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Interim Annual Report
October 1995–September 1996

by L. J. Simpson, D. J. Wronkiewicz,
and J. A. Fortner

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DEVELOPMENT OF TEST ACCEPTANCE STANDARDS FOR
QUALIFICATION OF THE GLASS-BONDED ZEOLITE WASTE FORM

INTERIM ANNUAL REPORT
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Chemical Technology Division

September 1997

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LIST OF ACRONYMS

AEM	Analytical electron microscopy
ANL	Argonne National Laboratory
ASTM	American Society for Testing and Materials
DIW	Deionized water
DOE	U.S. Department of Energy
EDS	Energy dispersive spectroscopy
EJ-13	Leachant made with Yucca Mountain tuff and J-13 water
ER	Elemental release
IC	Ion chromatography
ICP/MS	Inductively coupled plasma/mass spectroscopy
MCC-1	Materials Characterization Center test No. 1
ML	Mass loss
NL	Normalized elemental mass loss
NRR	Normalized release rate
PCT	Product Consistency Test
PCT-B	Modified Product Consistency Test-B
RE	Rare earth
SEM	Scanning electron microscopy
S/V	Ratio of the geometric surface area of the monolith to the volume of the leachant
TCLP	Toxicity Characteristic Leaching Procedure
TEM	Transmission electron microscopy
VHT	Vapor Hydration Test
WAPS	Waste Acceptance Product Specifications
WASRD	Waste Acceptance System Requirements Document
XRD	X-ray diffraction

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ABSTRACT

Glass-bonded zeolite is being developed at Argonne National Laboratory in the Electrometallurgical Treatment Program as a potential ceramic waste form for the disposition of radionuclides associated with the U.S. Department of Energy's (DOE's) spent nuclear fuel conditioning activities. The utility of standard durability tests [e.g. Materials Characterization Center Test #1 (MCC-1), Product Consistency Test (PCT), and Vapor Hydration Test (VHT)] are being evaluated as an initial step in developing test methods that can be used in the process of qualifying this material for acceptance into the Civilian Radioactive Waste Management System. A broad range of potential repository conditions are being evaluated to determine the bounding parameters appropriate for the corrosion testing of the ceramic waste form, and its behavior under accelerated testing conditions. In this report we provide specific characterization information and discuss how the durability test results are affected by changes in pH, leachant composition, and sample surface area to leachant volume ratios. We investigate the release mechanisms and other physical and chemical parameters that are important for establishing acceptance parameters, including the development of appropriate test methodologies required to measure product consistency.

I. INTRODUCTION

A. The Electrometallurgical Treatment Program

The Electrometallurgical Treatment Program at Argonne National Laboratory (ANL) is developing conditioning processes for the potential treatment of a diverse inventory of U.S. Department of Energy (DOE) spent nuclear fuel that may not be amenable for direct geologic disposal.[1] The electrometallurgical treatment uses molten anhydrous LiCl-KCl salt in an electrorefining process that accumulates Sr, Cs, Ba, Ce, La, Nd, and Y fission products, and some transuranics. The residue from the electrorefiner anode and the salt are treated as two separate waste streams. The anode residue is incorporated in a metal waste form, while fission products that accumulate in the salt are loaded into zeolites through a column ion exchange process.[1,2] Additional zeolites are blended with the salt-loaded zeolites to absorb any remaining free salt and pressed together with non-radioactive glass to produce glass-bonded zeolite, termed the "ceramic" waste form. Because the salt-loaded zeolite may have a high transuranic-element content, it may be considered a high-level waste. This report focuses on the development and evaluation of tests for acceptance of the ceramic waste form.

B. Requirements for Acceptance at a Geologic Repository

In accordance with the Nuclear Waste Policy Act of 1982, the Nuclear Regulatory Commission issued document 10 CFR 60 in 1983,[3] that regulated the disposal of high-level radioactive wastes. This document defines such items as licensing requirements, siting criteria, design criteria for the waste package, and performance objectives and requirements for the waste package and the engineered barriers. The two performance criteria of most concern here involve the containment and corrosion of the radioactive waste. Specifically, containment of the high-level waste within the waste package must be "substantially complete" for 300 to 1000 years. Following the containment period, the release rate from the engineered barrier

system cannot exceed 1 part in 100,000 per year for the inventory of that radionuclide calculated to be present at 1,000 years following permanent closure. Furthermore, the chemical, physical, and nuclear properties of the waste and its interaction with the repository environment cannot compromise the function of the waste packages, the performance of the underground facility, or the geologic setting. Therefore, the waste package design must consider solubility, oxidation/reduction reactions, corrosion, hydration, gas generation, thermal effects, mechanical strength, mechanical stress, radiolysis, radiation damage, radionuclide retardation, leaching, fire and explosion hazards, thermal loads, and synergistic interactions. It is likely that the total system performance assessment will consider the release of other elements besides radionuclides for their effect on spent fuel and other high-level waste form behavior. In this report, we will be concerned primarily with issues related to solubility, corrosion, hydration, thermal effects, and leaching.

In 1985, the Environmental Protection Agency issued document 40 CFR 191, establishing the environmental radiation protection standards for management and disposal of spent nuclear fuel, high-level waste, and transuranic radioactive waste.[4] Details in this document specify containment and individual protection requirements and standards for groundwater protection. Furthermore, a 10,000-year limit was established for the cumulative release of radionuclides into the accessible environment, whereby the amount of release is based upon performance assessments of the spent nuclear fuel or the high-level wastes.

The Energy Policy Act of 1992 mandated that, based upon recommendations from the National Academy of Sciences, the Environmental Protection Agency and the Nuclear Regulatory Commission provide a separate set of standards and regulations applicable only to the Yucca Mountain Project.[5] In 1995, the National Academy of Sciences released its report "Technical Basis for Yucca Mountain".[6] It recommended risk-based standards rather than release standards and the elimination of the groundwater protection provisions established by the Environmental Protection Agency. Neither the National Academy of Sciences report nor

the Nuclear Waste Policy Act of 1996[7] significantly changed the design criteria for the waste form or waste form packaging outlined in document 10 CFR 60.

The DOE Civilian Radioactive Waste Management System established the functions to be performed and the technical requirements for accepting spent nuclear fuel and high-level waste in the Waste Acceptance System Requirements Document (WASRD, DOE/RW-0351, revision 2, 1996).[8] This document adopts the waste form criteria of document 10 CFR 60 as it relates to the disposal of commercial spent fuel and the vitrified high-level waste form of borosilicate glass. Similar specifications are being developed for acceptance of other DOE spent nuclear fuels and other high-level waste forms. The Waste Acceptance Product Specifications (WAPS) were developed by the environmental management division of DOE to establish the technical specifications that the waste form producers must meet to ensure acceptance of their vitrified high-level waste into the Civilian Radioactive Waste Management System. These specifications include identification of chemical composition, radionuclide inventory, acceptance criteria, and compliance methods for product consistency, structural stability, and hazardous waste accountability.

The technical basis for acceptance of a waste form, other than borosilicate glass, into the DOE Civilian Radioactive Waste Management System must follow the guidelines presented in the WASRD for spent nuclear fuel and high-level waste and in the WAPS for borosilicate glass.[9] Waste forms must be reviewed and deemed acceptable.

C. Background on Zeolites as an Immobilization Matrix

Zeolites are a well-defined class of crystalline aluminosilicate minerals, with 45 naturally occurring forms and over 100 synthetic forms. Zeolites are characterized by their three-dimensional structures, with frameworks of $[\text{SiO}_4]^{4-}$ and $[\text{AlO}_4]^{5-}$ coordination polyhedra linked by the corners to form an open, negatively charged framework (with

channels and cavities) that is charge balanced by the presence of cations.[2] This open framework provides a relatively accessible passage for ion exchange and water transport. Presently, there is a tremendous amount of scientific and industrial interest in zeolites.[1,2,17-27] The diverse physical parameters of individual zeolites dictate the specific applications. For example, the structural size of the cavities and channels may be used for molecular size selectivity. The negative framework charge may be used for ion exchange selectivity.

The ceramic waste form from the ANL electrometallurgical treatment process is being designed to immobilize Sr, Cs, RE (rare earth) fission products, and transuranics from a molten salt. Chabazite[2,17] and zeolite A[2,10-16,18-27] have both been identified as excellent Sr and Cs absorbers, however, zeolite A has been chosen for the preliminary studies at Argonne National Laboratory because of its lower Si:Al ratio which provides for a greater negative framework charge.[10-14] The distinguishing characteristic of the zeolite A structure is the 1:1 ratio of Si and Al atoms, with twice the number of oxygen atoms.[2] The natural forms of zeolite A contain water, with a unit cell that has twice the number of atoms as sodalite (i.e., $\text{Na}_{12}[(\text{AlO}_2)_{12}(\text{SiO}_2)_{12}] \cdot 27\text{H}_2\text{O}$ versus $[\text{Na}_6[(\text{AlO}_2)_6(\text{SiO}_2)_6] \cdot 8\text{H}_2\text{O}]$). Zeolite A has sodalite cavities (beta cages) and larger, truncated cubo-octahedron, cavities between the sodalite units (alpha cages). The hydrated forms of zeolite A and sodalite are excellent ion exchangers.

Besides hydration and specific ion considerations, thermal stability effects of expansion and crystal degradation must also be considered in determining a viable nuclear waste immobilization matrix. One aspect of this is the possible use of sodalite as an immobilization matrix. Since zeolite A structurally reconfigures to sodalite at the high temperatures sometimes required for the glass-zeolite processing, sodalite has also been considered as a final immobilization matrix. In this process, zeolite A is used to concentrate the fission products and is then converted to sodalite.

The dehydrated form of zeolite A was investigated by Lewis and coworkers for the immobilization of nuclear waste materials.[10,11] Dehydrated zeolite A powder preferentially absorbed Sr, Ba, and Cs from the simulated chloride waste salt, at 500°C, and was radiation stable.[10,11] Zeolite A sorbed Sr and Ba better than zeolite IE95 (a combination of mordenite and chabazite), mordenite, or clinoptilolite. The normalized release rates (NRRs) from 90°C product consistency tests (PCTs) for components present in the samples were reported to be less than or equal to that of SRL-202-G borosilicate glass.[28] However, the difference in particle size between the high surface areas of the 4-μm diameter fine powdered zeolites and the relatively coarse-grained 75 to 150-μm diameter glass makes the validity of these comparisons difficult to evaluate.

