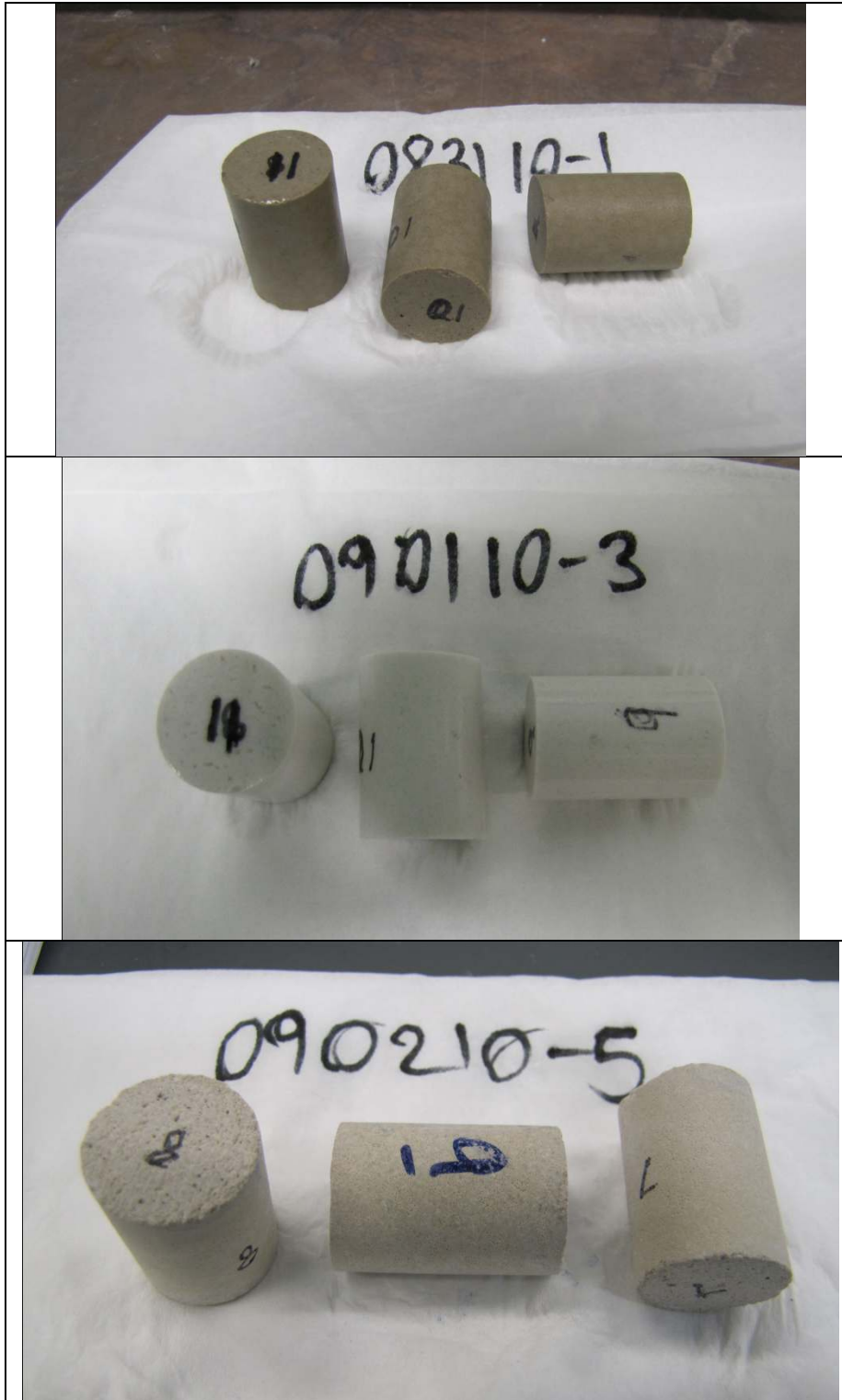


Figure 5-3. Waste form samples with cluster 2 waste simulant loading after 3 months water immersion: (top) 45 % class C fly ash filler + 20 % waste, (middle) 45 % CaSiO_3 filler + 20 % waste, and (bottom) 35% class C fly ash + 10 % slag + 15 % waste

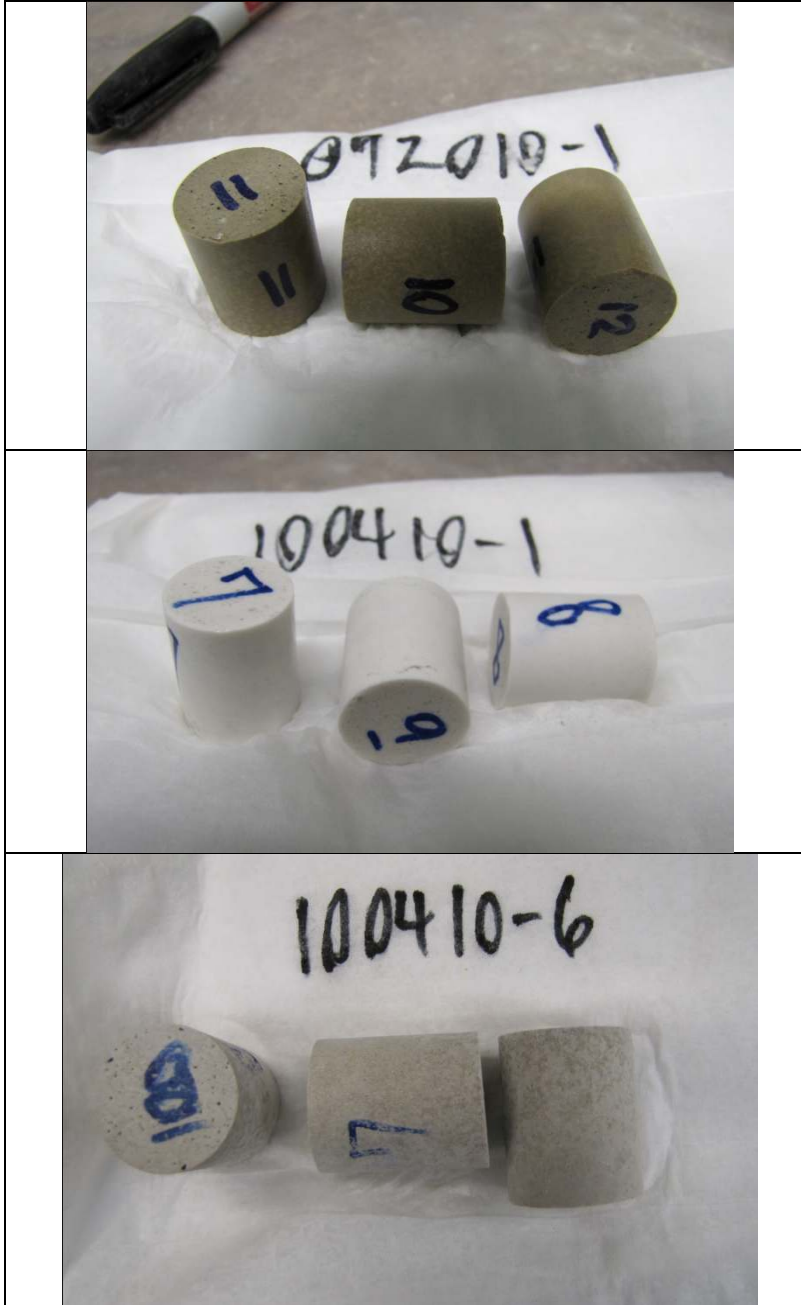


Figure 5-4. Waste form samples with mixed waste simulant loading after 3 months water immersion: (top) 45 % class C fly ash filler + 20 % waste, (middle) 45 % CaSiO_3 filler + 20 % waste, and (bottom) 35% class C fly ash + 10 % slag + 10 % waste

5.2 Weight Loss Measurements

As part of the water immersion testing, weight loss of the samples at 1, 2, and 3 month intervals was monitored in separate sets of samples. Samples were wiped with wipes and were allowed to air dry for several days in a hood before making measurements. Weight loss was determined as percent loss from the original sample weight before it was placed in water. The tests were done in triplicate at each condition.

Figures 5-5 through 5-9 show the weight loss for the various waste forms fabricated using the three filler materials: fly ash, CaSiO_3 , and mixture of fly ash and slag. It should be noted that the weight loss reported for different immersion times are on different samples that were left undisturbed. For the fly ash and CaSiO_3 samples waste loadings were 15 wt.% and 20 wt.% for all four waste simulants. For fly ash + slag filler, only 10 wt.% loading was tested.

The results show that during the first month, the weight loss is the highest, 5-14 wt.%. This weight loss is probably due to unreacted binder powders on the surface of the waste form being removed. There is not much weight change for the samples immersed for 2 and 3 months. Depending on the sample composition, there is approximately 2-3 wt.% change during the subsequent 2-month period, indicating the waste form is stable.

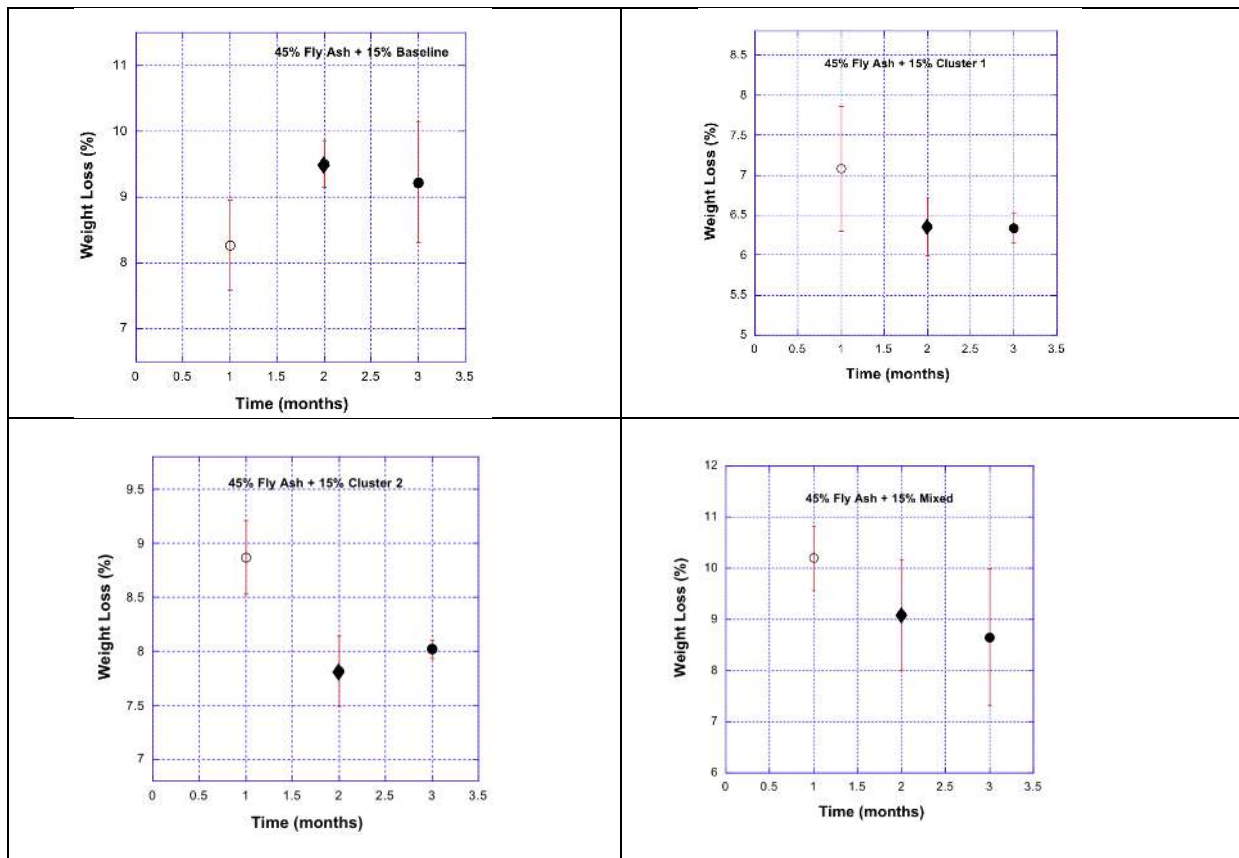


Figure 5-5. Weight loss of waste forms during water immersion tests as a function of time for 45% Class C fly ash and 15 wt.% of various waste loadings

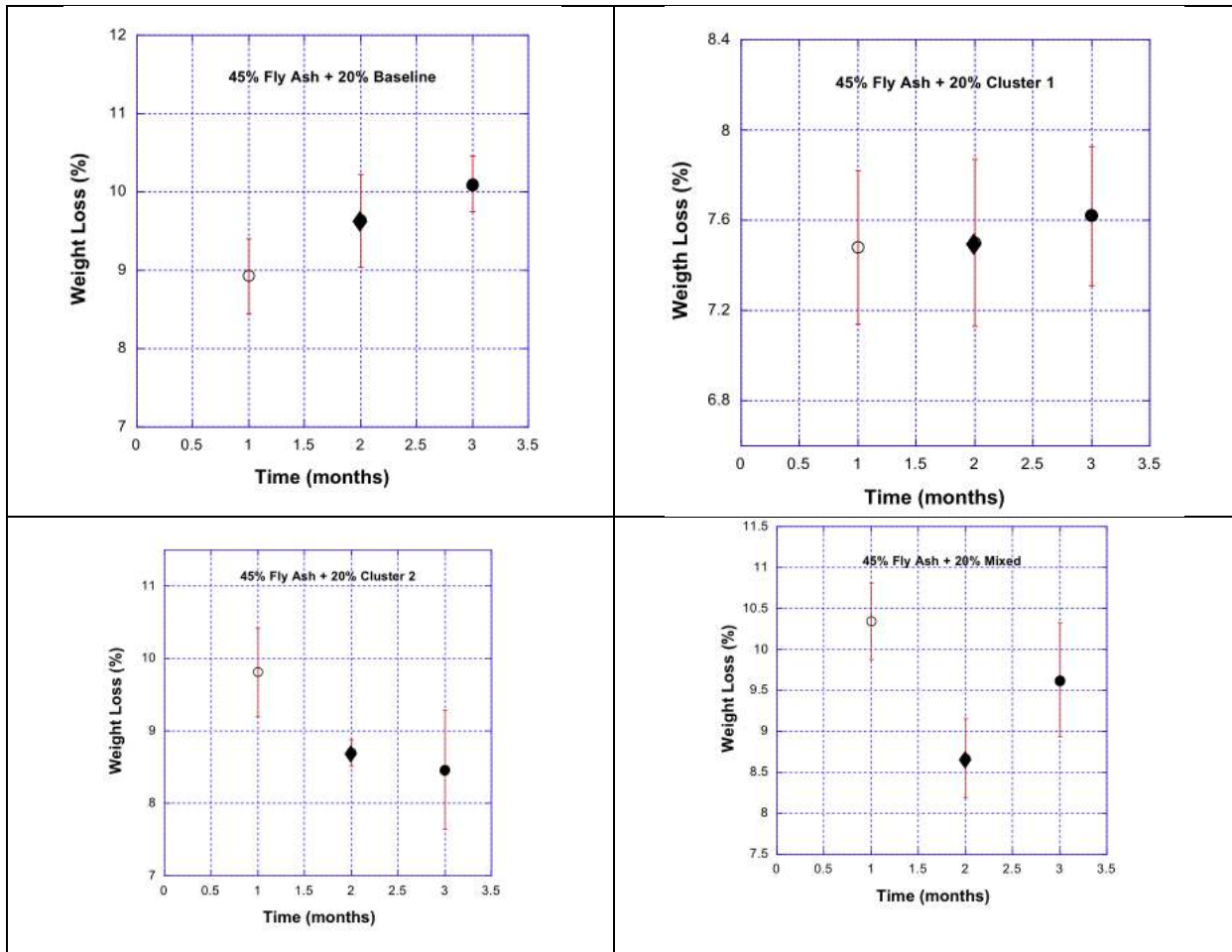


Figure 5-6. Weight loss of waste forms during water immersion tests as a function of time for 45% Class C fly ash and 20 wt.% of various waste loadings