The salt and other waste ions form a stable structure that may behave differently from the hydrated zeolites. Therefore, known ion exchange and transport properties of the hydrated zeolites may not be applicable to the salt-loaded zeolites. The relatively limited amount of available information on salt-loaded zeolites[10,11,21,22] may require extensive investigations to completely characterize their ion-exchange and transport properties.

Irradiation from a ^{60}Co source for 1280 h (9.9×10^8 rad) produced color centers in the salt-loaded zeolite A that changed the material color from white to a brown or gray. Comparisons between the release rates of irradiated and non-irradiated samples could not attribute any significant effect of γ radiation on the durability of the salt-loaded zeolite A.[10] However, gamma irradiation induced radiolysis of the leachant as opposed to internal damage of the solid has a greater effect on the release rates of glass and ceramic waste forms.[29]

The work performed by Lewis *et. al.*[10,11] on the salt-loaded zeolite A without the glass binder concluded: (1) Zeolite A ion exchanged Na with and occluded the molten waste salt constituents up to 45 wt% (under pressurized column loading conditions, typically, 20-22 wt% salt occlusion is achieved by blending the zeolite) to form a leach-resistant and

radiation-stable product. (2) Dissolution of the occluded salt was the predominant release mechanism and primarily involved Li and Cl (a large initial release of these ions was observed).

D. Previous Ceramic Waste Form Studies

Lewis *et. al.*[12-14] hot uniaxially pressed salt-loaded zeolites with borosilicate and boroaluminosilicate glass from 600 to 750°C and at 28 MPa to form glass-bonded zeolite samples with different mole fractions. These researchers[12-14] determined:

- (1) The NRRs from 28 and 56 d Materials Characterization Center #1 (MCC-1, "American Society for Testing and Materials" [ASTM] method C1220-92) tests were less than 1 g/(m²•d) for each element contained in samples with 50, 60, and 67 wt% zeolites (the remainder was glass) and decreased with time.
- (2) Ion exchange did not adversely affect leach resistance.
- (3) The glass may be less corrosion resistant than the zeolite.
- (4) Increased pressure above 28 MPa, free salt in the salt-loaded zeolite, incomplete densification, and higher pressing temperatures increased the NRRs.
- (5) The zeolite decreased the Poisson ratio and the Young's modulus of the glass matrix.
- (6) Boroaluminosilicate glass wetted the zeolite powders better and provided a more durable glass-bonded zeolite than soda-lime or high-silica glasses.

- (7) Results from MCC-1 tests indicated that neither the durability of the glass itself, its incipient fusion point, or its softening point can be used to predict the durability of the zeolite bonded with that glass.
- (8) For good durability, the zeolite must be encapsulated in the glass matrix or wetted with the glass.
- (9) The best results were related to glass silica concentrations between 52 and 61 wt% and a molar ratio of alkaline earths to alkalis greater than 1.
- (10) The hot pressing did not cause significant volatilization of Cs or any other component.

E. Acceptance Qualifications of the Ceramic Waste Form

The specific issues related to the ceramic waste form qualification for acceptance into the Civilian Radioactive Waste Management System include regulatory compliance, test methodology, and sample characterization. As outlined in the WAPS, the WASRD defines the borosilicate glass produced by programs directed from the DOE Office of Environmental Restoration and Waste Management as a standard high-level waste form. The WASRD specifications define the physical characteristics and consistency required for a waste form. The physical characterization specifications require: (1) Solids (no free liquids), with particulates consolidated, and combustible radioactive wastes must be reduced to non-combustible forms. (2) The waste materials cannot contribute to free liquids in the waste packages to an amount that could compromise the ability of the waste package to achieve the performance objectives related to containment of the waste form or result in spillage and spread of contamination in the event of waste package perforation during the period through permanent closure. (3) Chemical compositions (elemental and oxide) and crystalline phase

projections for the waste form must be determined. (4) Establishment of structural and compositional phase stability. Demonstration of waste form production control will also be required through a comparison of the waste form with the Environmental Assessment (EA) benchmark glass through the use of a PCT or another as yet to be determined method.[30] In the event that the PCT is deemed an inappropriate test methodology for the ceramic waste form, other test methods for measuring product consistency will also be explored. The waste form must be quantitatively measured for the presence of any hazardous wastes with a Toxicity Characteristic Leaching Procedure (TCLP). This test may be performed in the future, when sufficient samples are available.

To be accepted into the DOE Civilian Radioactive Waste Management System, the ceramic waste form must be reviewed and deemed "acceptable" with the identification and development of the appropriate qualification methods and criteria.[8] As an initial step in this process, the present work evaluates the utility of standard durability tests for providing the information needed to qualify the ceramic waste form.[8] The MCC-1[31], PCT, and Vapor Hydration Test (VHT) are the focus of these initial investigations because of their widespread use with borosilicate glass testing, the ASTM approval of the PCT and MCC-1 test, and their use in qualification of borosilicate glass (e.g., PCT). Additional test methods may be evaluated in the future, depending upon the final composition of the ceramic waste form and the results of these initial tests. A broad range of potential repository conditions is also being incorporated into the testing procedures to determine the bounding parameters appropriate for the corrosion testing of the ceramic waste form and its behavior under accelerated conditions. The effects of pH, leachant composition, sample surface-area-to-leachant volume ratios, and test temperature on the results from durability tests with the ceramic waste form are discussed.

In accordance with the acceptance criteria (WASRD[8] and WAPS), information about the chemical composition, crystalline structures, and structural integrity of the ceramic waste

form and its constituent parts were obtained by performing microanalytical characterization studies. This report includes: (1) scanning electron microscopy and energy dispersive spectroscopy (SEM/EDS) results of the ceramic waste form (with zeolite and sodalite) and the precursor material of zeolite loaded with simulated waste salt (salt-loaded zeolite); (2) transmission electron microscopy and analytical electron microscopy (TEM/AEM) results from the ceramic waste form with sodalite; (3) mass loss measurements of the salt-loaded zeolite and glass at high temperatures; and (4) X-ray diffraction (XRD) analysis.

II. EXPERIMENTAL METHODS AND DEVELOPMENT

A. Sample Production

The Ceramic Waste Form Development Group of the ANL Electrometallurgical Treatment Program provided the samples used in these experiments. The exact processing conditions and sample constituents based on the initial materials used are listed in Appendix A. The zeolite 4A ($\text{Na}_{12}[(\text{AlO}_2)_{12}(\text{SiO}_2)_{12}]$) or 5A ($[\text{Ca}_{4.5}\text{-Na}_3][(\text{AlO}_2)_{12}(\text{SiO}_2)_{12}]$) powders from UOP were dehydrated by heating stepwise up to 525°C in a nitrogen atmosphere. Dehydration was considered complete when zero relative humidity was measured. For the samples discussed here, 21 g of salt were added to 79 g of zeolite and mixed in a blending chamber, heated for 24-48 hours at 550°C. The entire blending process was performed in an Argon atmosphere.[10-13] Equal amounts by weight of glass and salt-loaded zeolite were thoroughly mixed and mechanically pressed into 2.54 cm diameter 304 stainless steel canisters.[12-14] The tops were welded to the canisters in air, and the canisters were evacuated at 500°C for 16 to 48 hours. The H005-2 sample with 311 glass (Appendix B) and salt-loaded zeolite 4A was hot isostatically pressed at 750°C for 2 hours. The H059-1,2 and H062-1,2,3 samples with 57 glass and salt-loaded zeolite 5A were heated to 750°C by means of a "bump and run" process outlined in Appendix A (Table I). The hot isostatic press reached a maximum isostatic load of 28 MPa. The samples were kept in a sealed desiccator to

TABLE I. Ceramic Waste Form Production Specifications

Sample Name	Glass Type	Salt-Loaded Zeolite Type	Pressing Characteristics	Comments
H005-2	311	4A	750°C, 2 hr	Converted to sodalite
H059-1	57	5A	750°C, bump and run	Converted to microsommite
H059-2	57	5A	750°C, bump and run	
H062-1	57	5A	750°C, bump and run	
H062-2	57	5A	750°C, bump and run	Partially converted to microsommite
H062-3	57	5A	750°C, bump and run	

prevent water absorption until they could be used for testing. The ceramic waste form sample monoliths were cut with a diamond saw and polished without the aid of water or oil lubricants to minimize leaching from and contamination of the samples.

B. Testing Methods

The MCC-1, PCT, and VHT, were the main durability tests evaluated here. The MCC-1 test is an ASTM method (C1220-92[31]) that is used to determine the release of ions from sample monoliths in solutions. The ratio of the geometric surface area of the monolith to the volume of the leachant (S/V) in an MCC-1 test is 10 m^{-1} . For the tests performed here, the samples were cut and polished with 240-grit SiC paper without the use of lubricants.

Crushed material with 75 to 150- μm grain sizes is used in the ASTM PCT.[30] The PCT is generally performed with an S/V ratio between 2,000 and 20,000 m^{-1} . The 7 d PCT

has been approved as an acceptance criteria test method for determining the consistency of high-level borosilicate waste glass samples with an S/V of 2000 m⁻¹. [8]

The PCT-B procedure [30] was used for the ion-exchange and preferential ion release study of the salt-loaded zeolites. The 4- μ m-diameter salt-loaded zeolite powders, used in these PCT-Bs have extremely high surface areas that are not representative of the actual material to be disposed, nor are the results directly comparable with standard PCTs conducted with materials that have grain sizes between 75 and 100 μ m. The results from these PCT-Bs provide information about the relative release rates and mechanisms among the different salt-loaded zeolite components.

Besides deionized water (DIW), the use of several other leachants was investigated to determine the relevant effects they may have on the durability tests. The EJ-13 groundwater is a potential leachant that the waste form may encounter at some point during the internment in the geologic repository at Yucca Mountain (Table II). Additionally, the compositions of potential leachants that the ceramic waste form may encounter could depend upon the dissolution of materials in the repository or in different parts of the waste package or the waste form. The most relevant process that may occur results from the dissolution materials of the glass component of the ceramic waste form interacting with the salt-loaded zeolites. This type of mechanism was investigated with a leachant made with dissolved glass (311 glass leachant, Appendix A, Table III) in the PCT-B experiments with the salt-loaded zeolites.

In an unsaturated repository, condensed fluid may react with the glass in the sample, creating thin liquid films of locally concentrated leachates (brines) on the ceramic waste form. The composition of such a fluid will be controlled by the composition of the solid waste form material. High-ionic-strength Mg-Na-K-Cl and Ca-Na-K-Cl brines, prepared according to ASTM standards (Appendix A), were used in the PCT-B experiments with the salt-loaded

TABLE II. Compositions (mg/L) of Leachants Used in the Durability Tests

Element	EJ-13	311 Glass	Ca-Na-K-Cl Brine	Mg-Na-K-Cl Brine
B	0.13	33	1.0	1.3
Na	47	93	34000	35000
Al	0.80	0.54	0	0
Si	46	58	4.8	1.9
Cl	7.3	6.6	100000	100000
K	5.1	3.3	23000	23000
Ca	4.0	8.8	44000	4.5
Mg	0	0	2.8	29000
Sr	0.02	0.34	29	0.05
Cs	0	0	0	0
Ba	0	0	1.3	0.03

TABLE III. The Color of the Different Ceramic Waste Form Samples

Sample Designation	Color of Top	Color of Middle	Color of Bottom
H059-1	Gray	Gray	Gray
H059-2	Light Tan	Light Tan	Light Tan
H062-1	Light Tan	Light Gray	Dark Gray
H062-2	Dark Gray	Dark Gray	Dark Gray
H062-3	Light Tan	Light Gray	Gray

zeolites and in the MCC-1 tests with ceramic waste form monoliths to examine the ion exchange properties of the zeolite material with the brines.

The preparation procedures for the leachants are outlined in Appendix A, and the initial compositions are listed in Table IV.

TABLE IV. Normalized Composition Results from EDS Analyses (Oxide wt%)

Component	Zeolite 4A ^b	311 Glass ^c	H005-2 Target ^d	H005-2 ^e Measured	La-Series Rich Region ^f dry sample	La-Series Rich Region ^f MCC-1 sample	3 d MCC-1 ^g	H005-2 ^h sodalite
Na ₂ O	16.2	6.8	11.5	10.8	7.7	7.0	10.0	1.7
Al ₂ O ₃	26.9	9.9	18.4	19.9	23.9	19.1	20.6	22.0
SiO ₂	33.6	55	44.3	44.8	25.3	20.5	45.4	43.0
Cl	12.0	0	6.0	5.6	1.4	5.8	4.9	6.2
K ₂ O	8.0	0.8	4.4	4.0	1.4	3.7	3.9	3.1
CaO	0	13.6	6.8	7.0	4.9	8.2	6.9	9.3
Y ₂ O ₃	0.0	0	0.0	0.2	1.6	0.4	0.4	0.2
SrO	0.1	0.1	0.1	0.4	1.4	1.6	0.0	2.7
Cs ₂ O	0.4	0	0.2	0.2	0.3	0.6	0.5	3.1
BaO	0.2	0	0.1	0.1	0.4	0.4	0.5	1.0
La ₂ O ₃	0.3	0	0.2	0.1	8.8	3.8	0.0	0.0
Ce ₂ O ₃	0.3	0	0.2	0.1	4.6	8.7	0.0	0.0
Nd ₂ O ₃	0.4	0	0.2	0.2	11.4	12.7	0.2	0.2
LiO ₂ ^a	1.2	0	0.6	0.6	0.6	0.6	0.6	0.6
B ₂ O ₃ ^a	0	13.9	6.9	6.9	6.9	6.9	6.9	6.9
Total	100	100	100	100	100	100	100	100

- Lithium and boron could not be accurately measured with EDS, but were accounted for in the compositional analysis from initial composition calculations
- Calculated composition of the salt-loaded zeolite from the initial masses.
- Calculated composition of the 311 glass from the initial elemental masses.
- Calculated composition from the initial elemental masses.
- Measured composition from the initial elemental masses.
- Measured composition from areas with enhanced La series elements.
- Measured composition of the MCC-1 sample after a 3 d test.
- Measured composition from areas with enhanced Sr and Cs.

Coupons from the H059-1 and H059-2 samples (Table I) were "dry" cut with a diamond saw. The flat sample surfaces were polished to 240 or 600 grit (without the use of any lubricant), sonicated for 5 minutes in isopropyl alcohol to remove any polishing residue, and dried in a 90°C oven for 10 minutes. Monolithic VHT samples were suspended with a Teflon thread in stainless steel vessels, with DIW, and placed in ovens at 120, 150, and 200°C. Enough DIW was added to the test vessel to provide 100% relative humidity inside the test chamber, but the amount was limited to prevent dripping of accumulated fluid from the samples. Thus all the dissolution products stay in contact with the sample surface in a thin film of water, where they precipitate to form alteration phases. The VHT is an accelerated test that may provide information on corrosion rates, alteration phase paragenesis, material toughness, structural integrity, and potential sorption of fission products on alteration phases. Thus, the VHT can provide information about ceramic waste form alteration phases that may be useful for the performance assessment modeling efforts required for geologic disposal.[32]

The use of additional durability tests, like the unsaturated drip test,[33] the accelerated dissolution test,[34] the Soxhlet test,[35] continuous flow-through tests,[36] and others[37] with the ceramic waste form may be investigated depending upon the final structure and composition chosen.

The information obtained from these investigations should be sufficient to evaluate the utility of these durability tests for determining the criteria that the ceramic waste form will need for acceptance into the Civilian Radioactive Waste Management System. However, for disposition in a geologic repository, evaluations of additional durability tests will be required to fulfill the repository relevant performance assessment criteria. This may require additional assessments of durability tests that use unsaturated conditions at elevated temperatures.

The presence of any hazardous material contained within the ceramic waste form must be quantified and the producer may be required to perform a Toxicity Characteristic Leaching Procedure (TCLP) or other relevant tests in accordance with the 55 Federal Register 26986, 6/29/90, of the Resource Conservation and Recovery Act.[38] This may be important for fission product and decay daughters, like Ba, Se, and As, that are present in the ceramic waste form.

C. Solids Analysis

Optical microscopy, SEM, AEM, and XRD analyses are being performed as initial survey techniques of the materials and provide background information about the morphology, composition, structure, bonding, and alteration phase formation of the ceramic waste form and the precursor material of salt-loaded zeolite.

A Topcon ABT-60 SEM operates with a focused electron beam, having electron energies between 5 and 30 keV (resolution approximately 0.1 μm), that interact with the sample in a number of elastic and inelastic processes which provide elemental and image information. X-ray microanalysis and digital imaging were obtained with a Noran Instruments Voyager system (Version 3.3). Energy dispersive X-ray spectroscopy has a resolution of approximately 3 μm (at 10 KeV) for the relatively low density glass and zeolite materials examined in this study and is sensitive to elements with atomic numbers higher than beryllium. This technique provides good characterization of samples without significant sample preparation requirements. In general, the accuracy of the EDS numbers range between ± 5 to 10%, with the lighter elements having the lower accuracy.[39] The instrument does have nominal detection limits of 0.1 to 0.5 wt%. However, the structural arrangement and distribution of a given element may raise these detection limits.[39]

Analytical electron microscopy is a combination of TEM, energy dispersive X-ray spectroscopy, selected area electron diffraction, and convergent beam electron diffraction. The samples were analyzed with a JEOL 2000 FXII TEM with an incident electron energy of 200 KeV and was equipped with 2 Noran energy dispersive spectroscopy detectors and a Gatan parallel detection electron energy loss spectrometer. Point-to-point resolution for images obtained with TEM approaches 0.3 nm. The resolution from electron diffraction, EDS, and electron energy loss spectroscopy from the AEM instrument is approximately 20 nm. Electron energy loss spectroscopy is sensitive to elements with atomic numbers higher than hydrogen and can detect RE and transuranic elements at levels as low as 50 mg/L. These techniques pass high-energy electrons through solid materials that interact strongly via a number of elastic and inelastic processes which provide structural and compositional information on the nanometer scale. This analytical technique requires the sample to be extremely thin (<70 nm) necessitating the use of advanced ultramicrotomy procedures and embedding.

Migration of Na and Cl due to the electron beam interaction was evaluated and factored into the experimental procedures. The ultramicrotomy processes, the inherent heterogeneity of the glass-crystal samples, and the thin cross section of the samples resulted in individual AEM sample mounts displaying significantly different material and structural compositions. Several AEM sample mounts were prepared, but the limited number analyzed may not be representative of the ceramic waste form as a whole.

Structural analysis was obtained with a Rigaku D/Max2400V X-ray diffractometer and selected area electron diffraction. The XRD should be accurate to within 0.1° for 2θ measurements.

D. Solution Analysis

Leachate analyses were performed with a Fisson Instruments Plasma Quad Inductively coupled plasma mass spectrometer (ICP/MS) for the cation analysis, Ion Chromatography (IC) for anion analysis, a Brinkmann 605 pH meter, and a Dohrmann Carbon Analyzer. The leachate cation, anion, and carbon analysis are accurate to within $\pm 15\%$. The pH measurements are accurate to within ± 0.2 . The analyses were performed at room temperature, and the cation solutions were acidified to prevent precipitate formation.

E. Standard Data Characterization

The leachate analysis provides information about the concentration of a specific ion in the solution (C_i) and is reported as mg/L or $\mu\text{g/L}$. The leachate concentrations can be converted to normalized elemental mass loss (NL_i) or the elemental release percent ($ER_i\%$) that are defined as (Appendix B):

$$NL_i = \frac{(C_i - C_{is}) V_s}{(f_i A)} = NRR \cdot t \quad (1)$$

$$ER_i\% = \frac{100 \cdot m_i}{M_i} = \frac{100 (C_i - C_{is}) V_s}{(f_i M)} \quad (2)$$

- where, C_i = concentration of i th element in the leachate (g/m^3)
 C_{is} = starting concentration of i th element in the leachant (g/m^3)
 V_s = volume of the leachate (m^3)
 f_i = fraction of i th element in the sample
 t = duration of tests (d)
 A = surface area of the sample (m^2)

m_i = mass of i th element released to the leachate (g)

M_i = total mass of the i th element in the sample (g)

M = total mass of the sample (g).

The NL_i provides a measure of the amount of released solids, accounting for the surface area of the sample interacting with the leachant. The surface areas are determined geometrically for monoliths and are estimated for crushed samples. However, if ion exchange or preferential ion release is occurring, then the external geometric surface area is less important and the correlation between the amount released compared to the total amount of an element in the sample (as given with $ER_i\%$) may be more useful. The latter is useful for the evaluation of ion release from the salt-loaded zeolites that have large surface areas which substantially decrease the NL_i values. However, geometric effects are important and must be considered for any meaningful evaluation. The total mass loss (ML) may provide a quick and easy method for evaluating the durability of a sample, but the geometric effects compounded by ion exchange or absorption processes limits the utility of this method.