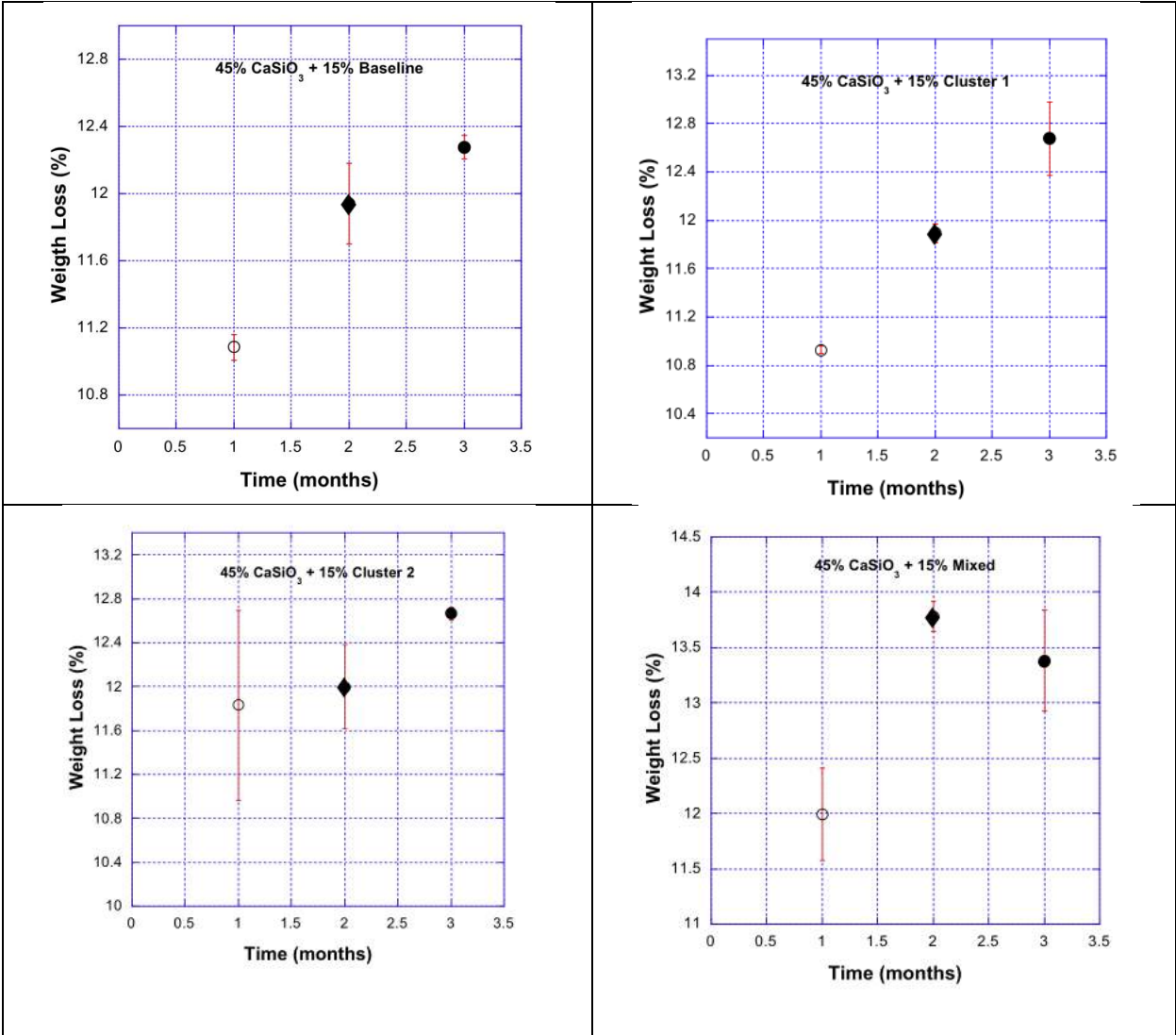


Figure 5-7. Weight loss of waste forms during water immersion tests as a function of time for 45% CaSiO₃ and 15 wt.% of various waste loadings

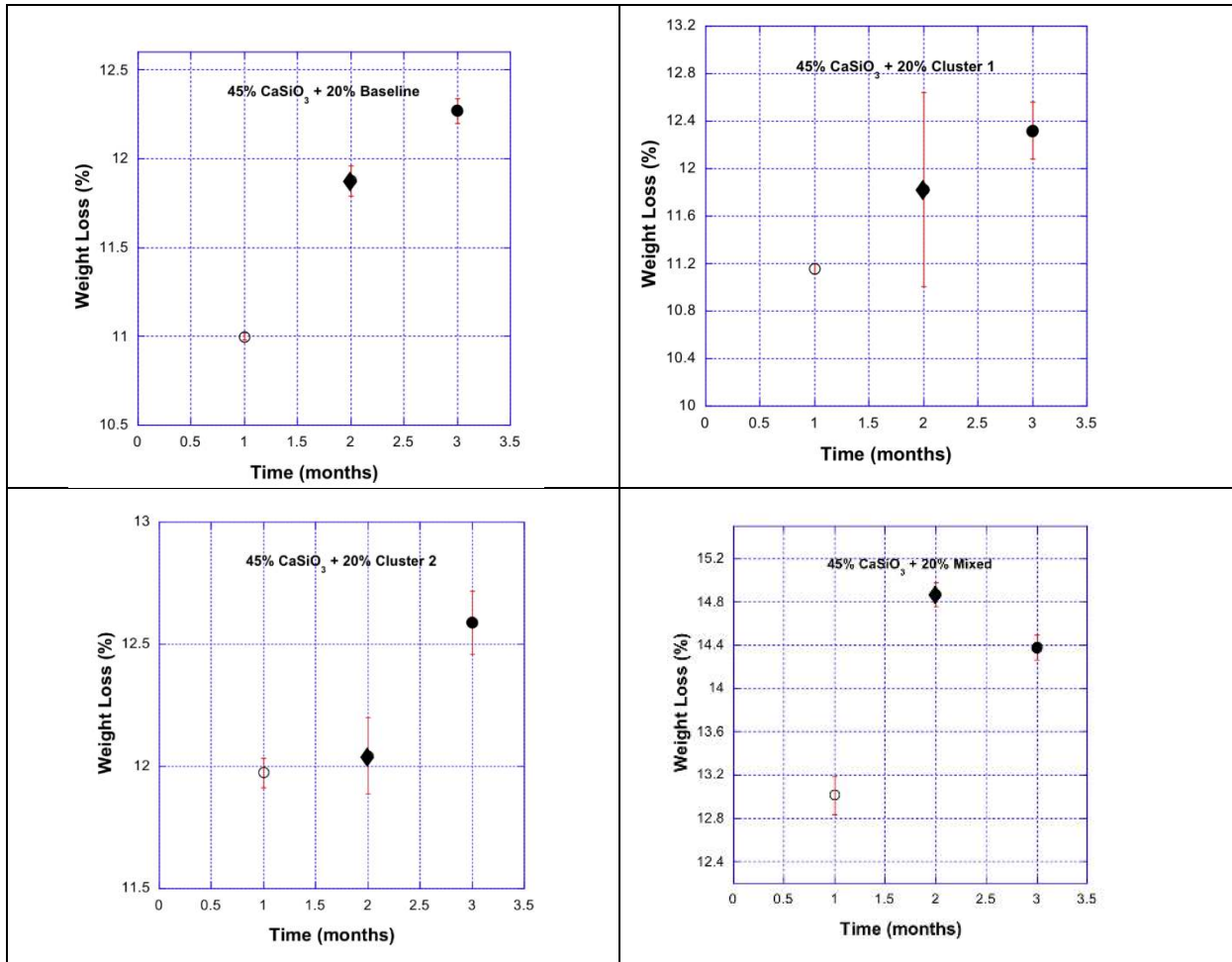


Figure 5-8. Weight loss of waste forms during water immersion tests as a function of time for 45% CaSiO₃ and 20 wt.% of various waste loadings

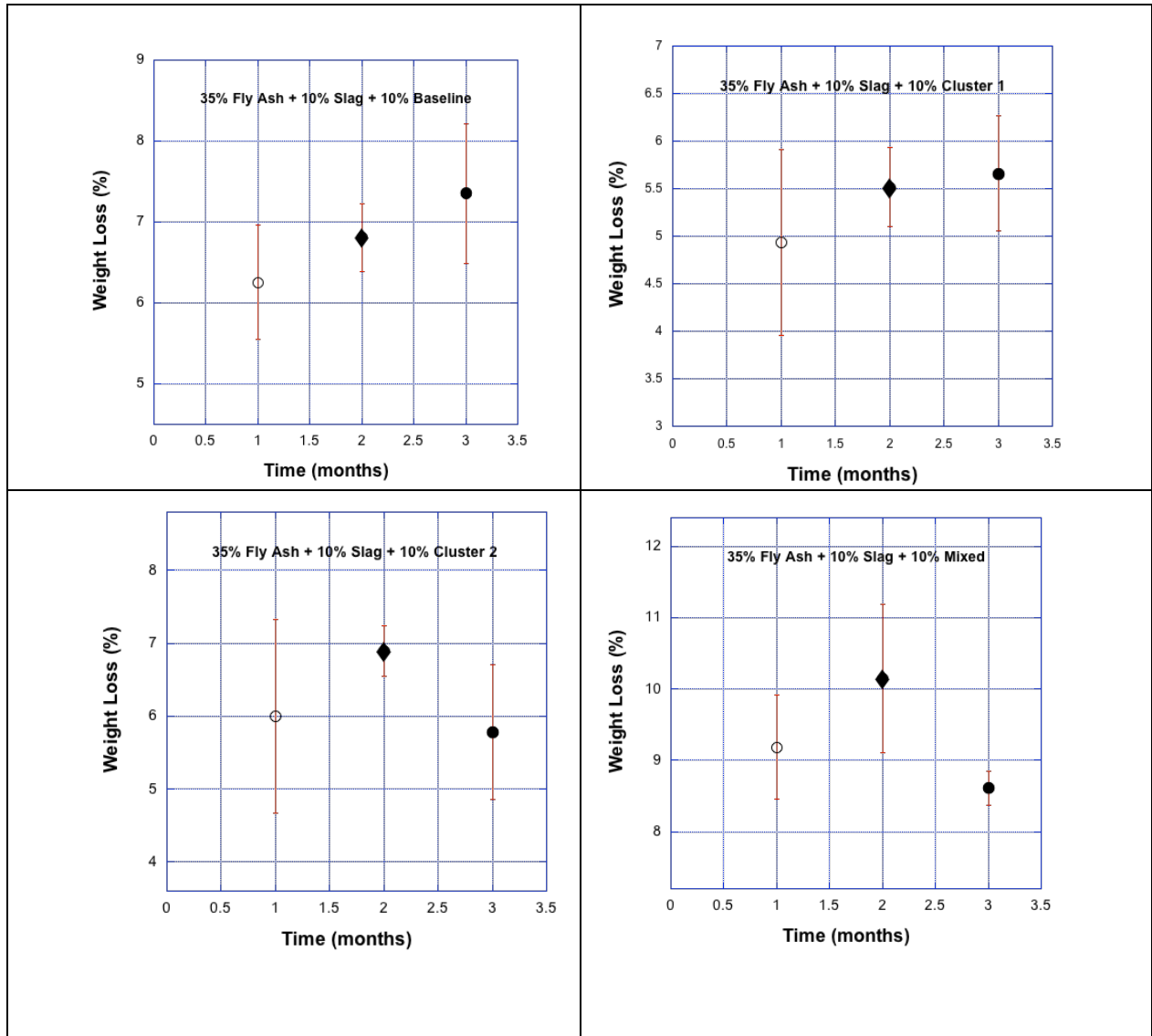


Figure 5-9. Weight loss of waste forms during water immersion tests as a function of time for 35% Class C fly ash, 10% slag, and 10 wt.% of various waste loadings

5.3 Compression Strength

Compression tests on the water-exposed samples were conducted in the same manner as for the as-fabricated samples described in Section 4.3. Results from the water immersion tests are shown in Figures 5-10 through 5-12 for baseline waste stream formulations with different filler. Similar test results are reported for cluster 1 formulations in Figures 5-13 through 5-15, cluster 2 formulations in Figures 5-16 through 5-18, and mixed formulations in Figures 5-19 through 5-21. The strengths of the water-immersed samples are compared with the as-fabricated strengths (15-20 day cure) of the waste forms as well.

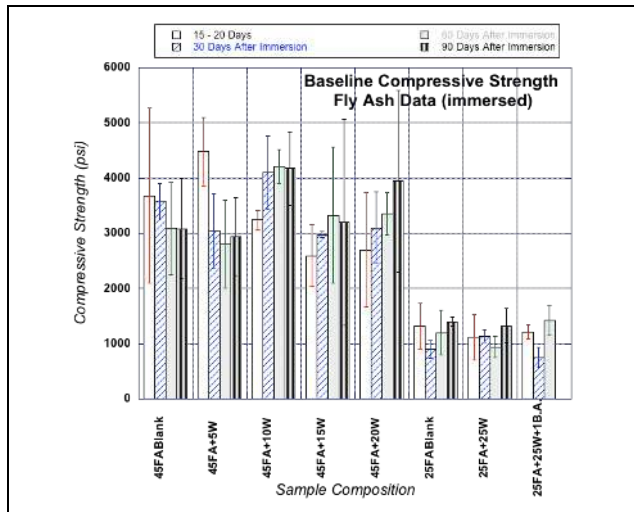


Figure 5-10. Compressive strength of waste forms with baseline waste simulant and fly ash filler as a function of immersion time

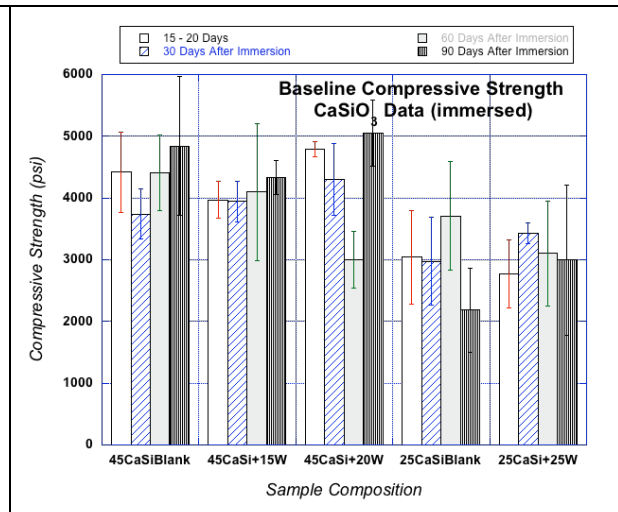


Figure 5-11. Compressive strength of waste forms with baseline waste simulant and CaSiO_3 filler as a function of immersion time

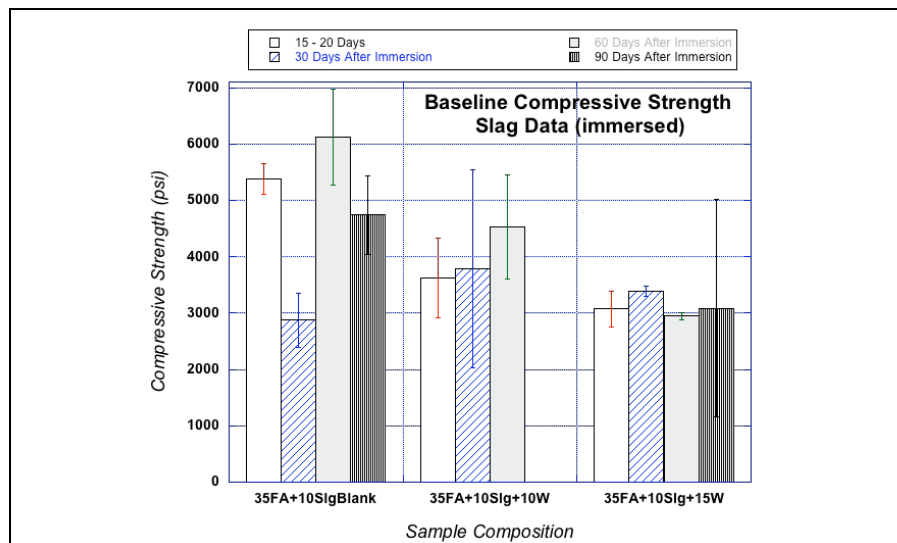


Figure 5-12. Compressive strength of waste forms with baseline waste simulant and slag and fly ash filler as a function of immersion time

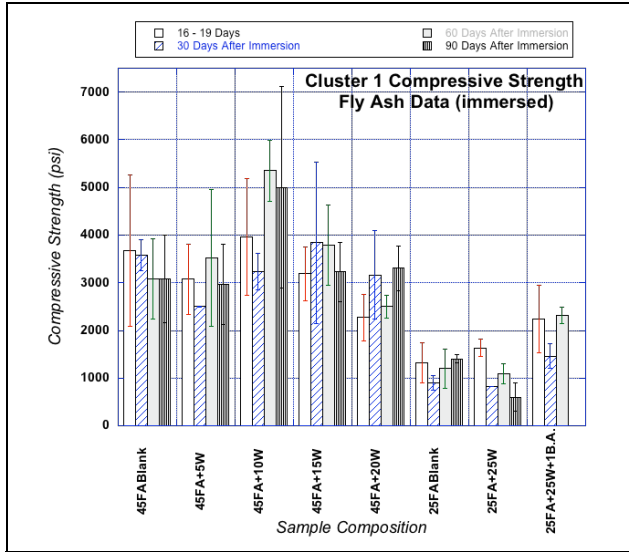


Figure 5-13. Compressive strength of waste forms with cluster 1 waste simulant and fly ash filler as a function of immersion time

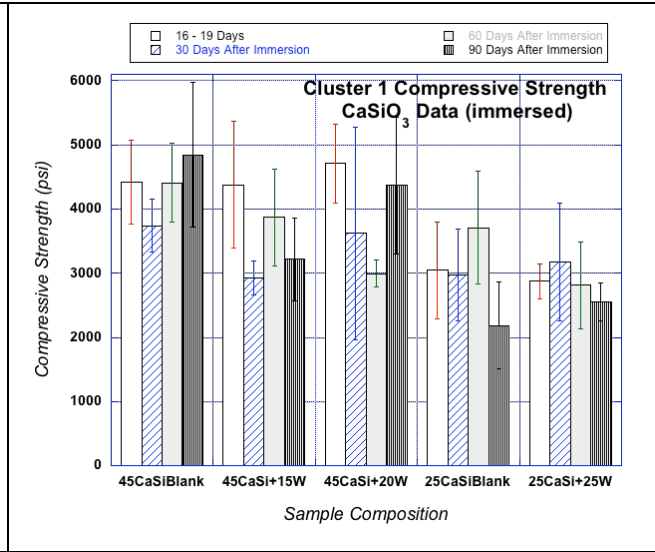


Figure 5-14. Compressive strength of waste forms with cluster 1 waste simulant and CaSiO₃ filler as a function of immersion time

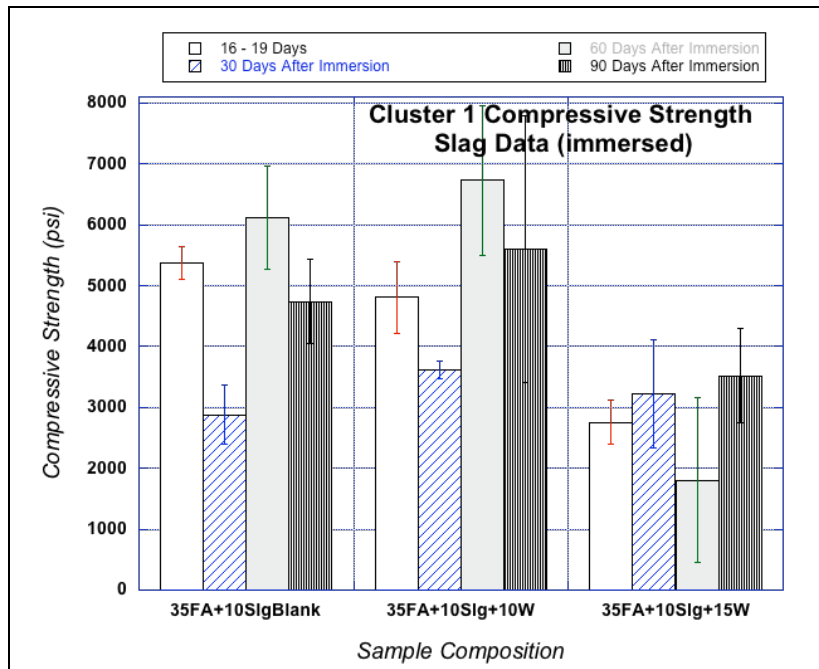


Figure 5-15. Compressive strength of waste forms with cluster 1 waste simulant and slag and fly ash filler as a function of immersion time

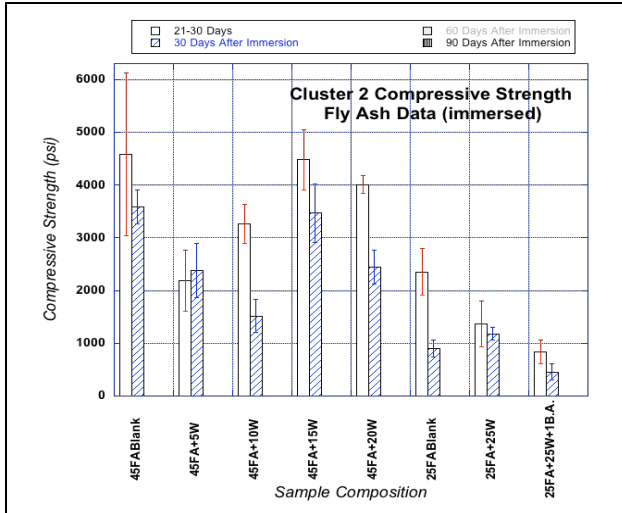


Figure 5-16. Compressive strength of waste forms with cluster 2 waste simulant and fly ash filler as a function of immersion time

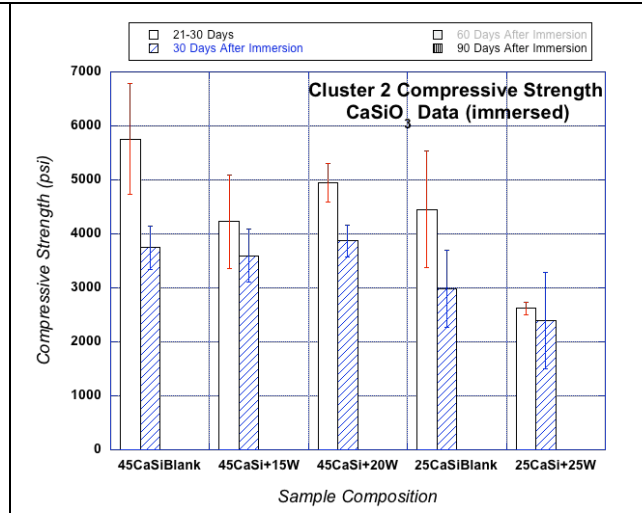


Figure 5-17. Compressive strength of waste forms with cluster 2 waste simulant and CaSiO₃ filler as a function of immersion time

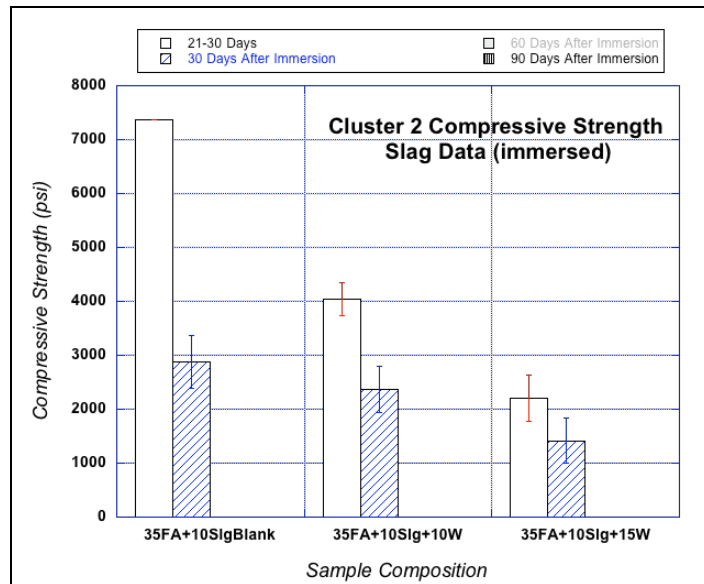


Figure 5-18. Compressive strength of waste forms with cluster 2 waste simulant and slag and fly ash filler as a function of immersion time

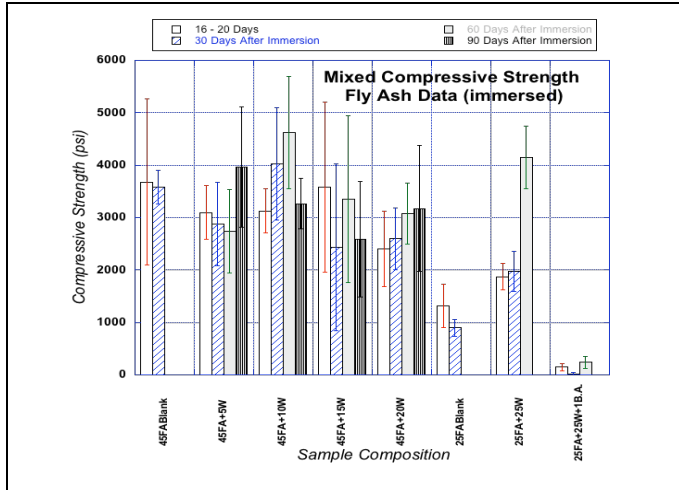


Figure 5-19. Compressive strength of waste forms with mixed waste simulant and fly ash filler as a function of immersion time

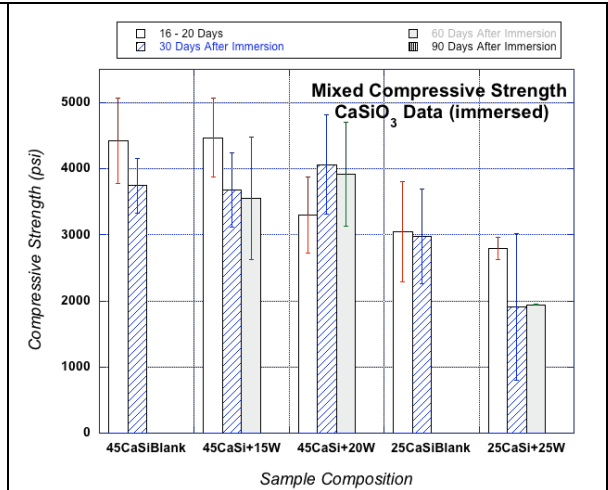


Figure 5-20. Compressive strength of waste forms with mixed waste simulant and CaSiO₃ filler as a function of immersion time

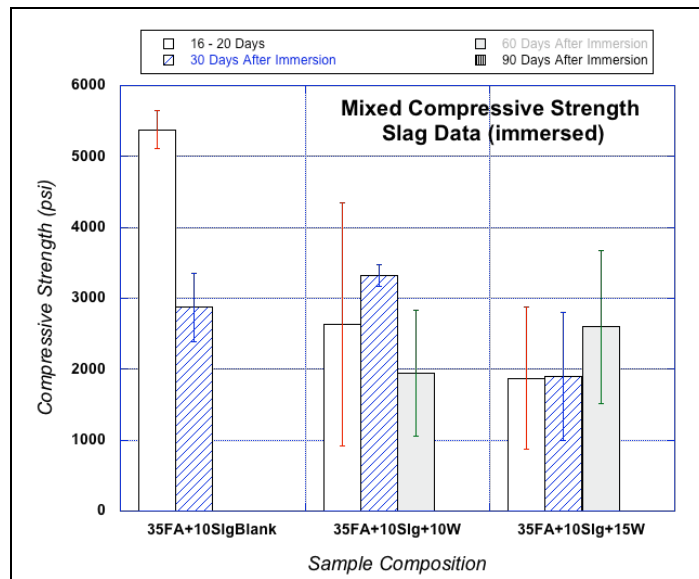


Figure 5-21. Compressive strength of waste forms with mixed waste simulant and slag and fly ash filler as a function of immersion time

5.4 Summary

Weight loss of the waste forms was measured on samples immersed in water for a period of 1, 2, and 3 months to determine the stability of the waste forms. The waste forms contained different filler materials (fly ash, CaSiO_3 , and fly ash + slag) and had waste loadings of 15 and 20 wt.%. These samples did not contain zeolite or any other additives. The following conclusions were reached from the weight loss data:

1. The majority of the weight loss (~5-14 wt.%) occurred in the first month of the test period. This loss is probably due to residual unreacted MgO and potassium phosphate on the waste form surface that washed off. Water from the immersion tests was not analyzed. However, based on the results from the analysis conducted on the ANS 16.1 leachate (Section 6.1), the concentration of Mg and P in the leachate was higher in the initial stages of the leach test as well, supporting the above conjecture. Weight loss trends for the forms fabricated with the four waste simulants were quite similar.

It should be noted that the initial weight loss is strongly dependent on the surface-to-volume ratio. The samples used in the study have a very high surface-to-volume ratio. For large waste forms, the surface to volume ratios will be significantly smaller, and the weight losses will thus be lower as well.

2. Subsequent immersion time intervals (2 & 3 months) did not show any additional significant drop in the weight loss over the first one-month water immersion.
3. It appears that the waste forms fabricated using CaSiO_3 filler exhibited slightly higher weight loss as compared to the other filler compositions.
4. Overall, the waste forms appeared to be stable over the 3-month water immersions tests. No evidence of any surface residue buildup was found.
5. The compressive strengths of the water-immersed samples were similar or even increased with immersion as compared to the as-fabricated samples for the same filler and waste loading for baseline, cluster 1, and mixed wastes.
6. For cluster 2 waste forms, the strength of the waste forms declined with immersion time as compared to the as-fabricated samples.
7. The compressive strengths of the waste forms are controlled by possibly two competing effects, namely, increase in strength as the phosphate phase cures with time and decrease in strength due to any loss of the matrix material in water. These effects could be controlling the overall strength of the waste forms. It is possible that for cluster 2 waste forms, because of the short curing time, the decrease in post immersion strength tests is more obvious.

8. Similar to the un-immersed samples, waste forms with 25 wt.% filler and 25 wt.% waste loading exhibited low compressive strengths and were excluded from further investigations.

6 AMERICAN NUCLEAR SOCIETY'S 16.1 LEACHING TEST

6.1 Leachability Index Determination

As per the ANS 16.1 test procedure, the leachability index (LI) for radionuclide species and surrogates is determined by conducting a test in which a cylindrical (1-in. length and 0.50-in. diameter) waste form sample is suspended in a known volume (10X the surface area of the sample) of leachant. The water is replaced at fixed time intervals (2, 5, 17, 24, 24, 24, 24, 336, 672, and 1032 h) to simulate dynamic leaching conditions. The contaminant species, n , in the leachate were analyzed by induction-coupled plasma–mass spectrometry (ICP-MS). The effective diffusivity (D_e) and leachability index (LI) are given by

$$D_e = \pi[(a_n/A_0)/(\delta t)_n]^2(V/S)^2(T)$$

$$LI = (1/n)\Sigma[\log (b/D_e)]_n$$

In the above equations, a_n represents the amount (g) of contaminant leached from the waste form during the leaching interval n ($t_{n-1} < t < t_n$), A_0 is the amount of contaminant in the waste form at the beginning of the leach test (g), $(\delta t)_n (= t_n - t_{n-1})$ is the duration of n^{th} leaching interval (s), D_e is the effective diffusivity (cm^2/s), V is the volume of the waste form (cm^3), S is the external surface area of the waste form (cm^2), T is the mean time of leaching interval (s) and is represented as $[0.5(t_n^{1/2} + t_{n-1}^{1/2})]^2$, and b is a constant ($1 \text{ cm}^2/\text{s}$).

Table 6-1 shows the ICP-MS (conducted by ANL's Analytical Chemistry Lab) results for the various waste simulants used to fabricate the waste forms for ANS 16.1 tests. The purpose of conducting this analysis was to determine the concentration of the various elemental species in the waste stream. Based on the waste loadings for the various ANS 16.1 samples, initial amounts of specific specie, A_0 , in the waste form at the start of the test were determined. Initial concentration used for leachability index (LI) calculation corrected for the amount of the specific specie lost during the initial "rinse" step of the test protocol.