Some corrosion rate information may also be obtained through the physical inspection of tested samples. If an altered layer can be observed and measured, as in the VHTs, then a corrosion rate may be calculated. Similarly, the identification of altered corrosion products or precipitate formations may be useful in identifying transformation pathways. In either case, these type of measurements are more qualitative than the analysis of the leachate, but provide valuable complimentary information about solid phase formations and potential ion-exchange processes. This is especially true for solid-phase identification in the interpretation of leachate data. Here, examination of the solids is required to ascertain whether reprecipitation has occurred, so that the material performance evaluation does not underestimate the amount of reacted material.

III. RESULTS

A. Sample Characterization

1. Blended Zeolite 4A Loaded with Simulated Waste Salt

a. SEM/EDS Characterization

A zeolite 4A sample blended with the simulated waste salt was provided by the ANL Ceramic Waste Form Development Group. The initial configuration of the constituents and the processing conditions of this sample are detailed in Appendix A. Fig. 1a is an SEM image of the pure salt-loaded zeolite 4A. Analyses from this type of SEM image indicated that the average particle size was approximately 4 μm . Furthermore, little difference was observed between the zeolite particles before and after a 2-minute sonication in DIW, suggesting that very little residual salt exists outside of the zeolite. This is consistent with the XRD results presented in Appendix E.

A sample of pure zeolite 5A that contained Ca and Na anions in the structure and a sample of zeolite 5A blended with the simulated waste salt were also provided by the ANL Ceramic Waste Form Development Group. Scanning electron microscopy results of the zeolite 5A revealed little difference in morphology, composition (except for the Ca and Na differences), structure, and size from the zeolite 4A.

Significant differences in morphology could not be observed between the salt-loaded zeolite 4A (Fig. 1a), sodalite (Fig. 1b), and nepheline ($\text{K}[\text{Na},\text{K}]_3\text{Al}_4\text{Si}_4\text{O}_{16}$, Fig. 1c) with SEM. The sodalite and nepheline were identified with XRD (Appendix E) and

(a)

15 μm

(b)

15 μm

(c)

15 μm

Fig. 1. Secondary Electron Images from a Scanning Electron Microscope. (a) Pure salt-loaded zeolite 4A powder. (b) Salt loaded zeolite 4A powder converted to sodalite after 4 h at 700°C in an open Pt crucible with 311 glass. (c) Salt loaded zeolite 4A powder converted to nepheline after 24 h at 700°C in an open Pt crucible.

were produced by holding samples of salt-loaded zeolite 4A at 700°C for 4 h in contact with the 311 glass and for 24 h by itself, respectively (Appendix D). The SEM/EDS analyses on the sodalite and nepheline were performed in an effort to identify any morphological, compositional, or structural differences between the salt-loaded zeolite 4A and its high-temperature transformation products.

b. Compositional Effects on Zeolite 4A to Sodalite Transformation

Part of the characterization studies with the salt-loaded zeolite 4A involved the effect of the glass matrix on the stability of the zeolite structure. Ion exchange, ion release, and water transport between the glass and the salt-loaded zeolite may influence phase compositional changes and the kinetics of transformation. At 700°C, salt-loaded zeolite transforms to nepheline and sodalite after 24 h. However, water transport and Ca and Na ion exchange were identified as potential processes that accelerated the salt-loaded zeolite 4A transformation to sodalite when mixed with 311 glass (transformation occurred within 4 h).

One potential mechanism involves the absorption of water from the 311 glass into the salt-loaded zeolite 4A at high temperatures. This absorbed water presumably decreases the zeolite stability, thus accelerating its transformation to sodalite. To fully investigate this hypothesis, a complete accounting of the water content in the glass and zeolite was required. The amount of water in the 311 glass was determined by recording the mass change of a powdered sample of 311 glass in a platinum crucible at various temperatures and times, (Appendix D). The results of these measurements indicated that only 0.5 and 0.6 wt% of the sample volatilized at 700 and 850°C, respectively. Furthermore, all the mass loss of the 311 glass at 700°C occurred within the first hour and is probably associated with water adsorbed on the surface of the glass particles (Appendix D).

Similar measurements were made with the salt-loaded zeolite 4A in a Pt crucible at 700°C. The salt-loaded zeolite 4A had a 2 wt% loss within the first hour and continued to lose approximately 2.2 wt% per day at 700°C for several days after the tests were initiated (Appendix D).

If we assume that the initial mass loss on ignition of these tests was due to volatile water, then in the ceramic waste form with a 50/50 mixture of zeolite and glass, the 311 glass could contribute 0.5 wt% to the salt-loaded zeolite's 2 wt% water at 700°C. Precluding an unforeseen catalytic effect of a 0.5 wt% absorbency of water by the salt-loaded zeolite, the fact that at 700°C the salt-loaded zeolite 4A releases 2 wt% water, and the fact that in open crucible tests water should vaporize out of the sample before it is absorbed into the zeolite, it is unlikely that water contributed to the zeolite 4A from the 311 glass could be responsible for the observed high-temperature acceleration of the zeolite transformation. This accelerated transformation of zeolite in contact with glass is observed in both the open crucible and the closed hot isostatically pressed samples.

A. Ellison and D. Strachan of the ANL Nuclear Waste Management Department suggested that the unbalanced Na (with respect to Al and Si concentrations) in the 311 glass may account for the accelerated salt-loaded zeolite 4A transformation through solid-state ion exchange. To test this theory, a set of four samples were prepared and heated at 700°C (Appendix D): (1) salt-loaded zeolite 4A, (2) equal amounts by weight of salt-loaded zeolite 4A and K-Li glass (Appendix A), (3) equal amounts by weight of salt-loaded zeolite 4A and NaCl, and (4) equal amounts by weight of salt-loaded zeolite 4A and 311 glass. Small portions of each sample were removed at regular intervals and mass loss measurements were taken. The timed sequence of samples were submitted for XRD (Appendix E) to evaluate the degree of transformation.

Only the salt-loaded zeolite 4A plus 311 glass sample converted to sodalite within 4 hours at 700°C (Fig. 2; Appendices D and E). In addition the same sample had partially converted from sodalite to nepheline after 24 h. The salt-loaded zeolite 4A plus K-Li glass sample converted to sodalite (with some nepheline present) and the salt-loaded zeolite 4A and salt-loaded zeolite 4A plus NaCl samples both converted to nepheline (sodalite was a minor phase) after 24 h at 700°C. These results indicate that the Na in the 311 glass was sufficient to create solid-state ion exchange with the salt-loaded zeolite 4A and accelerate the transformation of zeolite to sodalite. The complete removal of the Na in the K-Li glass or the use of a Na-containing material with a lower Na “activity,” like NaCl, increased the amount of time before the salt-loaded zeolite 4A transformed.

Fig. 2. Phase Composition Changes of the Salt-Loaded Zeolite 4A at 700°C as a Function of Time. The transformation of the salt-loaded zeolite 4A to sodalite was accelerated in the presence of the 311 glass. After 24 h, all of the samples had completely converted to a combination of sodalite and nepheline. The semi-quantitative identification of the phases was determined by XRD.

The glass in the salt-loaded zeolite 4A plus K-Li glass sample induced the same sodalite to nepheline transformation as the salt-loaded zeolite 4A plus 311 glass sample after 24 h, while the NaCl resulted in a transformation primarily to nepheline, similar to the pure salt-loaded zeolite 4A. This indicates that the glass either reduces the rate of the sodalite to nepheline transformation or changes the transformation path from nepheline to that of sodalite when certain (as yet unknown) components in the glass are present. The K-Li glass contained approximately the same overall composition as the 311 glass with the Na exchanged on a mole-per-mole basis with the eutectic K-Li mixture. Therefore, Ca, K, Li, B, and H₂O, which are present in both glasses, do not appear to be involved with the accelerated transformation of salt-loaded zeolite 4A to sodalite at high temperatures.

2. The Ceramic Waste Form with Zeolite 5A

Lewis *et. al.*[12-14] initiated the investigations of glass-bonded zeolite as a waste form with hot isostatically pressed salt-loaded zeolite in borosilicate and boroaluminosilicate glass. With this experience, five ceramic waste form samples (designated H059-1, H059-2, H062-1, H062-2, and H062-3) were provided by the Ceramic Waste Form Development Group to be used in evaluating durability and characterization tests. All these specimens were prepared in a similar way, and under similar processing conditions (Appendix A, Table I), with a glass designated as "57" (Appendix A) and zeolite 5A blended with simulated waste salt (salt-loaded zeolite 5A, all compositions given in Appendix B). The first two samples (H059-1 and 2) were loaded in air and hot isostatically pressed at the same time. The last three samples (H062-1,2,3) were loaded in a dry box and hot isostatically pressed at the same time.

From a processing and preparation standpoint, the five samples should be equivalent and therefore provide similar results in any of the durability tests. Initial inspection of the five samples, however, indicated a significant difference in the color of the

ceramic waste forms from sample to sample and even within different portions of the same sample. These color changes have been associated with slight changes in the sample densities ($\pm 0.1 \text{ g/cm}^3$) by the Ceramic Waste Form Development Group. The densities were measured with a pycnometer that uses Archimedes principle with octanol to measure open porosity. This type of density measurement can provide accuracies better than $\pm 0.05 \text{ g/cm}^3$. Generally, the samples varied from a light tan to a dark gray color, with the lighter samples being less dense than the darker samples[40]. The tops of the samples were lighter in color than the bottoms, suggesting an association between the color change and the mechanical packing process of the glass/zeolite into the stainless steel canisters (Table III, Appendix A). The identification of any structural or chemical differences associated with the different colors of the samples was investigated in order to determine potential effects on the durability tests.

A thorough examination of the as-received material was performed to establish a baseline for comparison with the samples that were used in the tests. The SEM images along with EDS analyses (Figs. 3 through 5) indicated a uniform distribution of zeolite and glass for dimensions greater than $100 \text{ }\mu\text{m}$. However, for dimensions smaller than $100 \text{ }\mu\text{m}$, the glass (with particles as large as $45 \text{ }\mu\text{m}$) and salt-loaded zeolite are not uniformly distributed. The salt-loaded zeolites are concentrated in regions between the large glass particles. This may be corrected by reducing the average glass particle size or by increasing the hot isostatic pressing times while the glass is soft, so that the zeolite can distribute more evenly. The increased concentration of zeolites in the ceramic waste form results in individual zeolite particles physically contacting other zeolites. This could affect the corrosion behavior of the waste form by allowing channel access to the zeolites. Detailed analysis of the ceramic waste form samples (Figs. 3 through 5) indicate that all the samples contain:

1. Regions with high concentrations of RE ions (center of Figs. 4d and 5b).

Fig. 3. Scanning Electron Microscopy Images of Ceramic Waste Form Sample. (a) H059-1, secondary electron image; (b) H059-1, backscatter electron image; (c) H059-2, secondary electron image; and (d) H059-2, backscatter electron image. The images show the non-uniform distribution of the zeolites (gray features) in the glass matrix (dark regions) for dimensions smaller than 100 μm , the formation of crystalline NaCl (small white features), and the presence of pores (small round dark features). The bright feature in the center of image (b) is a region with high concentrations of rare earths. Fractures in the material are observed in the secondary electron images, but not in the backscatter images of the same regions (compare a and b, and c and d).

Fig. 4. Secondary Electron Images from Scanning Electron Microscopy of Ceramic Waste Form Samples. (a) H062-1, top third of sample; (b) H062-1, middle third of sample; (c) H062-1, middle third of sample (zirconium rich phase present in the middle of the image); and (d) H062-1, bottom third of sample. The light gray regions of the images are salt-loaded zeolite, the darker areas are the glass, and the small bright (white) square features are NaCl crystals. The small dark irregular features in images (a) and (b) are due to pores in the ceramic. The large bright white sub-rounded features in the middle of image (c) and the smaller feature in image (d) are Zr rich phases, while the irregular feature in the middle of image (d) contains high concentrations of RE and chloride ions.

Fig. 5. Secondary Electron Images from Scanning Electron Microscopy of Ceramic Waste Form Samples. (a) H062-3, top third of sample; (b) H062-3, middle third of sample; (c) H062-3, bottom third of sample; and (d) H062-2. The light gray regions of the images are salt-loaded zeolite, the darker areas are the glass, and the small bright (white) square features are NaCl crystals. The small dark features are due to pores in the ceramic. The larger bright white feature in the top right of image (a) is a Zr rich phase, while the irregular feature in the middle of image (b) contains high concentrations of RE and chloride ions.

2. Zirconium rich phases (circular features, Figs. 4c, 4d, and 5a).
3. More small pores (1-5 μm diameter) at the top of the sample (Figs. 4a and 5a). This has been associated with the canister packing process.

Small NaCl crystals were observed on all the samples that contained zeolite as the major crystalline phase. Analysis of the XRD spectra (Appendix E) indicate that samples H059-2, H062-1, and H062-3 contained primarily salt-loaded zeolite 5A and that crystalline NaCl was more prevalent in coupons taken from the top of the samples than the bottom (Appendix E). Analysis of the XRD spectra in Appendix E indicate the partial and complete transformation of the salt-loaded zeolite 5A to microsommitite $[(\text{Ca}, \text{Na}, \text{K})_8(\text{Al}, \text{Si})_{12}\text{O}_{24}\text{Cl}_{2.5}]$ in the H062-2 and H059-1 samples, respectively (Table I). Since these two samples were processed under exactly the same conditions as the other three samples, the structural transformation of these samples was not anticipated. The structural transformation of part or all of a specific sample indicates the relatively high sensitivity that this ceramic waste form displays towards processing conditions. This may be an important acceptance issue, since the structure and composition of the crystal phases must be identified and the batch to batch consistency of the product must be established.

Analysis of SEM images of the samples that partially or fully converted to microsommitite indicated less crystalline NaCl at the surface than the salt-loaded zeolite 5A samples. This is easily observed in Fig. 3, where large NaCl crystals are observed on the H059-2 sample (Figs. 3c and 3d) but not on the H059-1 sample (Figs. 3a and 3b). This is the case even though both samples developed micro-fractures (Figs. 3a and 3c) with the NaCl crystals preferentially observed along the fractures in the zeolite regions of the H059-2 sample (Fig. 1c). The fracturing appears to cross natural material boundaries suggesting that the fracture formation is associated with intrinsic stress in the samples. This would suggest

that annealing may aid in the minimization of this fracturing and that the zeolite-glass bond is stronger here than in the ceramic waste form with sodalite and 311 glass (Sect. III.A.3). Furthermore, the presence of the large NaCl crystals along the fractures and at the fracture triple points in the zeolite samples along with the lack of NaCl formation in the H059-1 sample would suggest that the NaCl is being released from the ceramic waste form materials and crystallizing at the high mass transport regions along the fractures. The raised surface features of the salt, as exhibited by their bright secondary electron image response (Figs. 3c and 3d), suggest that salt migration and crystallization occurred after the samples were cut and dry-polished for SEM analysis. This is probably the result of the ethanol used during the ultrasonic cleaning of the samples, providing enhanced transport of the salt to the surface during the drying process at 90°C.

3. The Ceramic Waste Form with Sodalite

A ceramic waste form with salt-loaded sodalite (sample H005-2) was provided by the Ceramic Waste Form Development Group. The initial configuration and processing conditions of this sample (Appendix A) resulted in the initial salt-loaded zeolite being converted to sodalite in the final product (Table I). The zeolite conversion to sodalite will probably affect the ion-exchange selectivity and release behavior of the waste form. However, this sample provided an initial evaluation of durability and characterization tests while the ceramic waste form samples with zeolite were prepared. The ceramic waste form with sodalite was used to evaluate the effects of sample preparation and provide preliminary information about the utility of constituent and morphology analysis with SEM and AEM. As part of this initial characterization, VHT and MCC-1 tests were performed with this ceramic waste form (Sect. III.B.2 and III.B.3).

a. Scanning Electron Microscopy Characterization

The SEM/EDS results of the H005-2 sample indicated that the sodalite was uniformly distributed throughout the sample. However, significant sodalite pullout on the polished surfaces (pores in Fig. 6a), regions of enhanced RE element concentrations (Fig. 6b), and cesium and strontium accumulation were observed before and after the MCC-1 test (Table IV). The sodalite pullout suggests that a poorly bonded contact between the sodalite and glass exists which may allow preferential fluid access to these regions during corrosion testing. A heterogeneous accumulation of simulated fission products may indicate a potential processing problem with the distribution of these elements. The observation of cubic sodalite particles in the region of concentrated RE elements (Fig. 6b, Table V), suggests that the RE ions preferentially concentrate in a few crystalline particles that tend to stay together.

Fig. 6. Secondary Electron Images from a Scanning Electron Microscope. (a) A polished H005-2 sample. (b) Enlargement of a highly concentrated RE region in the polished H005-2 sample.

b. Analytical Electron Microscopy (AEM) Characterization

Analytical electron microscopy was performed on the ceramic waste form sample H005-2 to determine the mineralogy of the crystalline phases, the composition of both the glass and the crystals, and the bonding characteristics between the crystals and the glass.

The processing conditions of the H005-2 sample resulted in zeolite conversion to sodalite in the final ceramic waste form, as indicated by the AEM electron diffraction patterns (Table V; Figs. 7 and 11). Extra diffraction spots in some of the samples, and the entire diffraction pattern from block #1564 (Table V), suggest possible feldspar inclusions. An examination of micrographs like Fig. 9 and analysis of the diffraction patterns suggest that feldspar comprises 10 to 20% of the crystal phase in this sample. Due to the small sample size and potential non-representative nature of the specimens, accurate quantification of the overall feldspar quantity in the ceramic waste form samples was impossible to determine. The phases identified as feldspar contained far less Cs and RE ions than the sodalite (Fig. 13). The crystalline material was found to have varying levels of Cs, Ba, Ce, La, Nd, and Cl (Figs. 8, 10, 12 through 14). Extra diffraction spots at a d-spacing of 1.06 nm may indicate the formation of a supercell in the crystal structure (Table V).

TABLE V. Electron Diffraction Patterns from the H005-2 Crystalline Material. The diffraction patterns were consistent with sodalite with the possible presence of feldspar. Joint Committee on Powder Diffraction Standards (JCPDS) reference values given for intensity peak ratios (I/I_0) greater than 0.30.

Block 1559 d spacings (nm)	Block 1560 d spacings (nm)	Block 1564 d spacings (nm)	Block 1570 d spacings (nm)	Precision in d spacings (nm)	JCPDS 9-456 Feldspar, Albite, calcian, high (Na,Ca) (Si,Al) ₄ O ₈ d spacings (nm)	JCPDS 10-360 Feldspar, Anorthite, sodian, high (Ca,Na) (Si,Al) ₄ O ₈ d spacings (nm)	JCPDS 23-651 Feldspar, Banalsite, Na ₂ Ba (Al ₂ Si ₂ O ₈) ₂ d spacings (nm)	JCPDS 10-360 Sodalite Na ₄ Al ₃ Si ₃ - O ₁₂ Cl d spacings (nm)
		1.060	1.090	0.030			0.850	
0.646	0.624	0.630	0.624	0.010	0.643	0.649	0.650	0.630
0.572			0.546					
0.502								
0.459	0.442	0.470	0.476 0.442	0.005	0.469	0.468	0.520 0.420	0.445
0.404					0.402	0.403 0.390		
0.372				0.003	0.374	0.375	0.377	
	0.361	0.354	0.364	0.003	0.363 0.346	0.363 0.346	0.354	0.363
0.338			0.330			0.336		
	0.312	0.323		0.003	0.326 0.320 0.317	0.326 0.323 0.320 0.317	0.321	
0.288	0.279		0.275	0.003			0.310 0.300 0.290	0.281
		0.267		0.002	0.265	0.265	0.265	0.257
0.262		0.264	0.264	0.002			0.260	
0.246	0.254	0.249	0.244	0.001	0.252	0.251	0.256 0.250	
0.238	0.237	0.238		0.001	d<0.252 not reported	d<0.251 not reported	0.239	0.238
0.215	0.209	0.210	0.209	0.001	"	"	0.209	0.210
0.187	0.189				"	"		0.109
	0.186		0.180		"	"		0.181
		0.169		0.001	"	"	0.169	
		0.158		0.001	"	"	0.158	0.157

Fig. 7. Electron Diffraction Image from Block 1559.

Fig. 8. Energy Dispersive Spectrum of the Sodalite in Block 1559. Note the very weak signal due to neodymium. The inset is the same spectrum on a linear, rather than logarithmic, scale. The Cu in the energy dispersive spectra is from the analytical electron microscopy mount.

Fig. 9. Micrograph of the Crystalline Material in Block 1559. Note the small (< 100 nm) included phase, indicated by the arrow. Diffraction data suggest it may be a feldspar.

Fig. 10. Electron Energy Loss Spectroscopy of the Sodalite Crystal in Block 1559, Revealing the Presence of Cs, Ba, La, Ce, and Nd.