Table 6-2 presents the leachability indices (average values over the test duration) results for the various samples evaluated, including the blank samples. Leachate solutions were not only analyzed for the radioactive surrogates I and Re, but also for Mg, Na, P, Si, and Ag. The LI of the non-radioactive surrogates provides insight into the stability of the waste form itself. All samples were run in duplicate. Expected target values for LI's for Re (Tc), I and Na are 9, 11, and 6, respectively [10,11]. Appendix C presents the diffusivity values for the various species at each leach interval for selected waste forms.

It should be noted that for cluster 2 waste stream, only CaSi filler based waste forms were evaluated. This was because, as discussed in Section 4.1, of discernible expansion of the samples observed for fly ash and fly ash + slag based waste form at higher waste loadings.

Results in Table 6-2 are for tests conducted for 60 days, except for the baseline samples (in bold), which were conducted for 90 days. There are three major results: (a) iodine was below the detection limits (5 ppb) for all the waste forms at each leaching interval, (b) rhenium LI ranged

from 7.9 to 9, and (c) LI for Na ranged from 7.6-10.4. There was no specific trend observed with respect to the waste simulant or the filler types.

Leachability indices for iodine were calculated by assuming the detection limit of 5 ppb at each leaching interval, including initial rinse. This is an overly conservative estimate and may not be an accurate representation since in most cases, by taking 5 ppb at each time interval, the cumulative fraction of I leached out exceeds 1. Nevertheless, actual values of iodine LI are expected to be greater than those listed in Table 6-2.

Leachability index for Re for all the samples tested range from 8.1 to 9. The target LI is met for mixed waste forms with 20 wt.% waste loading. In general, for Class C fly ash waste forms, Re LI is on the higher side (>8.5). Blast furnace slag filler did not show any distinct advantage.

Sodium LI for all samples were above the target value of 6, indicative of the containment of the specie. There was no distinct trend for Na leaching as a function waste stream type.

LI for binder phase elements such as Mg, P, and Si were also analyzed. These values are listed in Table 6-2. LI for Mg and Si are in 10-12 range, however, P LI is 8-9. This suggests that there may be some un-reacted potassium phosphate in the waste form that egresses out during the leaching tests. This could also explain the weight losses observed in the initial 1-month period of the water immersion testing.

Table 6-1. Results (in µg/L) from ICP-MS conducted on the waste stimulants used for preparing waste forms for ANS 16.1 tests

Simulant	Na	Mg	Si	P	Ag	Re	I
Baseline	1.96E+07	207	9.06E+04	2.05E+05	< 3.1	5.30E+03	1.63E+03
Cluster 1	1.92E+07	< 70	9.54E+04	1.59E+05	< 6.1	3.14E+04	1.35E+03
Cluster 2	2.04E+07	160	4.04E+04	1.87E+05	< 3.1	4.02E+03	1.58E+03
Mixed	2.13E+07	1.33E+03	4.12E+04	1.39E+05	< 30.5	1.28E+05	9.76E+03

Table 6-2. Average leachability indices from ANS 16.1 tests conducted on the various waste forms

Sample	Waste Simulant	Mg	Na	P	Si	Ag	Re	I ^a
45FA+Blank	Blank	10.5	-	8.4	12.5	16.3	-	-
45FA+Blank	Blank	10.5		8.4	12.5	16.2	-	-
45FA+20WS(B)^b	B	11.0	8.1	8.5	12.4	16.6	8.6	>6.5
45FA+20WS(B)	B	10.8	8.1	8.3	12.2	14.5	8.6	6.5
45FA+15WS(B)	B	10.6	8.2	8.3	12.2	16.1	8.6	>6.2
45FA+15WS(B)	B	10.7	8.2	8.4	12.3	16.4	8.6	>6.2
45CaSi+20WS(B)	B	10.9	7.6	8.4	11.3	17.2	8.4	>6.6
45CaSi+20WS(B)	B	11.0	7.6	8.5	11.3	17.7	8.4	>6.6
45CaSi+15WS(B)	B	11.0	7.7	8.5	11.3	16.9	8.3	>6.2
45CaSi+15WS(B)	B	11.0	7.7	8.5	11.4	17.8	8.3	>6.3
45CaSi+Blank	Blank	11.8	10.2	9.0	11.9	17.7	-	-
45CaSi+Blank	Blank	11.9	10.4	9.1	11.7	17.7	-	-
35FA+10Slag+15WS(B)	B	11.3	8.0	8.5	12.2	17.2	8.5	>6.3
35FA+10Slag+15WS(B)	B	11.3	8.0	8.5	12.2	17.1	8.5	>6.3
35FA+10Slag+Blank	Blank	11.5	9.6	8.8	12.2	17.2	-	-
35FA+10Slag+Blank	Blank	11.6	9.7	8.9	12.6	17.2	-	-
45FA+20WS(C1)	C1	10.8	8.0	8.4	12.3	16.0	8.7	>6.2
45FA+20WS(C1)	C1	11.0	8.0	8.4	12.3	16.1	8.8	>6.2
45FA+15WS(C1)	C1	10.7	8.0	8.3	12.2	16.1	8.6	>5.8
45FA+15WS(C1)	C1	10.7	8.0	8.3	12.2	16.8	8.6	>5.8
45CaSi+20WS(C1)	C1	11.2	7.6	8.5	11.8	17.4	8.1	>6.2
45CaSi+20WS(C1)	C1	11.2	7.6	8.5	11.6	17.5	8.1	>6.2
45CaSi+15WS(C1)	C1	11.3	7.6	8.6	11.7	17.7	8.0	>5.9
45CaSi+15WS(C1)	C1	11.3	7.6	8.5	11.6	17.7	8.1	>5.9
35FA+10Slag+15WS(C1)	C1	11.4	10.0	8.5	12.3	17.7	8.6	>8.0
35FA+10Slag+15WS(C1)	C1	11.6	8.1	8.6	12.4	17.7	8.5	>5.8
45CaSi+20WS(C2)	C2	11.3	7.6	8.5	11.6	17.0	8.1	>6.4
45CaSi+20WS(C2)	C2	11.4	7.6	8.6	11.7	17.0	7.9	>6.4
45CaSi+15WS(C2)	C2	11.3	7.6	8.6	11.6	17.0	8.0	>6.1
45CaSi+15WS(C2)	C2	11.4	7.7	8.7	11.8	17.0	7.9	>6.1
45FA+20WS(M)	M	10.6	8.1	8.5	12.3	17.0	8.9	>8.1
45FA+20WS(M)	M	10.6	8.1	8.5	12.3	15.0	9.0	>8.2
45FA+15WS(M)	M	10.7	8.1	8.5	12.3	16.7	8.7	>7.9
45FA+15WS(M)	M	10.6	8.1	8.5	12.3	16.8	8.7	>7.9
45CaSi+20WS(M)	M	11.0	7.7	8.5	11.5	17.4	8.2	>8.2
45CaSi+20WS(M)	M	11.1	7.7	8.5	11.5	17.3	8.1	>8.2
35FA+10Slag+15WS(M)	M	10.9	8.1	8.7	12.5	17.1	8.8	>7.9
35FA+10Slag+15WS(M)	M	10.9	8.1	8.6	12.4	17.1	8.9	>7.9

^a. Iodine was found below the detection level (<5 ppb). Detection level value was used to determine the LI.

^b. Bold results are for tests conducted over 90-day period; the rest are based on 60-day tests.

6.2 Analysis of Waste Forms after ANS 16.1 Test

After the 90-day tests, the waste form samples were removed and analyzed. While the waste form samples had superior LIs, minor degradation (cracks, discoloration, and slight residue formation) was observed in some waste forms. Figure 6-1 shows the post-90-day ANS 16.1 blank samples with the three different fillers and zeolite and SnCl_2 additives. The samples with CaSiO_3 and fly ash + slag fillers appear to be unchanged; however, the sample with 45 wt.% fly ash shows a fine crack in the middle. Since the water immersion samples (Section 5) did not show formation of any such cracks, we speculate that addition of zeolite in the waste form is the probable cause.

Figure 6-2 shows the waste forms with 15 wt.% baseline simulant loadings. Samples with fly ash and CaSiO_3 filler formed fine cracks. However, fly ash + slag filler showed more pronounced cracking. Note that cracking is lateral only (along the diameter).

Figure 6-3 shows the waste forms with 15 wt.% cluster 1 simulant loadings. Samples with fly ash and CaSiO_3 filler also formed fine cracks. However, the fly ash + slag filler sample had more pronounced cracking.

Figure 6-4 shows the waste forms with 15 wt.% cluster 2 simulant loadings. The sample with fly ash shows one crack. The sample with CaSiO_3 filler did not crack. The fly ash + slag filler sample had some cracking at the sample end.

Figure 6-5 shows the waste forms with 15 wt.% mixed simulant loadings. Samples with fly ash and CaSiO_3 did not crack. However, the fly ash + slag filler sample had some fine cracks at the sample end.

Similar trends were observed for waste forms with 20 wt.% loading. Based on these observations, several conclusions can be drawn:

1. Degradation during the ANS 16.1 was significantly more than that observed during the water immersion tests as reported in section 5-3. This enhanced degradation could be possibly due to the more severe test conditions of ANS 16.1, in terms of periodic change of immersion fluid.

The other possible reason could be the addition of zeolite (silicate) beads (for the iodine containment) in the samples fabricated for the ANS 16.1 test. Zeolite can react with potassium phosphate during the synthesis of waste forms with high pH (>12) simulants. Thus, there would be unreacted MgO powder in the waste form. During ANS 16.1 or water immersion tests, MgO can hydrate to form $\text{Mg}(\text{OH})_2$ that could cause swelling and cracking.

Second possibility is that since the waste simulants are rich in Na, there may be a reaction between zeolite and any pore fluid to form hydrous amorphous sodium silicate. This can also lead to volumetric expansion and cracking. Similar effect has been observed in Portland cements and is referred as “alkali-aggregate interactions” [12].

2. Cracking was more dominant for the 35% fly ash + slag samples followed by 45% fly ash samples.
3. The least amount of degradation occurred for CaSiO_3 filler samples.
4. For mixed waste simulant, no degradation was observed for both Class C fly ash and CaSiO_3 filler samples. Note that the mixed waste simulant had a pH of 8.6 as compared to >12 for the other three simulants. Prior neutralization of the baseline, cluster 1, and 2 waste simulants could possibly eliminate the cracking observed in some of the samples after ANS 16.1 tests.
5. Since iodine release was below detection levels, it not clear if zeolite addition is required in the waste forms. The Ceramicrete process itself could be sufficient to contain iodine. Under such circumstance, use of zeolite beads would be unnecessary. However, this needs to be established with further experimentation.

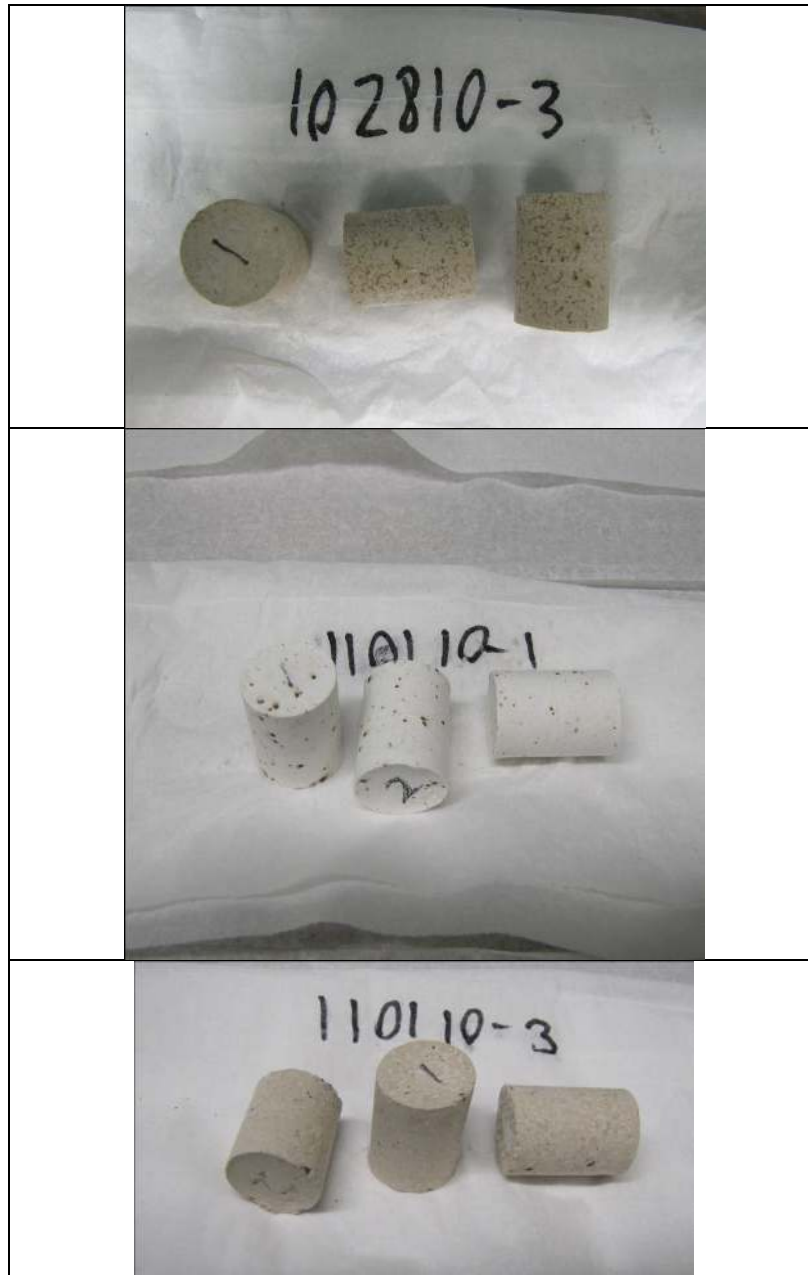


Figure 6-1. Blank waste forms after 90-day ANS 16.1 tests: (top) 45 wt.% Class C fly ash filler, (middle) 45 wt. % CaSiO_3 filler, and (bottom) 35 wt.% Class C fly ash + 10 wt.% slag

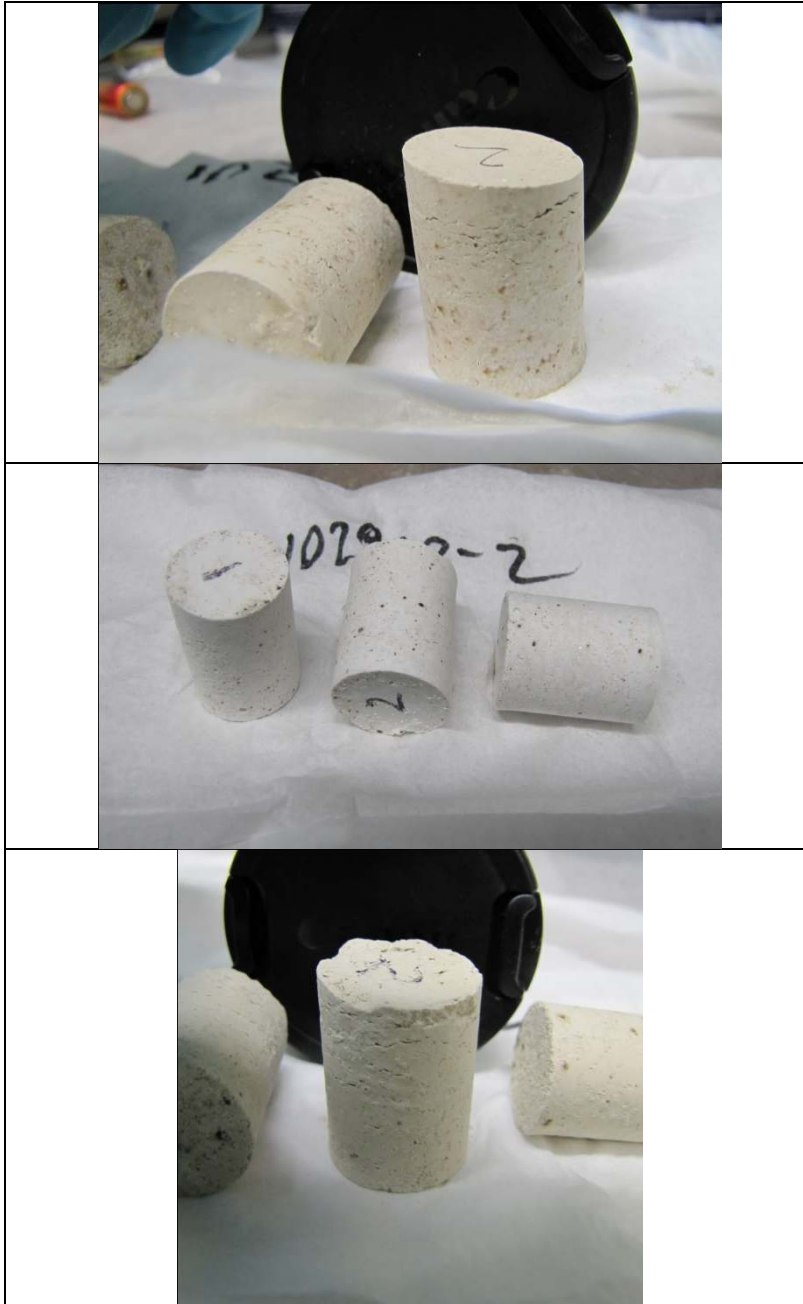


Figure 6-2. Waste forms with 15 wt.% waste baseline waste simulant loading after 90-day ANS 16.1 test: (top) 45 wt.% Class C fly ash, (middle) 45 wt. CaSiO_3 , and (bottom) 35 wt.% Class C fly ash + 10 wt.% slag