Fig. 11. Electron Diffraction Image from Block 1560.

Fig. 12. Energy Dispersive Spectrum from the Crystalline Material in Block 1560. Excess Si was detected over that expected for the sodalite composition, possibly indicating spectral interference resulting from the presence of silicate glass. The Cu in the energy dispersive spectra is from the analytical electron microscopy mount.

Fig. 13. Second Difference Electron Energy Loss Spectroscopy Spectra from Block 1564. The spectrum at left shows a barium-containing phase, possibly the feldspar Banalsite (see Table V), with possible cerium incorporation. The spectrum at right, from another spot on the sample, shows the presence of fluorine (possibly substituted for chlorine in a sodalite), with very low levels of neodymium and no detectable barium.

Fig. 14. Second Difference Electron Energy Loss Spectroscopy Spectra from Block 1566 that Contained Only Glass. The Cu in the energy dispersive spectra is from the analytical electron microscopy mount.

While the precision in measuring the electron diffraction patterns is relatively high (Table V, column 5), systematic errors associated with beam stability, lattice heating, and alignment may allow only a $\pm 5\%$ accuracy (Table V, column 1).

The EDS spectrum from the crystalline material in Figs. 8 and 12 indicate the presence of Ca, Cu, and excess Si. More Si was detected than expected for the sodalite composition, possibly indicating spectral interference resulting from the presence of silicate glass. The Cu in the EDS spectra is from the AEM mount. The presence of Ca in the EDS spectra, initially only in the 311 glass, suggest that electron interaction with glass is creating a spectral interference or that solid-state ion exchange between the glass and sodalite occurred.

The crystals were not sensitive to the electron beam, which is consistent with very low incorporation of water in the samples. Crystalline phases containing sodium were robust in the electron beam, even during electron energy loss spectroscopy acquisition. This behavior is somewhat unexpected, since sodium is usually mobile in many materials under the flux of the high-energy electrons used in electron energy loss spectroscopy. The sodium in these materials appears to be sufficiently anchored such that sodium migration was not a problem.

Glass-crystalline interfaces were not preserved in any of the AEM samples prepared, again suggesting that a weak bond exists between the glass and sodalite (Fig. 15). This is consistent with the sodalite pullout discussed above (Sect. III.3.a). Preparation of ceramic waste form samples with zeolites for AEM analysis by ultramicrotomy ion milling procedures may more fully address this problem.

Fig. 15. Micrograph Shows Both Glass and Crystalline Material Coexisting in Block 1570. The interface between the crystalline and glass phases was not preserved. This would have provided information about the bonding.

B. Corrosion Testing

Vapor hydration, product consistency, and MCC-1 tests with the ceramic waste form material and its constituent parts are ongoing. Some of these tests will require a year to complete. However, initial tests have been terminated and the results are reported below.

Special preparation procedures and tests were performed with the H005-2 sample to establish the initial leach rates and leach mechanisms of the ceramic waste form. Analysis with SEM/EDS, AEM, and ion chromatography techniques (ICP/MS, carbon, and IC) were performed on the dry cut, wet cut, and MCC-1 H005-2 samples (Table VI, Appendix B) to establish the effects of sample preparation and the sensitivity of these different characterization techniques to the different test samples. All but one of the samples were dry cut with a diamond saw to ensure that no leaching occurred during the sample preparation.

TABLE VI. Element Concentrations in the DIW Lubricant
Used to Wet Core the H005-2 Sample

Element	Solution Concentration (mg/L)
Li	4.5
B	10
Na	175
Si	10
K	15
Ca	<4
Cl	269

To investigate the effect of DIW on sample preparation, the DIW cutting fluid from a sample cut with a diamond coring saw and the solution used to wash a dry cut MCC-1 coupon (washed and ultrasonicated in DIW for 2 minutes) were analyzed. An analysis of the wash fluid indicated that all the glass and sodalite components in solution were at or below the detection limit of the standard ICP/MS analysis. However, the cutting fluid contained a relatively high concentrations of Li, B, Na, Si, K, and Cl (Table VI). Both solutions were passed through a 0.45- μ m filter before analysis. The solution concentrations of the alkali metals, chloride, and silicon are comparable to those observed in PCTs with the salt-loaded zeolites without the glass binder (Appendix B). The relatively harsh environment associated with cutting the sample and the possibility of fine particles going through the filter suggest that these data should only be used to identify a potential problem that should be investigated further.

Scanning electron microscopy and EDS analysis of the samples could not distinguish any significant difference between the compositions of the dry-cut, wet-cored and DIW-ultrasonically cleaned MCC-1 samples. This is probably related to the limited sensitivity of SEM/EDS to small changes in composition at the surface of a sample (Table IV). Scanning

electron microscopy photographs indicated very little difference between the surfaces that were dry or wet polished, or surfaces that had been exposed to DIW for short amounts of time.

1. Product Consistency Test (PCT)

Approximately 30% of the ceramic waste form samples crushed for PCTs had the correct size distribution (-100 to +200 mesh; 75 to 150 μm). Of the five initial ceramic waste form samples received, only the three with pure zeolite 5A (H059-2, H062-1, H062-3) will be used in the PCTs. With most of the H059-2 sample used for VHTs and MCC-1 tests, the limited material that remained from all the samples was insufficient to perform all the PCTs. Additional samples have been provided by the Ceramic Waste Form Development Group and are currently being prepared for analysis.

The precursor material of salt-loaded zeolite was also provided by the Ceramic Waste Form Development Group and PCT-Bs with the 4- μm diameter material were initiated while additional ceramic waste form material was being processed.

- a. Product Consistency Test-B (PCT-B) with Salt-Loaded Zeolite

The effects of several leachant compositions on the 4- μm diameter salt-loaded zeolite 4A powders have been examined with the PCT-B procedure. The leachants include DIW, EJ-13, 311 leachant, Ca-Na-K-Cl brine, Mg-Na-K-Cl brine, and phosphate buffers at different pH values. The incorporation of these fluids, that represent a broad range of leachants to which the waste form may be exposed, into durability test procedures should provide information about the bounding parameters and mechanistic release scenarios appropriate for the corrosion testing of the ceramic waste form and about the behavior of the waste form under accelerated testing conditions.

The effect of brines on the ceramic waste form may be important when considering disposition in an unsaturated repository, where fluids that condense on the waste form surface may react with the glass in the sample creating a concentrated leachate (brines). The composition of such a fluid in a high solid/liquid ratio environment will be controlled by the composition of the solid waste form material. Additionally, dehydrated material deposited near the repository during the initial hot phase, with temperatures at or above the boiling point of water, will create a brine upon the initial flooding of the cooled repository.[41] This initial brine should be short lived and contact only the waste containers, not the waste form. However, if canisters are breached, then this repository brine would interact with the waste form. Appendix C contains tables with all the PCT-B solution results.

The relatively small size and interconnected channels of the zeolite framework may be extremely important in evaluating results from the different durability tests. Therefore, a detailed understanding of the ion-exchange behavior of the salt-loaded zeolite is required. The PCT-Bs were initiated with salt-loaded zeolite 4A in DIW to investigate the time-dependent release, size-fraction release, and solubility of specific components. Tests were performed on samples that were held at 90°C; for 2 minutes, 1 h, 3, 7, 28, and 91 d; with $S/V = 2000 \text{ m}^{-1}$ (Fig. 16). Additional $S/V = 20,000 \text{ m}^{-1}$ tests in DIW were also performed at 90°C for 3, 7, 28, and 91 d (Fig. 17). Results from $S/V = 2000 \text{ m}^{-1}$ and $20,000 \text{ m}^{-1}$ PCT-Bs with the salt-loaded zeolite 4A in EJ-13, 311 leachant, and ASTM Ca-Na-K-Cl and Mg-Na-K-Cl brines (Figs. 18 through 23) at 90°C for 7, 28, and 91 d were also performed. The leachate results are reported in $\mu\text{g/L}$, normalized elemental mass loss, and elemental release percent (Appendix B). A test matrix, including completed and ongoing tests may be found in Appendix B and C.

Fig. 16. Elemental Release from the 2000 m⁻¹ PCT-Bs with Salt-Loaded Zeolite 4A in DIW at 90°C (leachate pH = 10.0 after 91 days).

Fig. 17. Elemental Release from the 20,000 m⁻¹ PCT-Bs with Salt-Loaded Zeolite 4A in DIW at 90°C (leachate pH = 10.1 after 91 days).

Fig. 18. Elemental Release from the 2000 m⁻¹ PCT-Bs with Salt-Loaded Zeolite 4A in 311 Glass Leachant at 90°C (leachate pH = 9.7 after 91 days).

Fig. 19. Elemental Release from the 20,000 m⁻¹ PCT-Bs with Salt-Loaded Zeolite 4A in 311 Glass Leachant at 90°C (leachate pH = 10.1 after 91 days).

Fig. 20. Elemental Release from the 2000 m⁻¹ PCT-Bs with Salt-Loaded Zeolite 4A in EJ-13 Simulated Groundwater at 90°C (leachate pH = 9.7 after 91 days).

Fig. 21. Elemental Release from the 20,000 m⁻¹ PCT-Bs with Salt-Loaded Zeolite 4A in EJ-13 Simulated Groundwater at 90°C (leachate pH = 10.2 after 91 days).

Fig. 22. Elemental Release from the 2000 m⁻¹ PCT-Bs with Salt-Loaded Zeolite 4A in ASTM Mg-Na-K-Cl Brine at 90°C (leachate pH = 5.6 after 91 days).

Fig. 23. Elemental Release from the 2000 m⁻¹ PCT-Bs with Salt-Loaded Zeolite 4A in ASTM Ca-Na-K-Cl Brine at 90°C (leachate pH = 5.8 after 91 days).

The $ER_i\%$ results of these PCT-B experiments indicate that:

1. 20% of the Li and Cl was released within 1 h of contact with DIW.
2. 60+% of the Li was released within 3 d (all leachants except brines).
3. Release of Na and K occurred at a slower rate, with 20+% released after 3 d (all leachants except brines).
4. 10% of the zeolite Al-Si framework was also released after 3 d in DIW at 2000 m^{-1} .
5. An inverse correlation may exist between $ER_i\%$ and ionic radius and/or charge in DIW (Table VII).
6. The release rates decreased substantially after 7 d in all leachants.
7. The preferential cation release order in all the leachants except the brines at 90°C is $\text{Li} > \text{Na} \geq \text{K} > \text{Cs} > \text{Al} > \text{Si} > \text{RE} > \text{Sr} > \text{Ba}$.
8. Preferential ion exchange of Ca and Mg species from solution occurred with the Na and K in the zeolite (observed with SEM of reacted solids and solution calculations with Geochemist Workbench[42]).