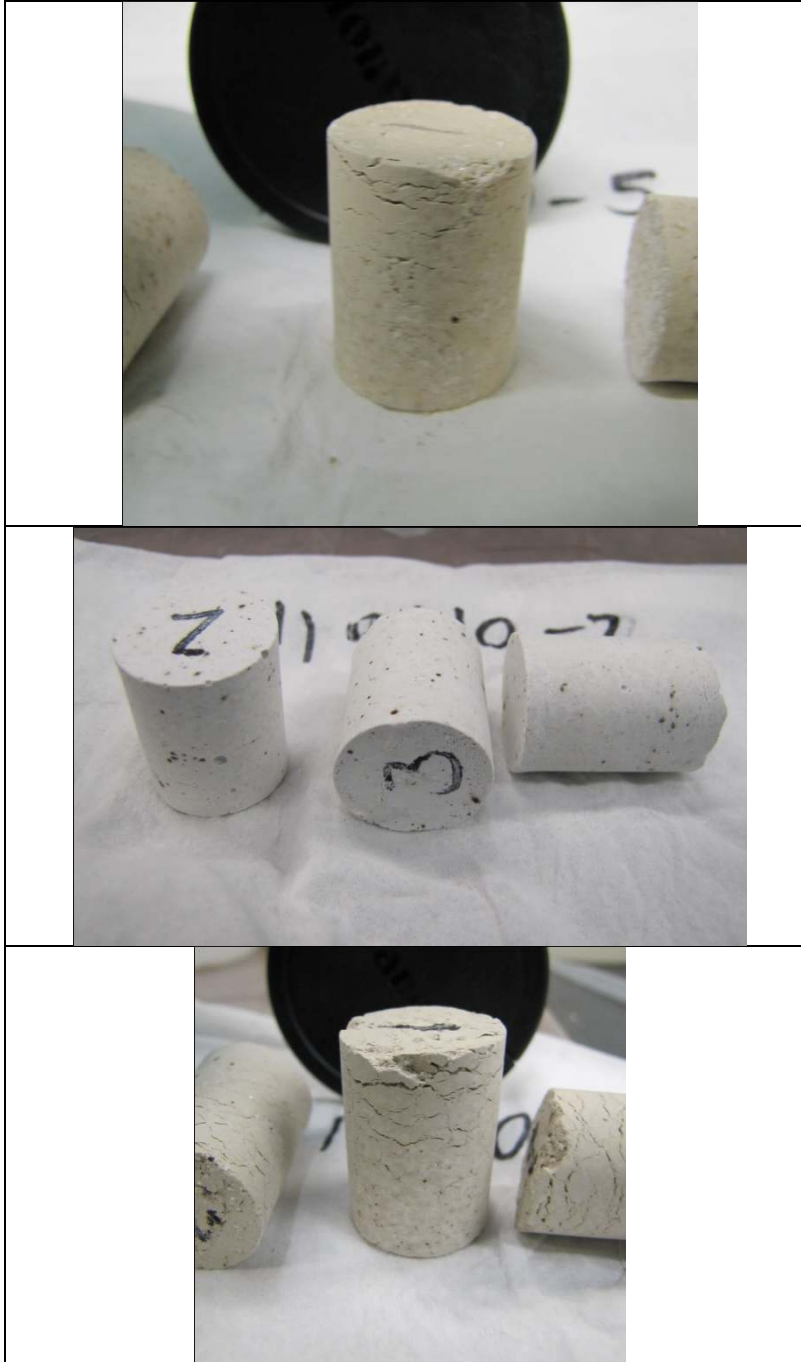


Figure 6-3. Waste forms with 15 wt.% waste cluster 1 waste simulant loading after 90-day ANS 16.1 test: (top) 45 wt.% Class C fly ash, (middle) 45 wt. CaSiO_3 , and (bottom) 35 wt.% Class C fly ash + 10 wt.% slag

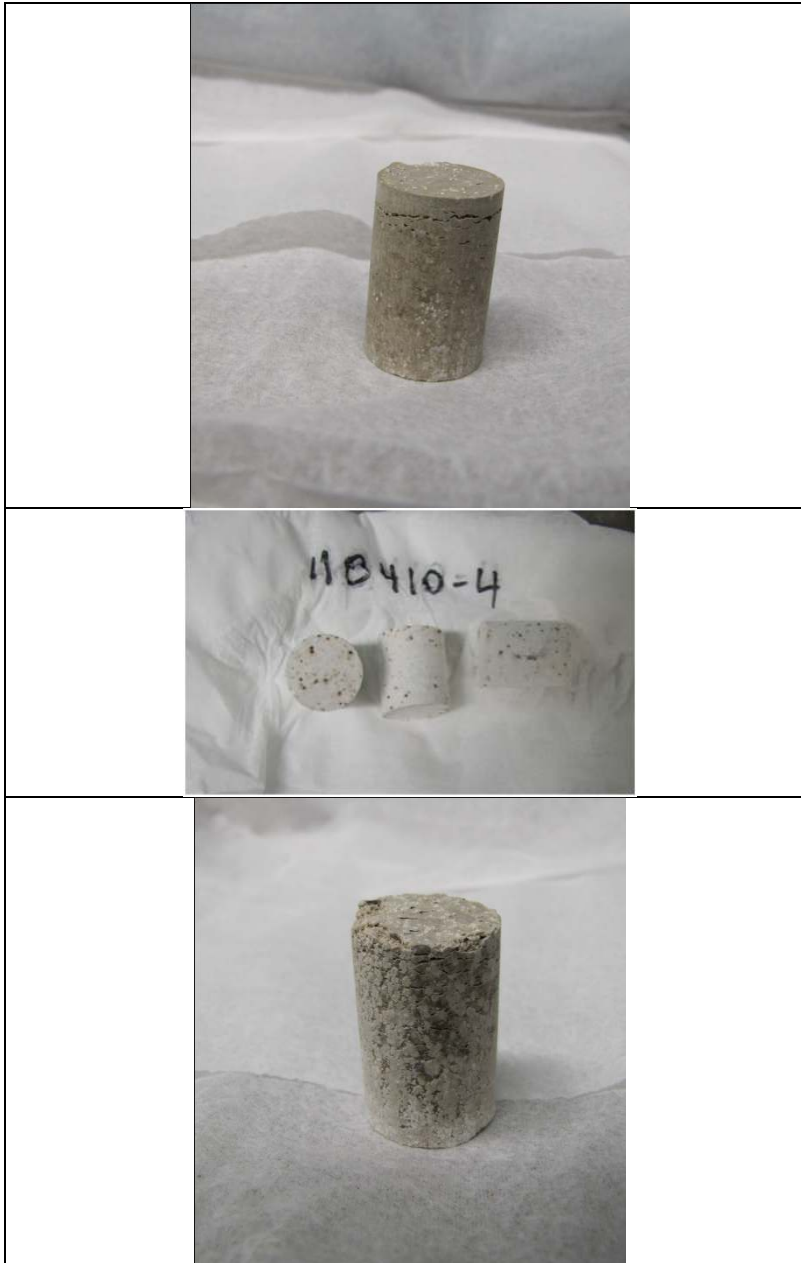


Figure 6-4. Waste forms with 15 wt.% waste cluster 2 waste simulant loading after 90-day ANS 16.1 test: (top) 45 wt.% Class C fly ash, (middle) 45 wt. CaSiO_3 , and (bottom) 35 wt.% Class C fly ash + 10 wt.% slag

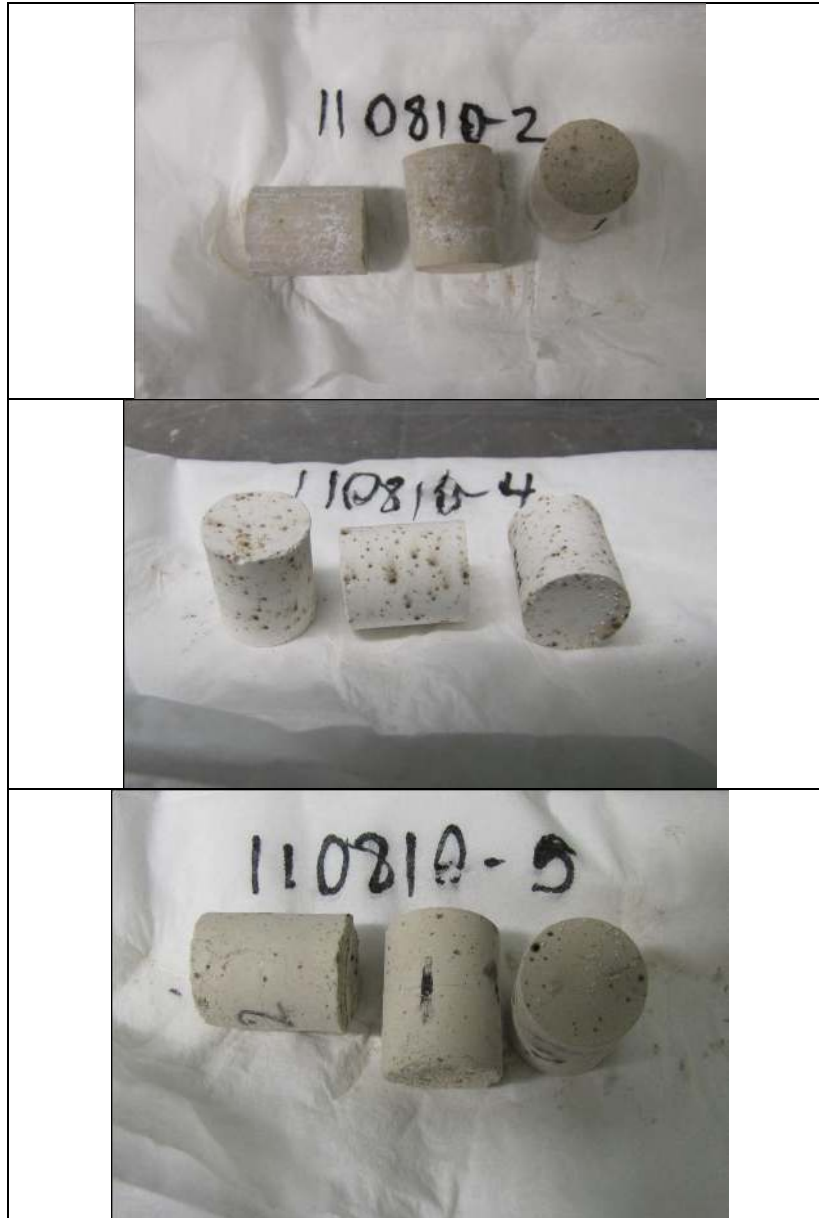


Figure 6-5. Waste forms with 15 wt.% waste mixed waste simulant loading after 90-day ANS 16.1 test: (top) 45 wt.% Class C fly ash, (middle) 45 wt. CaSiO_3 , and (bottom) 35 wt.% Class C fly ash + 10 wt.% slag

6.3 Summary

The following conclusions were reached from the ANS 16.1 tests:

- (a) There was no iodine detected in any of the leachate samples. The detection limit was 5 ppb. The calculated iodine LIs are based on taking 5 ppb as the iodine concentration in each leaching interval.
- (b) The Re LIs range from 7.9 to 9. There was no specific trend observed with respect to the waste simulant or the filler types. These values are somewhat lower than than the target LI value of 9. One possible way to enhance the LI of Re is to eliminate the swelling/cracking of the waste forms. As discussed before, this could be accomplished by eliminating the use of zeolite in the waste form mix.
- (c) The LIs for Mg, and Si are 10-12, indicative of the durability of the waste form. The LI for Mg in the waste forms appears to be better than that of the blank samples. LI for P is 8-9, indicative of some unreacted potassium phosphate in the waste forms.
- (d) The LI for Na exceeds the target value of 6 for all samples, indicative of containment of this specie.
- (e) Based on the ANS 16.1 tests, the waste forms are successful in containing the radionuclide surrogate species (Re and I) to as high as 20 wt.% waste simulant loadings.
- (f) After the 90-day ANS 16.1 tests, some waste forms showed degradation, particularly waste forms with fly ash + slag filler. This degradation is attributed to the zeolite additions. It is quite possible to eliminate the addition of zeolite in the various formulations since I in the leachate solutions was below the detection limits. However, additional study to evaluate I leaching in the absence of zeolite will be needed to confirm this hypothesis.

7 TOXICITY CHARACTERISTIC LEACHING PROCEDURE

7.1 Analysis of Hazardous Contaminants in the Waste Simulants

Table 7-1 lists the concentration of the various hazardous metals targeted in the four waste streams. Chromium, cadmium, and silver were significantly higher in the mixed waste as compared to the other three waste simulants.

Table 7-2 shows the concentrations of the hazardous metals in the waste streams as determined by ICP-MS (ANL's Analytical Chemistry Lab). For very low concentrations of metal there was a discrepancy in the targeted and observed concentrations. This discrepancy could be a result of the uncertainty in measuring very small amounts of metal salts (in tenths of a milligram), in spite of preparing a fairly large volume (~3.5 L) of the simulant. In particular, the discrepancy was observed for Ag and Cd additions to the baseline, cluster 1, and cluster 2 formulations. The second possibility could be some precipitation of the insoluble compounds of the hazardous elements as observed by some sedimentation in the prepared waste simulants (Fig. 3-2). Although, the waste streams were thoroughly mixed before preparing the waste forms, there is still a possibility of reduced hazardous element concentration from sedimentation. We are uncertain why the Pb concentration was low in the mixed waste simulant.

7.2 Summary

Table 7-3 lists the TCLP results for the eight metals conducted on blanks and the waste forms as reported from the GEL Labs.

Several key results to note:

- (a) All the results are below the Universal Treatment Standard limits (UTS).
- (b) Fly ash contained detectable levels of Cr and Se.
- (c) The "<" sign means that the metal concentration was below the detection limit.
- (d) For the waste forms fabricated with mixed waste simulants, where targeted amounts of Ag and Cd were present in the simulant, TCLP results were below the detection limit. It is rationalized that similar results would be expected for the baseline, cluster 1 and cluster 2 waste forms, although these waste streams did not have the target concentrations for Ag and Cd.
- (e) Lead is well contained for the baseline, cluster 1, and cluster 2 waste forms.
- (f) Containment of RCRA metals does not appear to be an issue at the waste stream loading levels studied.

Table 7-1. Hazardous metal concentrations targeted in waste simulants

Elements	Concentration (ppm)			
	Baseline	Cluster 1	Cluster 2	Mixed
Cr	11.38	11.42	11.31	58.49
Ag	0.62	0.63	0.62	2.27
As	2.44	2.44	2.42	1.07
Cd	0.17	0.17	0.17	2.17
Hg	2.00	2.01	1.99	0.90
Pb	1.75	1.76	1.74	1.54

Table 7-2. Hazardous metal concentrations in the in-house prepared waste simulants

Elements	Concentration (ppm)			
	Baseline	Cluster 1	Cluster 2	Mixed
Cr	9.32	11.30	10.00	46.5
Ag	0.0015	0.0013	0.0007	2.10
As	3.23	3.37	3.35	0.963
Cd	0.0069	0.078	0.0057	1.80
Hg	1.37	1.73	1.68	0.496
Pb	1.57	1.63	1.60	0.096

Table 7-3. TCLP results on various waste forms and blank samples (mg/L)

	As	Ba	Cd	Cr	Pb	Se	Ag	Hg
45FA Blank	0.183	<0.01	<0.01	0.375	<0.033	0.172	<0.01	<0.0007
45 CaSi Blank	<0.05	0.012	<0.01	0.014	<0.033	0.182	<0.01	<0.0007
35FA+10Slag Blank	0.074	0.011	<0.01	0.011	<0.033	0.268	<0.01	<0.0007
45FA+20WS(B)	0.15	<0.01	<0.01	0.013	<0.033	0.303	<0.01	<0.0007
45FA+15WS(B)	0.13	<0.01	<0.01	0.015	<0.033	0.129	<0.01	<0.0007
45CaSi+20WS(B)	<0.05	0.31	<0.01	0.016	<0.033	0.227	<0.01	<0.0007
45CaSi+15WS(B)	<0.05	<0.01	<0.01	0.019	<0.033	0.182	<0.01	<0.0007
35FA+10Slag+15WS(B)	0.15	<0.01	<0.01	0.016	<0.033	0.327	<0.01	<0.0007
35FA+10Slag+10WS(B)	0.14	0.01	<0.01	0.019	<0.033	0.397	<0.01	<0.0007
45FA+20WS(C1)	0.20	<0.01	<0.01	0.015	<0.033	0.262	<0.01	<0.0007
45FA+15WS(C1)	0.12	<0.01	<0.01	0.018	<0.033	0.457	<0.01	<0.0007
45CaSi+20WS(C1)	<0.05	<0.01	<0.01	<0.01	<0.033	0.226	<0.01	<0.0007
45CaSi+15WS(C1)	<0.05	<0.01	<0.01	0.011	<0.033	0.208	<0.01	<0.0007
35FA+10Slag+ 15WS(C1)	0.076	0.012	<0.01	0.020	<0.033	0.260	<0.01	<0.0007
35FA+10Slag+ 10WS(C1)	0.060	<0.01	<0.01	0.014	<0.033	<0.06	<0.01	<0.0007
45FA+20WS(C2)	0.14	0.011	<0.01	0.013	<0.033	0.297	<0.01	<0.0007
45FA+15WS(C2)	0.15	<0.01	<0.01	0.015	<0.033	0.173	<0.01	<0.0007
45CaSi+20WS(C2)	0.065	<0.01	<0.01	<0.01	<0.033	0.191	<0.01	<0.0007
45CaSi+15WS(C2)	<0.05	<0.01	<0.01	0.012	<0.033	0.085	<0.01	<0.0007
35FA+10Slag+ 15WS(C2)	0.31	0.019	<0.01	0.028	<0.033	0.398	<0.01	<0.0007
35FA+10Slag+ 10WS(C2)	0.25	0.013	<0.01	0.016	<0.033	0.283	<0.01	<0.0007
45FA+20WS(M)	0.096	0.011	<0.01	0.017	<0.033	0.267	<0.01	<0.0007
45FA+15WS(M)	0.182	<0.01	<0.01	0.011	<0.033	0.257	<0.01	<0.0007
45CaSi+20WS(M)	<0.05	<0.01	<0.01	0.0144	<0.033	0.219	<0.01	<0.0007
45CaSi+15WS(M)	<0.05	<0.01	<0.01	<0.01	<0.033	<0.06	<0.01	<0.0007
35FA+10Slag+ 10WS(M)	<0.05	<0.01	<0.01	0.011	<0.033	<0.06	<0.01	<0.0007
35FA+10Slag+ 15WS(M)	0.072	<0.01	<0.01	0.013	<0.033	0.138	<0.01	<0.0007
Universal Treatment Standard	5	21	0.11	0.6	0.75	5.7	0.14	0.025

8 SCALEUP STUDIES

A limited study was undertaken to understand the process variables for the setting of Ceramicrete on a more realistic waste form scale (2 gal). As part of this work, various formulations were tested to stabilize the baseline simulant. Specifically, we studied the following process parameters: (a) working time before pouring, (b) temperature rise during setting and curing of the waste form, and (c) appearance of the waste form.

Based on several laboratory-scale trials, we selected a dry powder formulation that consisted of 12.5 wt.% MgO, 42.4 wt.% KH_2PO_4 , 44.8 wt.% Class C fly ash, and 0.35 wt.% boric acid. To prepare a 2-gallon sample, 10 kg of the dry powder was mixed with 2.57 kg of baseline waste simulant. Approximately 350 g of additional water was used. Net waste loading was 20 wt.% of the simulant. Process steps were similar to those used before. First, 141 g of Ag-zeolite was added to the waste simulant and mixed for 3-5 minutes. Thereafter, the dry powders were added continuously as the slurry was mixed. Mixing continued for almost 35 minutes, and the slurry was poured into a plastic bucket, as shown in Figure 8-1. As the slurry was mixed, periodically (at 3-5 minute intervals) temperature was monitored. The slurry temperature rose to about 53°C before it was poured. A thermocouple was placed at the center of the waste form to monitor the temperature during the setting period.



Figure 8-1. Ceramicrete slurry formed during stabilization of baseline waste simulant at 20 wt.% loading



Figure 8-2. Waste form (2 gal) after 3 days of curing

Figure 8-2 shows the 2-gal waste form after 3 days of curing. Figure 8-3 shows it out of the bucket after only 4 days of curing. The temperature peaked around 56°C during the curing process and declined to ambient temperature in about 2 days. The waste form appeared to be uniform and dense. There was no visible appearance of bleed water. There was slight expansion on the top surface, as evident from the side view. Also, there was some residual white residue powder, probably unreacted MgO resulting from incomplete reaction/mixing or from the powder that was stored in the bucket that was used as a mold in this experiment. These findings clearly show that the process can be easily scaled up. However, some process optimization will be required.



Figure 8-3. Waste form (2 gal) after removal from the mold. Notice slight expansion on the top surface

9 CONCLUSIONS

The Ceramicrete process has been demonstrated to stabilize four secondary waste formulations that are expected to be generated as a result of treatment of Hanford liquid wastes. Based on the recipes provided by PNNL, the four waste simulants (baseline, cluster 1, cluster 2, and mixed) were prepared in-house. The waste forms were fabricated with three filler materials: Class C fly ash, CaSiO_3 , and Class C fly ash + slag. For the fly ash and CaSiO_3 , waste loadings were as high as 20 wt.%; for fly ash + slag filler, waste loadings were as high as 15 wt.%.

The samples were subjected to physical property characterizations, ANS 16.1 leach tests, and TCLP tests. For the physical property characterizations, the waste forms were fabricated with no additives, hazardous contaminants, or radionuclide surrogates. For the ANS 16.1 test samples, the waste simulants contained appropriate amounts of rhenium and iodine as radionuclide surrogates, and the additives were silver-loaded zeolite and tin chloride. The TCLP waste forms contained the requisite amounts of hazardous metals and potassium sulfide additive.

The physical property characterizations (density, compressive strength, and 90-day water immersion) showed that the waste forms are stable and durable. Compressive strengths were $>2,500$ psi, and these strengths were retained after 90-day water immersions test for the three waste simulants. Cluster 2 waste forms showed some drop in strength after the immersion tests, but they were still $>2,000$ psi. Fly ash and CaSiO_3 filler waste forms appeared to be superior to the waste forms with fly ash + slag. Waste form weight loss was ~ 5 -14 wt.%, which occurred during the initial phase of the immersion test, indicative of washing-off of residual unreacted powder.

The ANS 16.1 tests yielded a leachability index for Re of 7.9-9.0 for all the samples evaluated. Iodine was not detected in any of the leachate samples. Further, leaching of Na was low, as indicated by the leachability indices for Na ranging from 7.6-10.4.

There was degradation of the waste forms in the samples after the ANS 16.1 tests. Cracking of the samples was observed for some of the waste forms, particularly for the fly ash + slag and fly filler samples. Cracking could be possibly due to the reaction of zeolite to the potassium phosphate or to the sodium rich waste simulant. This cracking may be contributing to the LI's of the various species.

The TCLP results showed that all the hazardous contaminants are contained, and their concentration are below the UTS limits.

Preliminary scale-up of the process was conducted with 2-gal waste forms. Minimal amounts of boric acid as a set retarder were used to control the mixing time.

10 RECOMMENDATIONS

Based on the results and observations from this study, Ceramicrete technology has the potential to stabilize and contain the four secondary waste simulants investigated. Waste loadings as high as 20 wt.% can be attained. In terms of filler materials, Class C fly ash at 45 wt.%, on a dry powder basis, is adequate.

The present study was focused on 1 M Na concentration in the waste simulants. Higher Na concentrations (2M) are being investigated in a parallel project at PNNL.

The initial weight loss of the waste forms during 90-day water immersion test is attributed to the un-reacted binder phase materials such as potassium phosphate. The residual un-reacted binder phase needs to be minimized. This could be accomplished by improving slurry mixing, optimizing particle size and reactivity of binder phase starting powders (especially, magnesium oxide), and increasing curing times before evaluation.

Leachability indices of Re need to be improved to meet or exceed the target value of 9 for all the waste stream formulations. Since Re is present in the waste simulant in its highly mobile ReO_4^- form, it may be prudent to first reduce it to Re (+4) state and then incorporate the waste stream in the Ceramicrete. Eh-pH conditions of the slurry need to be optimized for Re containment. Use of other reducing agent(s) needs to be explored for improving Re LI. Further, any improvement in minimizing the initial weight loss of the waste form to water exposure will help in improving the containment of Re.

It appears that zeolite bead addition is contributing to cracking of some of the waste forms post-leaching test. Since iodine is below detection limit for all leaching intervals, optimization of zeolite loading or its complete elimination from the process needs to be explored. Preventing any cracking during the leaching tests will also help in improving the LI of contaminants as well.

It appears that reaction of acid to the carbonates and bicarbonates in the waste simulant can generate CO_2 that can lead to expansion of the waste form. To minimize expansion effects, especially for larger scale waste forms, pretreatment of the waste simulants to break down the carbonates/bi-carbonates prior to their incorporation in Ceramicrete would be needed.

For scale-up of the waste forms, use of small amounts (0.5 wt.% or less on dry powder basis) of boric acid is recommended. Use of boric acid retards the phosphate binder reactions and extends the working time of the slurry.

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APPENDIX A: COMPRESSIVE STRENGTH DATA

A.1 As-fabricated Waste Forms

A.1.1 Baseline Waste Stream

Waste Simulant	Baseline	
Sample Composition (16-20 days)	Average Compressive Strength (psi)	STDEV
45FA+20W	2698	1032
45FA+15W	2595	560
45FA+10W	3239	175
45FA+5W	4479	616
45FABlank	3680	1584
25FA+25W	1113	415
25FABlank	1317	420
25FA+25W+1B.A.	1215	134

Waste Simulant	Baseline	
Sample Composition (16-20 days)	Average Compressive Strength (psi)	STDEV
45CaSi+20W	4794	126
45CaSi+15W	3968	300
45CaSiBlank	4423	651
25CaSi+25W	2768	553
25CaSiBlank	3044	760

Waste Simulant	Baseline	
Sample Composition (16-20 days)	Average Compressive Strength (psi)	STDEV
35FA+10Slg+10W	3623	703
35FA+10SlgBlank	5378	266
35FA+10Slg+15W	3076	314

Waste Simulant	Baseline	
Sample Composition (30-35 days)	Average Compressive Strength (psi)	STDEV
45FA+20W	3203	691
45FA+15W	3755	1753
45FA+10W	4332	132
45FA+5W	4150	602
45FABlank	4582	1546
25FA+25W	1321	398
25FABlank	2351	438
25FA+25W+1B.A.	1358	42

Waste Simulant	Baseline	
Sample Composition (30-35 days)	Average Compressive Strength (psi)	STDEV
45CaSi+20W	6196	1342
45CaSi+15W	5473	811
45CaSiBlank	5760	1029
25CaSi+25W	3152	968
25CaSiBlank	4454	1077

Waste Simulant	Baseline	
Sample Composition (30-35 days)	Average Compressive Strength (psi)	STDEV
35FA+10Slg+10W	4834	1035
35FA+10SlgBlank	7376	0
35FA+10Slg+15W	4103	586

Waste Simulant	Baseline	
Sample Composition (60 days)	Average Compressive Strength (psi)	STDEV
45FA+20W	4260.013	541.70
45FA+15W	5582.71	346.92
45FA+10W	4567.86	856.58
45FA+5W	5555.03	1033.89
45FABlank	4189.45	376.01
25FA+25W	1582.46	292.03
25FABlank	1235.87	27.88
25FA+25W+1B.A.	1819.05	33.75

Waste Simulant	Baseline	
Sample Composition (60 days)	Average Compressive Strength (psi)	STDEV
45CaSi+20W	5901.27	192.24
45CaSi+15W	4994.34	92.45
45CaSiBlank	5036.89	196.64
25CaSi+25W	2524.67	547.38
25CaSiBlank	3217.84	441.72

Waste Simulant	Baseline	
Sample Composition (60 days)	Average Compressive Strength (psi)	STDEV
35FA+10Slg+10W	3726.65	1314.84
35FA+10SlgBlank	5681.63	2162.22
35FA+10Slg+15W	3018.54	996.15

A.1.2 Cluster 1 Waste Stream

Waste Simulant	Cluster 1	
Sample Composition (16-19 days)	Average Compressive Strength (psi)	STDEV
45FA+20W	2275	485
45FA+15W	3193	562
45FA+10W	3960	1230
45FA+5W	3075	733
45FABlank	3680	1584
25FA+25W	1631	180
25FABlank	1317	420
25FA+25W+1B.A.	2242	702

Waste Simulant	Cluster 1	
Sample Composition (16-19 days)	Average Compressive Strength (psi)	STDEV
45CaSi+20W	4712	615
45CaSi+15W	4381	985
45CaSiBlank	4423	651
25CaSi+25W	2873	268
25CaSiBlank	3044	760

Waste Simulant	Cluster 1	
Sample Composition (16-19 days)	Average Compressive Strength (psi)	STDEV
35FA+10Slg+10W	4806	585
35FA+10SlgBlank	5378	266
35FA+10Slg+15W	2760	360

Waste Simulant	Cluster 1	
Sample Composition (30-39 days)	Average Compressive Strength (psi)	STDEV
45FA+20W	4259	639
45FA+15W	6477	0
45FA+10W	4820	687
45FA+5W	4018	1395
45FABlank	4582	1546
25FA+25W	1376	571
25FABlank	2351	439
25FA+25W+1B.A.	2181	712

Waste Simulant	Cluster 1	
Sample Composition (30-39 days)	Average Compressive Strength (psi)	STDEV
45CaSi+20W	3632	154
45CaSi+15W	4932	736
45CaSiBlank	5760	1029
25CaSi+25W	3540	1477
25CaSiBlank	4454	1076

Waste Simulant	Cluster 1	
Sample Composition (30-39 days)	Average Compressive Strength (psi)	STDEV
35FA+10Slg+10W	5005	1146
35FA+10SlgBlank	7376	0
35FA+10Slg+15W	3503	791

Waste Simulant	Cluster 1	
Sample Composition (90-96 days)	Average Compressive Strength (psi)	STDEV
45FA+20W	3798.94	1900.41
45FA+15W	3909.97	2201.25
45FA+10W	4837.65	2365.61
45FA+5W	3764.69	1297.27
45FABlank	NT	NT
25FA+25W	1532.65	465.20
25FABlank	NT	NT
25FA+25W+1B.A.	2122.40	171.42

Waste Simulant	Cluster 1	
Sample Composition (90-96 days)	Average Compressive Strength (psi)	STDEV
45CaSi+20W	4391.45	954.15
45CaSi+15W	4672.32	789.62
45CaSiBlank	NT	NT
25CaSi+25W	3547.82	89.52
25CaSiBlank	NT	NT

Waste Simulant	Cluster 1	
Sample Composition (90-96 days)	Average Compressive Strength (psi)	STDEV
35FA+10Slg+10W	4959.06	1131.44
35FA+10SlgBlank	NT	NT
35FA+10Slg+15W	2378.36	835.79