TABLE VII. Comparison of ER_i% 7 d PCT-B Data with Ionic Charge and Radii

Ions	Ionic Radius (nm)	7 d ER _i % 2,000 m ⁻¹	7 d ER _i %[3] 7,500 m ⁻¹	7 d ER _i % 20,000 m ⁻¹
Li ⁺	0.060	75	68	79
Na ⁺	0.095	39	26	35
K ⁺	0.133	39	25	34
Cs ⁺	0.169	15	2.8	10
Cl ⁻	0.181	65		84
Sr ²⁺	0.113	0.05		<0.01
Ba ²⁺	0.135	<0.01		<0.01
Nd ³⁺	0.108	0.5		<0.01
Ce ³⁺	0.111	0.4		<0.01
La ³⁺	0.115	0.16		<0.01
Al ³⁺	0.050	8.9	5.3	1.2
Si ⁴⁺	0.041	7.3	4.7	0.34
pH		10.2	10.9	10.9

9. Substantial increases in RE, Sr, and Ba release were observed in the brines at 90°C, relative to the tests in DIW, EJ-13, and 311 leachants.
10. The RE elements that were released in the Ca brine leachate formed a RE-Ca-Si-Cl precipitate (observed with SEM/EDS, Table VIII).
11. The Si, Sr, Ba, and RE release rates displayed a significant pH dependence in the phosphate-buffered solutions.

TABLE VIII. Normalized Composition Results from SEM/EDS Analysis^a

Element	Salt-loaded Zeolite 4A	SLZ ^b , DIW 28d, PCT	SLZ, Mg Brine 28d, PCT	SLZ, Ca Brine 28d, PCT	SLZ, P Buffer, 3d pH = 3	RE-Ca-Si-Cl Precipitate
Na ₂ O	15.3	15.7	4.1	0.55	6.2	1.4
Al ₂ O ₃	29.6	36.5	38.9	30.3	40	4.5
SiO ₂	33.5	38.3	30.6	28.4	32	3.7
Cl	13.3	0.3	3.2	8.3	1.2	25.7
K ₂ O	5.7	3.5	6.1	0.6	1.6	2.2
CaO	0.4		0.2	26.3		13.8
Y ₂ O ₃	0.0	0.0	0	0.0	0.0	1.0
SrO	0.6	0.6	0.6	0.6	0.6	0.5
Cs ₂ O	0.0	0.0	0.0	0.0	0.1	0.1
BaO	0.0	0.0	0.0	0.0	0.0	0.4
La ₂ O ₃	1.9	1.3	1.7	1.1	0.0	2.4
Ce ₂ O ₃	0.3	3.8	0.9	1.45	0.0	17.0
Nd ₂ O ₃	0.0	0.0	0.0	0.0	0.51	20.3
LiO ₂				1.3		
P ₂ O ₃				2.3	17.6	6.8
MgO			13.6	0.18	0.4	
Total	100	100	100	100	100	100

a. Values in oxide wt% except for Cl

b. SLZ = salt-loaded zeolite

The 4- μ m diameter salt-loaded zeolite powders used in these PCT-Bs have extremely large surface areas that are not representative of the actual final waste form material nor are the results directly comparable with standardized PCTs conducted with materials that have grain sizes between 75 and 100 μ m. The results from these PCT-Bs provide information about the relative release mechanisms among the different salt-loaded zeolite components. Table VII lists the specific ER_i% data for leachate ions from 7 d PCTs (in DIW at 90°C with S/V = 2000

and $20,000 \text{ m}^{-1}$) as a function of ionic charge and radius. Elemental release results from these tests are consistent with previously published $S/V = 7500 \text{ m}^{-1}$ results[10,11] and demonstrate a possible correlation between release rate and ionic size and charge of the salt-loaded zeolite constituents, along with possible solubility effects with the $ER_i\%$ decrease of some of the components with increasing S/V . The RE, Sr, and Ba appear to be released slower than the Al and Si that comprise the zeolite framework. This suggests that preferential re-sorption of RE, Sr, and Ba ions into the remaining zeolites occurred after the dissolution of zeolites that originally contained these components.

The zeolites were somewhat corroded and a few spalled particulates were observed after a 28 d 2000 m^{-1} PCT-B in DIW at 90°C (Fig. 24). These results are consistent with the corresponding release results that indicate a significant fraction of the Li and Cl salt was released from the zeolite, while the Si and Al leachate concentrations indicate that approximately 10% of the zeolite matrix dissolved. However, the zeolites maintained much of their original cubic shape and size (Fig. 24). The amount of salt released from the zeolite is not surprising given the aggressive nature of this PCT-B with the high zeolite surface area and the initially high unsaturated conditions with the DIW.

Energy dispersive spectroscopy results from PCT-Bs with salt-loaded zeolite 4A in Mg-Na-K-Cl and Ca-Na-K-Cl brines for 28 d at 90°C indicate that some of the Na and K in the zeolite structure was replaced by Mg and to a greater extent by Ca (Table VIII). This is consistent with observed preferential absorption and ion exchange of $\text{Ca} > \text{Mg} > \text{Na}, \text{K}$ in zeolite 4A[43,44] and suggests that high-ionic-strength fluids may increase the rate at which these components are removed from the zeolites, which may be important should brines contact the waste form in the repository.[41]

Fig. 24. Secondary Electron Images from Scanning Electron Microscopy of Salt-Loaded Zeolite 4A Powder after a 28 d 2000 m^{-1} PCT-B in DIW.

In most of the tests, the pH of the system was unconstrained. However, in order to investigate the effect of pH on the durability tests, buffered leachates with defined pH were used. The $S/V = 2000 \text{ m}^{-1}$ $ER_i\%$ results of PCT-Bs with the salt-loaded zeolite 4A in phosphate pH buffered leachants at 90°C for 3 d (Figs. 25 and 26) indicate that the Si release rate was pH dependent.[45] The $ER_i\%$ of Si increased from 3 to 40% between

Fig. 25. Elemental Release from the 2000 m⁻¹ 3 day PCT-Bs with Salt-Loaded Zeolite 4A in Phosphate Buffer Solutions at 90°C.

Fig. 26. Elemental Release from the 2000 m⁻¹ 3 day PCT-Bs with Salt-Loaded Zeolite 4A in Phosphate Buffer Solutions at 90°C. Expanded y-axis provides increased separation of the RE elements.

pH 9.5 and 3, respectively. Correspondingly, the RE, Ba, and Sr $ER_i\%$ increased significantly when the pH dropped from 9.5 to 3. The Ba, Sr, and Re ions released from the sample passed through a 0.45- μm filter but not through a 0.02- μm filter, suggesting that a colloidal precipitate was forming. This is consistent with the fact that Ba, Sr, and RE ions form insoluble phosphate compounds, especially at the higher pH values. Since phosphate was also found in the zeolite after the tests (as much as 5 wt% at pH 3) it may not be an appropriate buffer for this system (Table VIII). The release rates of the singly charged ions (Li, Na, K, Cl, and Cs) were not effected by pH. This may be due to preferential ion release processes dominating the loss mechanism of these ions.

The above findings indicate that leachant composition, including pH, may significantly affect the evaluation of the durability test results for the ceramic waste form. The disproportionately high release of the alkali metals and chloride from the salt-loaded zeolites in the PCT-Bs indicates that preferential ion release (as opposed to bulk dissolution of the entire material) is the primary loss mechanism. Furthermore, the increased release of Ba, Sr, and RE ions in the presence of high-ionic-strength fluids may be important should brine-like fluids contact the waste form in the repository.

2. Material Characterization Center Test #1 (MCC-1 Test)

a. The Ceramic Waste Form with Zeolite 5A

The 3, 7, and 28 d MCC-1 tests were performed with the ceramic waste form in DIW, EJ-13, and ASTM Mg-Na-K-Cl brine leachants (Appendix III and IV). Ninety one-day MCC-1 tests in DIW were initiated but are not finished. The tests were performed in duplicate with coupons cut from the H059-1 and H059-2 samples (tests designated with a "1" or "2", respectively, in Appendix B). The leachants were passed through 0.02 and 0.45- μm filters and analyzed by ICP/MS. Leachate results are tabulated in Appendix B and

indicate that there was little difference between the two filter sizes except with RE release in the brine solutions. The 0.02- μm filters were tested and used primarily in PCT-Bs with the salt-loaded zeolite 4A (Sect. III.B.1.a) to determine the effects of the very small zeolite particles on the leachate results. In general no significant difference in leachate results can be attributed to zeolite particles smaller than 0.45 μm in these MCC-1 tests. However, sample to sample variations result in significant differences in the ionic release from the ceramic waste form.

The H059-2 sample had crystalline NaCl present (Fig. 1) and had higher alkali metal and chloride release rates in DIW relative to the H059-1 sample. This is consistent with the SEM analyses previously discussed (Sect. III.A.1), where the microsommite sample (H059-1) incorporated chloride into the crystalline structure of the material, thus providing an enhanced barrier for alkali metal ion release.

Scanning electron microscopy images of the monolithic surfaces from the 28 d MCC-1 tests revealed significantly more corrosion pitting in the regions that contained zeolites occurred in DIW, compared to EJ-13 or Mg-Na-K-Cl brine leachants (Fig. 27). The leachate analysis indicated that the Ba, Sr, and RE ions had very low release rates (at or below detection limits) in DIW and EJ-13 leachants. However, the release of these ions increased substantially in the Mg-Na-K-Cl brine. As was evident by the results obtained from the PCT-Bs with the zeolite powders, this increase has been associated with enhanced ion exchange between the zeolites and the high-ionic-strength leachant (Sect. III.B.1.a). These trends suggest that the zeolite in the ceramic waste form should preferentially retain nearly all of the contained divalent and trivalent fission products, except in the presence of a brine where retention of these ions may be more dependent on the leach resistance of the glass matrix.