A.1.3 Cluster 2 Waste Simulant

Waste Simulant	Cluster 2	
Sample Composition (21-30 days)	Average Compressive Strength (psi)	STDEV
45FA+20W	4005	167
45FA+15W	4483	569
45FA+10W	3269	368
45FA+5W	2186	579
45FABlank	4582	1546
25FA+25W	1370	431
25FABlank	2351	439
25FA+25W+1B.Acid	840	224

Waste Simulant	Cluster 2	
Sample Composition (21-30 days)	Average Compressive Strength (psi)	STDEV
45CaSi+20W	4950	355
45CaSi+15W	4230	868
45CaSiBlank	5760	1029
25CaSi+25W	2618	115
25CaSiBlank	4454	1076

Waste Simulant	Cluster 2	
Sample Composition (21-30 days)	Average Compressive Strength (psi)	STDEV
35FA+10Slg+10W	4039	304
35FA+10SlgBlank	7376	0
35FA+10Slg+15W	2201	426

A.1.4 Mixed Waste Stream Simulant

Waste Simulant	Mixed	
Sample Composition (16-20 days)	Average Compressive Strength (psi)	STDEV
45FA+20W	2402.71	720.64
45FA+15W	3577.22	1621.85
45FA+10W	3130.33	415.16
45FA+5W	3100.58	519.98
45FABlank	3680.02	1584.28
25FA+25W	1874.05	249.46
25FABlank	1316.42	419.38
25FA+25W+1B.Acid	146.60	65.67

Waste Simulant	Mixed	
Sample Composition (16-20 days)	Average Compressive Strength (psi)	STDEV
45CaSi+20W	3295.66	572.70
45CaSi+15W	4468.93	601.56
45CaSiBlank	4421.85	651.17
25CaSi+25W	2792.04	170.03
25CaSiBlank	3042.88	760.00

Waste Simulant	Mixed	
Sample Composition (16-20 days)	Average Compressive Strength (psi)	STDEV
35FA+10Slg+10W	2631.27	1712.00
35FA+10SlgBlank	5378	266
35FA+10Slg+15W	1874.74	997.14

Waste Simulant	Mixed	
Sample Composition (30-40 days)	Average Compressive Strength (psi)	STDEV
45FA+20W	4561	1907.42
45FA+15W	3891.98	1873.33
45FA+10W	4299.44	1586.69
45FA+5W	2626.02	756.44
45FABlank	4581.60	1546.92

A.2 Post Water Immersion

A.2.1 Baseline Waste Stream Simulant

Waste Simulant	Baseline	
Sample Composition (30 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
45FA+20W	3095	652
45FA+15W	2974	51
45FA+10W	4102	666
45FA+5W	3049	676
45FABlank	3582	326
25FA+25W	1138	115
25FABlank	900	160
25FA+25W+1B.A.	748	180

Waste Simulant	Baseline	
Sample Composition (30 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
45CaSi+20W	4299	578
45CaSi+15W	3943	333
45CaSiBlank	3743	410
25CaSi+25W	3434	170
25CaSiBlank	2977	714

Waste Simulant	Baseline	
Sample Composition (30 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
35FA+10Slg+10W	3784	1758
35FA+10SlgBlank	2878	483
35FA+10Slg+15W	3393	87

Waste Simulant	Baseline	
Sample Composition (60 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
45FA+20W	3353.77	375.93
45FA+15W	3329.56	1238.92
45FA+10W	4209.51	308.74
45FA+5W	2801.73	792.13
45FABlank	3087.89	839.05
25FA+25W	942.21	190.92
25FABlank	1200.80	411.34
25FA+25W+1B.A.	1430.61	276.19

Waste Simulant	Baseline	
Sample Composition (60 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
45CaSi+20W	2999.58	464.46
45CaSi+15W	4101.59	1110.40
45CaSiBlank	4412.19	610.71
25CaSi+25W	3104.04	848.77
25CaSiBlank	3709.43	877.06

Waste Simulant	Baseline	
Sample Composition (60 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
35FA+10Slg+10W	4531.19	926.77
35FA+10SlgBlank	6116.87	852.72
35FA+10Slg+15W	2945.97	68.97

Waste Simulant	Baseline	
Sample Composition (90 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
45FA+20W	3948.36	1645.07
45FA+15W	3205.04	1867.71
45FA+10W	4172.85	662.82
45FA+5W	2944.24	702.23
45FABlank	3082.60	911.01
25FA+25W	1324.77	308.19
25FABlank	1400.86	85.11
25FA+25W+1B.A.	NT	NT

Waste Simulant	Baseline	
Sample Composition (90 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
45CaSi+20W	5051.42	537.22
45CaSi+15W	4336.80	276.15
45CaSiBlank	4845.26	1123.90
25CaSi+25W	2995.43	1219.98
25CaSiBlank	2187.42	680.92

Waste Simulant	Baseline	
Sample Composition (90 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
35FA+10Slg+10W	NT	NT
35FA+10SlgBlank	4742.19	700.39
35FA+10Slg+15W	3083.99	1926.03

A.2.2 Cluster 1 Waste Simulant

Waste Simulant	Cluster 1	
Sample Composition (30 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
45FA+20W	3167	924
45FA+15W	3840	1700
45FA+10W	3244	383
45FA+5W	2500	3
45FABlank	3582	326
25FA+25W	825	1
25FABlank	900	160
25FA+25W+1B.A.	1465	263

Waste Simulant	Cluster 1	
Sample Composition (30 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
45CaSi+20W	3625	1656
45CaSi+15W	2925	264
45CaSiBlank	3743	410
25CaSi+25W	3170	918
25CaSiBlank	2977	714

Waste Simulant	Cluster 1	
Sample Composition (30 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
35FA+10Slg+10W	3617	148
35FA+10SlgBlank	2878	483
35FA+10Slg+15W	3222	886

Waste Simulant	Cluster 1	
Sample Composition (60 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
45FA+20W	2501.38	235.98
45FA+15W	3788.38	838.44
45FA+10W	5349.24	642.42
45FA+5W	3523.51	1432.19
45FABlank	3087.89	839.05
25FA+25W	1093.36	209.81
25FABlank	1200.80	411.34
25FA+25W+1B.A.	2317.43	164.31

Waste Simulant	Cluster 1	
Sample Composition (60 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
45CaSi+20W	2991.70	208.59
45CaSi+15W	3874.14	753.66
45CaSiBlank	4412.19	610.71
25CaSi+25W	2815.35	678.18
25CaSiBlank	3709.43	877.06

Waste Simulant	Cluster 1	
Sample Composition (60 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
35FA+10Slg+10W	6734.44	1226.22
35FA+10SlgBlank	6116.87	852.72
35FA+10Slg+15W	1807.75	1358.61

Waste Simulant	Cluster 1	
Sample Composition (90 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
45FA+20W	3306.73	464.88
45FA+15W	3227.87	624.79
45FA+10W	4996.76	2114.32
45FA+5W	2973.98	842.97
45FABlank	3082.59	911.01
25FA+25W	603.24	293.43
25FABlank	1400.86	85.11
25FA+25W+1B.A.	NT	NT

Waste Simulant	Cluster 1	
Sample Composition (90 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
45CaSi+20W	4370.70	1065.77
45CaSi+15W	3220.26	647.57
45CaSiBlank	4845.26	1123.90
25CaSi+25W	2549.92	294.15
25CaSiBlank	2187.42	680.92

Waste Simulant	Cluster 1	
Sample Composition (90 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
35FA+10Slg+10W	5596.54	2184.95
35FA+10SlgBlank	4742.19	700.39
35FA+10Slg+15W	3522.57	777.30

A.2.3 Cluster 2 Waste Simulant

Waste Simulant	Cluster 2	
Sample Composition (30 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
45FA+20W	2440.53	320.03
45FA+15W	3466.80	559.21
45FA+10W	1518.67	312.37
45FA+5W	2376.90	517.32
45FABlank	3582.30	325.91
25FA+25W	1179.81	119.85
25FABlank	900.41	159.91
25FA+25W+1B.Acid	456.43	154.52

Waste Simulant	Cluster 2	
Sample Composition (30 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
45CaSi+20W	3867.91	290.36
45CaSi+15W	3594.05	489.04
45CaSiBlank	3742.74	410.12
25CaSi+25W	2400.41	890.98
25CaSiBlank	2977.19	714.23

Waste Simulant	Cluster 2	
Sample Composition (30 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
35FA+10Slg+10W	2364.45	429.95
35FA+10SlgBlank	2877.59	483.11
35FA+10Slg+15W	1412.86	419.79

Waste Simulant	Cluster 2	
Sample Composition (60 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
45FA+20W	1612.03	299.27
45FA+15W	2695.02	126.16
45FA+10W	2124.48	35.21
45FA+5W	2271.78	264.06
45FABlank	3087.89	839.05
25FA+25W	1207.47	167.24
25FABlank	1200.80	411.34
25FA+25W+1B.Acid	615.15	203.92

Waste Simulant	Cluster 2	
Sample Composition (60 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
45CaSi+20W	3807.05	202.45
45CaSi+15W	4146.61	1097.87
45CaSiBlank	4412.19	610.71
25CaSi+25W	1926.69	223.97
25CaSiBlank	4412.19	610.71

Waste Simulant	Cluster 2	
Sample Composition (60 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
35FA+10Slg+10W	1973.03	783.39
35FA+10SlgBlank	6116.87	852.72
35FA+10Slg+15W	1477.18	403.15

Waste Simulant	Cluster 2	
Sample Composition (90 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
45FA+20W	2296.68	257.29
45FA+15W	2759.34	537.25
45FA+10W	2515.21	570.09
45FA+5W	2203.32	281.19
45FABlank	3082.59	911.01
25FA+25W	1178.42	96.69
25FABlank	1400.86	85.11
25FA+25W+1B.Acid	840.94	223.45

Waste Simulant	Cluster 2	
Sample Composition (90 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
45CaSi+20W	3233.75	642.93
45CaSi+15W	4053.25	323.52
45CaSiBlank	4845.26	1123.90
25CaSi+25W	2401.80	799.74
25CaSiBlank	2187.42	680.92

Waste Simulant	Cluster 2	
Sample Composition (90 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
35FA+10Slg+10W	2177.73	238.04
35FA+10SlgBlank	4742.19	700.39
35FA+10Slg+15W	1537.34	474.70

A.2.4 Mixed Waste Simulant

Waste Simulant	Mixed	
Sample Composition (30 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
45FA+20W	2598.20	585.78
45FA+15W	2430.84	1589.88
45FA+10W	4029.74	1072.94
45FA+5W	2878.98	796.78
45FABlank	3582.30	325.91
25FA+25W	1976.49	376.12
25FABlank	900.41	159.91
25FA+25W+1B.Acid	19.16	33.18

Waste Simulant	Mixed	
Sample Composition (30 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
45CaSi+20W	4061.55	754.75
45CaSi+15W	3672.20	560.18
45CaSiBlank	3742.74	410.11
25CaSi+25W	1908.71	1109.83
25CaSiBlank	2977.18	714.23

Waste Simulant	Mixed	
Sample Composition (30 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
35FA+10Slg+10W	3315.35	152.84
35FA+10SlgBlank	2877.59	483.11
35FA+10Slg+15W	1895.57	902.73

Waste Simulant	Mixed	
Sample Composition (60 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
45FA+20W	3078.15	585.78
45FA+15W	3352.70	1589.88
45FA+10W	4625.17	1072.94
45FA+5W	2739.97	796.78
45FABlank	3087.89	839.05
25FA+25W	4154.56	597.08
25FABlank	1200.80	411.34
25FA+25W+1B.Acid	242.48	112.61

Waste Simulant	Mixed	
Sample Composition (60 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
45CaSi+20W	3912.17	782.46
45CaSi+15W	3551.18	923.47
45CaSiBlank	4412.19	610.71
25CaSi+25W	1939.83	5.87
25CaSiBlank	3709.43	877.06

Waste Simulant	Mixed	
Sample Composition (60 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
35FA+10Slg+10W	1949.52	890.53
35FA+10SlgBlank	6116.87	852.72
35FA+10Slg+15W	2597.51	1081.94

Waste Simulant	Mixed	
Sample Composition (90 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
45FA+20W	3173.58	1204.31
45FA+15W	2585.75	1100.25
45FA+10W	3266.25	483.93
45FA+5W	3966.11	1148.34
45FABlank	3082.59	911.01
25FA+25W	2472.34	548.13
25FABlank	1400.86	85.11
25FA+25W+1B.Acid	196.40	62.73

Waste Simulant	Mixed	
Sample Composition (90 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
45CaSi+20W	3172.89	184.22
45CaSi+15W	3334.02	1443.18
45CaSiBlank	4845.26	1123.90
25CaSi+25W	1773.86	1043.14
25CaSiBlank	2187.42	680.92

Waste Simulant	Mixed	
Sample Composition (90 days H ₂ O Immersion)	Average Compressive Strength (psi)	STDEV
35FA+10Slg+10W	2671.51	648.00
35FA+10SlgBlank	4742.19	700.39
35FA+10Slg+15W	1186.72	225.47

(NT in above tables denotes as Not Tested)

APPENDIX B: WEIGHT LOSS DATA

Sample Composition	Waste Stream	Weight Loss (%-1 Month)	STDEV (1 Month)	Weight Loss (%-2 Month)	STDEV (2 Month)	Weight Loss (%-3 Month)	STDEV (3 Month)
45FA+20W	Baseline	8.93	0.48	9.63	0.59	10.10	0.36
45FA+15W	Baseline	8.27	0.68	9.50	0.35	9.22	0.92
45FA+10W	Baseline	7.61	0.43	9.03	0.28	8.56	0.67
45FA+5W	Baseline	6.98	0.19	8.74	0.46	8.70	0.59
45FABlank	Blank	7.39	0.13	8.05	0.90	7.83	0.86
45CaSi+20W	Baseline	11.00	0.02	11.87	0.08	12.27	0.070
45CaSi+15W	Baseline	11.09	0.08	11.94	0.24	12.28	0.07
45CaSiBlank	Blank	9.80	0.05	10.70	0.04	10.94	0.16
25CaSi+25W	Baseline	10.39	0.10	10.45	0.36	11.09	0.35
25CaSiBlank	Blank	8.43	0.13	8.89	0.25	9.20	0.46
25FA+25W	Baseline	12.86	0.44	12.51	1.10	12.18	1.14
25FABlank	Blank	11.69	0.39	13.11	0.14	13.24	0.54
25FA+25W+1B.Acid	Baseline	12.24	0.22	14.18	1.29	12.58	0.72
35FA+10Slg+10W	Baseline	6.26	0.70	6.81	0.42	7.35	0.86
35FA+10SlgBlank	Blank	6.29	0.14	7.38	0.71	7.23	0.80
35FA+10Slg+15W	Baseline	6.62	0.77	8.66	0.34	8.27	0.80

Sample Composition	Waste Stream	Weight Loss (%-1 Month)	STDEV (1 Month)	Weight Loss (%-2 Month)	STDEV (2 Month)	Weight Loss (%-3 Month)	STDEV (3 Month)
45FA+20W	Cluster 1	7.48	0.34	7.50	0.37	7.62	0.31
45FA+15W	Cluster 1	7.079	0.78	6.35	0.36	6.33	0.18
45FA+10W	Cluster 1	5.17	0.24	4.46	0.09	4.31	0.20
45FA+5W	Cluster 1	6.82	0.31	7.34	0.83	7.84	1.51
45FABlank	Blank	6.91	0.31	7.32	0.61	7.63	0.56
45CaSi+20W	Cluster 1	11.16	0.04	11.82	0.82	12.32	0.24
45CaSi+15W	Cluster 1	10.93	0.04	11.89	0.08	12.68	0.30
45CaSiBlank	Blank	10.18	0.072	11.38	0.12	12.11	0.58
25CaSi+25W	Cluster 1	11.35	0.48	12.29	0.39	12.50	0.51
25CaSiBlank	Blank	8.78	0.21	9.67	0.34	10.14	0.61
25FA+25W	Cluster 1	13.77	0.74	14.71	0.20	14.89	0.22
25FABlank	Blank	12.23	0.52	11.37	1.37	12.13	0.82
25FA+25W+1B.Acid	Cluster 1	14.30	0.49	15.32	0.80	15.06	0.24
35FA+10Slg+10W	Cluster 1	4.93	0.98	5.51	0.42	5.66	0.60
35FA+10SlgBlank	Blank	6.29	0.14	7.38	0.71	7.23	0.80
35FA+10Slg+15W	Cluster 1	6.12	0.20	7.31	0.44	6.81	0.09

Sample Composition	Waste Stream	Weight Loss (%-1 Month)	STDEV (1 Month)	Weight Loss (%-2 Month)	STDEV (2 Month)	Weight Loss (%-3 Month)	STDEV (3 Month)
45FA+20W	Cluster 2	9.81	0.61	8.70	0.18	8.47	0.82
45FA+15W	Cluster 2	8.87	0.34	7.82	0.33	8.02	0.08
45FA+10W	Cluster 2	9.02	1.84	9.06	0.30	8.21	1.35
45FA+5W	Cluster 2	8.85	1.79	7.32	0.66	6.56	1.02
45FABlank	Blank	7.39	0.13	8.05	0.59	7.83	0.86
45CaSi+20W	Cluster 2	11.97	0.06	12.04	0.16	12.59	0.13
45CaSi+15W	Cluster 2	11.83	0.86	12.00	0.38	12.66	0.06
45CaSiBlank	Blank	9.80	0.05	10.70	0.04	10.94	0.16
25CaSi+25W	Cluster 2	11.75	0.23	11.44	0.69	13.61	2.77
25CaSiBlank	Blank	8.43	0.13	8.89	0.08	9.20	0.46
25FA+25W	Cluster 2	11.62	1.82	14.93	5.96	13.44	0.84
25FABlank	Blank	11.69	0.39	13.11	0.08	13.24	0.54
25FA+25W+1B.Acid	Cluster 2	11.32	3.83	10.65	5.10	17.42	7.42
35FA+10Slg+10W	Cluster 2	5.99	1.33	6.89	0.35	5.78	0.93
35FA+10SlgBlank	Blank	6.29	0.14	7.38	0.71	7.23	0.80
35FA+10Slg+15W	Cluster 2	7.62	0.59	7.30	0.27	7.89	0.54

Sample Composition	Waste Stream	Weight Loss (%-1 Month)	STDEV (1 Month)	Weight Loss (%-2 Month)	STDEV (2 Month)	Weight Loss (%-3 Month)	STDEV (3 Month)
45FA+20W	Mixed	10.35	0.47	8.67	0.49	9.63	0.69
45FA+15W	Mixed	10.20	0.63	9.09	1.08	8.65	1.34
45FA+10W	Mixed	9.33	0.54	8.28	0.15	8.79	0.92
45FA+5W	Mixed	9.47	0.41	8.45	0.83	8.79	0.72
45FABlank	Blank	7.39	0.13	8.05	0.59	7.83	0.86
45CaSi+20W	Mixed	13.01	0.18	14.87	0.11	14.38	0.12
45CaSi+15W	Mixed	12.00	0.42	13.78	0.14	13.38	0.46
45CaSiBlank	Blank	9.80	0.05	10.70	0.04	10.94	0.16
25CaSi+25W	Mixed	12.56	1.28	14.27	1.24	12.21	1.33
25CaSiBlank	Blank	8.43	0.13	8.89	0.08	9.20	0.46
25FA+25W	Mixed	12.24	1.58	18.89	12.63	11.06	0.31
25FABlank	Blank	11.69	0.39	13.11	0.08	13.24	0.54
25FA+25W+1B.Acid	Mixed	20.67	1.63	17.13	4.34	20.80	0.24
35FA+10Slg+10W	Mixed	9.19	0.739	10.149	1.04	8.61	0.24
35FA+10SlgBlank	Blank	6.29	0.14	7.38	0.71	7.23	0.80
35FA+10Slg+15W	Mixed	10.00	1.27	10.33	0.84	10.88	0.45

APPENDIX C: DIFFUSIVITIES OF SELECTED CERAMICRETE WASTE FORMS AS OBTAINED FROM ANS 16.1 TEST

45% FA + 20% Baseline Waste (average value from two samples)

Leach Interval (h)	Cumulative Leach Duration (h)	D_{Mg} (cm^2/s)	D_{Na} (cm^2/s)	D_P (cm^2/s)	D_{Si} (cm^2/s)	D_{Ag} (cm^2/s)	D_{Re} (cm^2/s)
2	2	1.2E-10	7.4E-08	2.2E-08	3.7E-13	9.6E-16	8.5E-10
5	7	1.9E-10	7.2E-08	2.6E-08	8.0E-13	1.3E-15	1.0E-09
17	24	1.2E-10	4.9E-08	1.7E-08	9.3E-13	3.8E-16	1.3E-09
24	48	7.0E-11	3.3E-08	1.3E-08	1.1E-12	4.7E-16	2.1E-09
24	72	4.6E-11	2.4E-08	1.1E-08	1.3E-12	7.9E-16	3.1E-09
24	96	2.5E-11	1.6E-08	7.7E-09	1.2E-12	1.1E-15	3.9E-09
24	120	1.7E-11	1.2E-08	6.3E-09	1.3E-12	1.4E-15	4.8E-09
336	456	1.1E-12	2.1E-09	1.1E-09	2.3E-13	1.8E-15	5.7E-09
672	1128	1.5E-13	2.4E-10	2.3E-10	1.1E-13	1.8E-15	5.3E-09
1032	2160	3.4E-13	5.4E-11	9.2E-11	1.0E-13	8.2E-14	3.2E-09

45% FA + 20% Cluster 1 Waste (average value from two samples)

Leach Interval (h)	Cumulative Leach Duration (h)	D_{Mg} (cm^2/s)	D_{Na} (cm^2/s)	D_P (cm^2/s)	D_{Si} (cm^2/s)	D_{Ag} (cm^2/s)	D_{Re} (cm^2/s)
2	2	2.5E-11	3.2E-08	8.6E-09	2.7E-13	9.2E-17	4.2E-10
5	7	5.8E-11	3.9E-08	1.1E-08	5.0E-13	1.2E-16	7.2E-10
17	24	4.4E-11	3.0E-08	9.6E-09	6.2E-13	3.6E-17	1.3E-09
24	48	3.2E-11	2.1E-08	7.3E-09	7.3E-13	4.5E-17	2.0E-09
24	72	2.2E-11	1.9E-08	6.5E-09	9.1E-13	7.6E-17	3.0E-09
24	96	1.3E-11	1.3E-08	4.7E-09	9.1E-13	1.1E-16	3.2E-09
24	120	1.1E-11	9.9E-09	3.8E-09	1.0E-12	1.4E-16	3.5E-09
336	456	1.7E-12	2.6E-09	9.1E-10	2.3E-13	6.7E-17	3.9E-09
672	1128	7.5E-13	3.8E-10	2.9E-10	1.0E-13	1.5E-16	2.7E-09

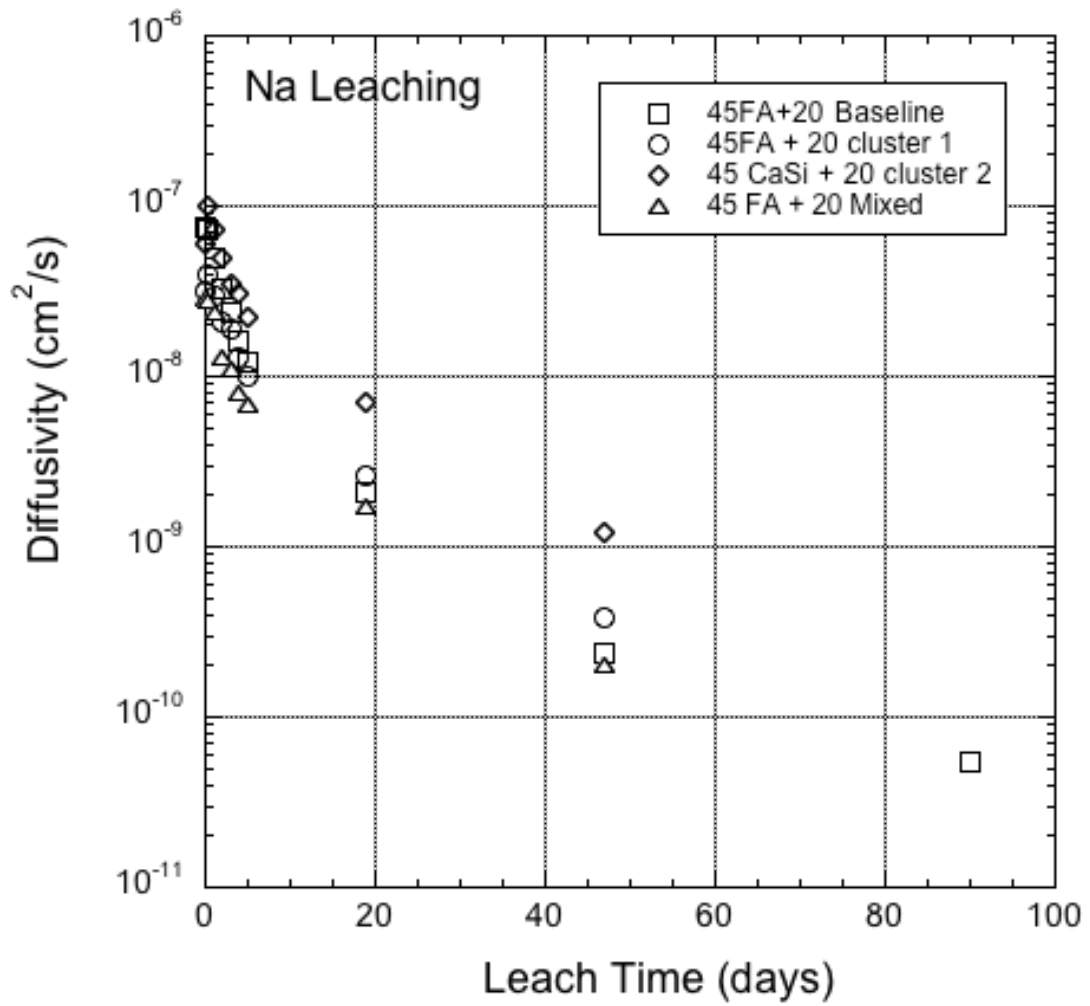
45% CaSi + 20% Cluster 2 Waste (average value from two samples)

Leach Interval (h)	Cumulative Leach Duration (h)	D_{Mg} (cm^2/s)	D_{Na} (cm^2/s)	D_P (cm^2/s)	D_{Si} (cm^2/s)	D_{Ag} (cm^2/s)	D_{Re} (cm^2/s)
2	2	6.8E-12	6.1E-08	4.9E-09	3.1E-13	3.3E-17	3.7E-09
5	7	1.5E-11	1.0E-07	9.7E-09	1.3E-12	4.3E-17	1.3E-08
17	24	9.3E-12	7.3E-08	6.8E-09	2.5E-12	1.3E-17	1.9E-08
24	48	8.2E-12	4.9E-08	4.9E-09	3.6E-12	1.6E-17	2.1E-08
24	72	7.1E-12	3.5E-08	3.7E-09	4.3E-12	2.7E-17	2.1E-08
24	96	5.8E-12	3.1E-08	3.4E-09	5.1E-12	3.8E-17	2.2E-08
24	120	4.8E-12	2.2E-08	2.5E-09	5.1E-12	4.9E-17	1.8E-08
336	456	6.4E-13	7.0E-09	6.5E-10	2.4E-12	6.0E-19	9.1E-09
672	1128	6.9E-13	1.2E-09	1.7E-10	1.7E-12	1.2E-19	1.7E-09

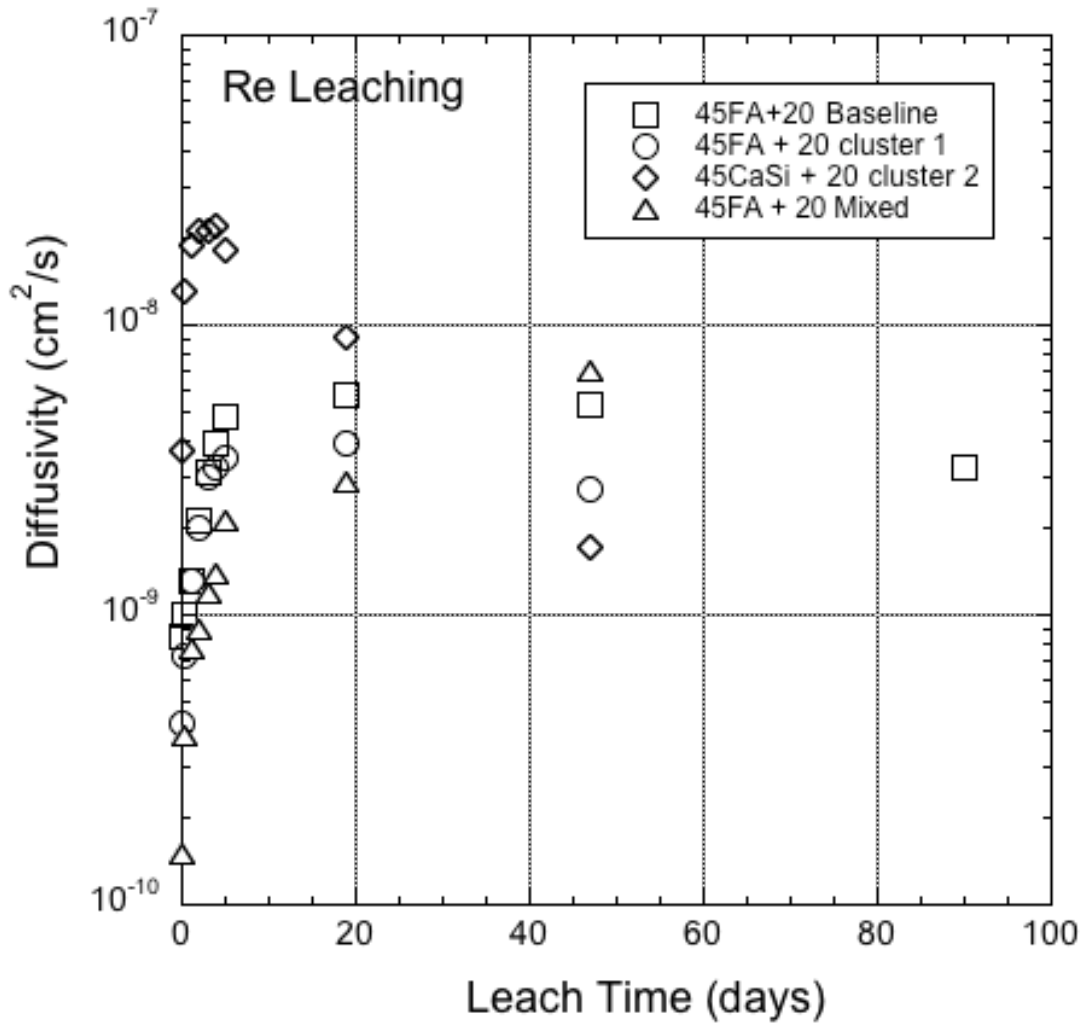
45% FA + 20% Mixed Waste (average value from two samples)

Leach Interval (h)	Cumulative Leach Duration (h)	D_{Mg} (cm^2/s)	D_{Na} (cm^2/s)	D_P (cm^2/s)	D_{Si} (cm^2/s)	D_{Ag} (cm^2/s)	D_{Re} (cm^2/s)
2	2	9.9E-11	2.9E-08	1.0E-08	1.9E-13	4.8E-16	1.5E-10
5	7	1.1E-10	2.8E-08	9.2E-09	3.3E-13	6.4E-16	3.8E-10
17	24	6.9E-11	2.4E-08	7.5E-09	5.3E-13	1.9E-16	7.6E-10
24	48	4.2E-11	1.3E-08	4.9E-09	6.2E-13	2.4E-16	8.9E-10
24	72	3.7E-11	1.1E-08	4.9E-09	9.2E-13	4.0E-16	1.2E-09
24	96	2.4E-11	8.1E-09	3.8E-09	9.2E-13	5.6E-16	1.4E-09
24	120	2.1E-11	6.9E-09	3.4E-09	1.0E-12	7.2E-16	2.1E-09
336	456	4.6E-12	1.7E-09	8.8E-10	4.6E-13	8.0E-18	2.9E-09
672	1128	1.5E-12	2.0E-10	2.6E-10	2.1E-13	6.6E-14	6.9E-09

Diffusivity of Na as a function of cumulative leaching time



Diffusivity of Re as a function of cumulative leaching time



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