One of the WAPS criteria for borosilicate glass is a comparison between the release of Li, B, and Na from the test glass and from the environmental assessment (EA)

Fig. 27

Secondary Electron Images from Scanning Electron Microscopy of H059-2 Sample Surfaces after 28 d MCC-1 Tests. (a) Deionized water; (b) EJ-13 leachant; (c) Mg-Na-K-Cl Brine. The greatest amount of corrosion occurs in DIW and manifests itself as pits in the surface regions containing zeolites.

glass. While the WAPS protocol established the PCT procedure for comparison between individual glasses, the use of the $S/V = 10 \text{ m}^{-1}$ MCC-1 test was investigated in these tests as a possible alternative procedure. The 28 d MCC-1 NL_i results obtained for the ceramic waste form compare favorably to similar results for the EA glass[46] and a Savannah River

Laboratory (SRL) composite glass[45] (Tables IX and X). The SRL composite glass was chosen for comparison here because its composition is similar to the EA glass, it contains Cs (an element not found in the EA glass), and it has reported results for brine leachants. Since B is highly soluble, it is often used as an indicator of the glass matrix reaction with the leachate. For the ceramic waste form, B is present only in the glass and thus is ideally suited to monitor the dissolution of the glass matrix. Similarly, Li is only contained in the zeolite and should provide a useful maximum measure of the alkali metal release. Cesium is contained only in the salt-loaded zeolite and the PCT-Bs with the salt-loaded zeolites indicated that Cs has the greatest release to solution of all the simulated fission products and should provide a reasonable measure for monitoring maximum radionuclide release via ion exchange with the zeolite crystals. Table IX indicates that the NL_i results of Cs, Si, Li, Na, and B from the ceramic waste form are lower than either the EA or the SRL composite glass when reacted in DIW. Similarly, the Cs, Si, and B release for the ceramic waste form are equivalent to the SRL composite glass in brine solutions (Table X).

TABLE IX. Comparison of 28 d NL_i Results (g/m^2)
from ($S/V = 10\ m^{-1}$) $90^\circ C$ MCC-1
Tests in DIW

Element	Ceramic Waste Form*	Benchmark EA Glass[46]	SRL Glass Composite[45]
Li	14.3	40.9	na
B	4.5	49.3	22.1
Na	10.9	49.3	na
Si	4.5	34.4	15.4
Cs	15.4	np	38.9

*Leachate pH = 9.

na - test data not available

np - element is not present in material

TABLE X. Comparison of 28 d NL_i Results (g/m^2) from ($S/V = 10\ m^{-1}$) $90^\circ C$ MCC-1 Tests in Mg-Na-K-Cl Brines ($\approx 25\%$ total dissolved solids)

Element	Ceramic Waste Form*	Benchmark EA Glass[46]	SRL Glass Composite[45]
B	8.7	na	8.7
Si	7.6	na	5.9
Cs	10.6	na	6.7

*Leachate pH = 5.5

na - test data not available

b. The Ceramic Waste Form with Sodalite

Leachate results from a 28 d MCC-1 test with DIW are reported in Appendix B in the row designated "MCC1A-28, F45: $\mu g/L$." The original wt% of the starting materials are reported in the rows designated Zeolite A, 311 Glass, and H005-2. These are the calculated initial configurations of the different materials. None of the elements had normalized release rates that exceeded $1\ g/(m^2 \cdot d)$. The total mass loss of the MCC-1 samples was 0.23 wt% after 3 days and 0.32 wt% after 28 days. The results of this test are similar to published data from Lewis *et. al.* [13] (MCC1ML94-28, Appendix B). A significant difference was noted for structural features of the sample surface after a 28 d MCC-1 test at $90^\circ C$ (Fig. 28). The surface was corroded with small pits ($10\ \mu m$) covering about 70% of the surface and approximately 20% of the surface had corrosion regions on the order of 100 mm (Fig. 28).

Fig. 28. Secondary Electron Images from Scanning Electron Microscopy of the H005-2 Sample after a 28 d MCC-1 Test. (a) Bright region contains high concentrations of RE elements. The surface has significant pitting due to corrosion (compare to the unreacted surface shown in Fig. 3). (b) Regions with pitting and large scale corrosion.

3. Vapor Hydration Test

a. The Ceramic Waste Form with Zeolite 5A

Coupons from the H059-1 and H059-2 samples were used to determine the utility of the VHT for providing acceptance criteria for the ceramic waste form. After termination of several of these tests at set time intervals (Appendix C), examination of the coupons indicated that the VHT samples were significantly affected by structural integrity differences in the samples. All of the H059-1 coupons were severely fractured at 120, 150, and 200°C after just 3 days of testing (Fig. 29d). These fractures are larger and are present in far greater quantity than was observed in the pretested material (Sect. III.A.2). Most of the

Fig. 29. Secondary Electron Images from Scanning Electron Microscopy of Cross-Sectioned Ceramic Waste Form Samples after VHTs at 200°C. (a) H062-3, top third of sample, 3 days; (b) H062-3, bottom third of sample, 3 days; (c) H059-2, 21 days; and (d) H059-1, 21 days. These images illustrate the amount of fracturing that occurred during the vapor hydration tests. The bright object in the bottom right corner of image (d) is due to an air bubble in the epoxy mount.

H059-1 coupons had broken apart and fallen to the bottom of the stainless steel vessels, but small parts of some coupons remained suspended, and SEM examinations were performed on those. All of the H059-2 samples held together and remained suspended in the vessels (Figs. 29c and 30). Additional 3 d VHTs on coupons from the H062-1, 2, and 3 samples (Figs. 29a and 29b) indicate that the "lighter colored" samples (Fig. 29a) had less fracturing than the darker samples (Fig. 29b). The entire test matrix for the VHTs is listed in Appendix C.

Analyses from SEM images of the cross sectioned coupons indicate that severe fracturing occurred in the samples, with longer and more fractures developing in the samples tested for longer times (compare Figs. 29a and 29c) and at higher temperatures (compare Figs. 29c with 30a and 30b).

Crystals of NaCl formed as a precipitate on the surface of the samples as a result of the VHTs (cubic or dendritic growth features in Fig. 31). Some KCl crystals, Ca-Al-Si spherical phases (small round bright spots in Fig. 30c through 30d), and Na-Al-Si-RE "stick" phases (bright "stick" features in Fig. 31d) were also observed on the surface of the samples after the VHTs. Because of the EDS detector limitations, precipitates containing Li and B that should be present on the VHT samples could not be identified.

If the relatively small quantities of the simulated fission products in the ceramic waste form are uniformly distributed throughout the sample, then they should be below the detection limits of the EDS element quantification. However, the RE elements seem to concentrate in small regions, allowing detection with EDS measurements. Patches of RE phases were present before and after the VHTs and were associated with high concentrations of chloride (bright region in Fig. 31a). Stick-like RE phases were present only after the VHTs (Fig. 31d).

Fig. 30. Secondary Electron Images from Scanning Electron Microscopy of H059-2 Ceramic Waste Form Samples after 21 d VHTs. (a) Cross-sectional view, 120°C; (b) cross-sectional view, 150°C; (c) surface, 120°C; and (d) surface, 150°C. Images (a) and (b) illustrate the increased amount of fracturing that occurs with increased temperature. The little bright round features in images (c) and (d) are alteration phases that contain high concentrations of Ca, Al, and Si.

Fig. 31. Secondary Electron Images from Scanning Electron Microscopy of the Surfaces of Ceramic Waste Form Samples after 21 d VHTs. (a) H059-2, 150°C, the large bright feature in the middle contain high concentrations of RE and chloride ions; (b) H059-2, 200°C, the dendritic growth features are NaCl crystals; (c) H059-1, 200°C, the sample illustrates the amount of cracking and NaCl formation (darker gray regions) present at the surface of the H059-1 sample; and (d) H059-1, 200°C, a magnified view of the H059-1 surface shown in image (c): (i) The large feature on the left is a dendritic growth of NaCl; (ii) The bright stick-like feature (middle right); (iii) the small gray square features of image (b) contain above average concentrations of RE and chloride ions; and (iv) a small Ca-Al-Si rich spherical feature (middle top) is also present. Pores due to corrosion are present throughout.

In general, higher temperatures increased the amount of precipitates observed on the surface. This may be associated with the increased amount of fracturing observed at higher temperatures, since the increase in exposed surface area provides more material that may be transported to the surface, or it may be due to the increase in the dissolution kinetics associated with increases in temperature.

Preliminary measurements of the rates of alteration layer formation associated with these initial VHTs are comparable to that of the SRL 202 glass.[47] Quantitative measurements of this alteration layer formation were very difficult to perform due to the excessive fracturing of the samples. The VHT provided information about the toughness and structural integrity of the ceramic waste form. However, the qualitative nature of layer thickness measurements limits the usefulness of VHT corrosion rates for qualification studies. The VHT can provide information about the ceramic waste form alteration phases that may be useful for the performance assessment modeling efforts required for geologic disposal.

b. The Ceramic Waste Form with Sodalite

Preliminary 7 and 35 d 200°C VHTs were performed with the H005-2 ceramic waste form sample with sodalite. The VHT samples developed a visible precipitation layer that covered all of the outer surfaces. Cross sections of the samples revealed the formation of three distinct layers. The interior core remained unaltered from the pretest sample. However, an intermediate region (the alteration layer) between the unaltered and precipitate regions was present in both of the VHT samples and was characterized by an increase in porosity (Fig. 32). No significant chemical composition difference between the unaltered interior of the sample and the 42 μm thick alteration layer could be detected in the

Fig. 32. Secondary Electron Images from Scanning Electron Microscopy of a H005-2 35 d VHT Sample in Cross Section. The image displays the unaltered interior, the alteration layer, and the precipitation layer of the VHT sample. The alteration layer is more porous than the unaltered material, with regions of concentrated NaCl and analcime. Analcime, calcium-silicate material (the light gray areas in the precipitation layer), and large B-Ca containing crystals precipitated at the surface of the VHT sample.

7 d VHT sample. An 18 μm thick calcium-silicate precipitate layer was also present on the 7 d VHT sample and, in combination with the high porosity layer, formed a 60 μm thick altered layer (Table XI).

TABLE XI. Normalized Energy Dispersive Microscopy of the 7 d VHT H005-2 Sample*

Element	Interior	Alt. Layer	Prec. Layer
Na ₂ O	11.5	10.0	2.7
Al ₂ O ₃	21.3	23.0	13.0
SiO ₂	44.2	46.0	48.2
Cl	5.2	3.2	2.2
K ₂ O	3.6	4.2	3.0
CaO	6.5	5.3	20.4
Y ₂ O ₃	0.2	0.2	0.6
SrO	0.1	0.0	1.6
Cs ₂ O	0.4	0.3	0.4
BaO	0.4	0.4	0.6
La ₂ O ₃	0.1	0.0	0.3
Ce ₂ O ₃	0.1	0.1	0.1
Nd ₂ O ₃	0.2	0.1	0.1
LiO ₂	0.6	0.6	0.6
B ₂ O ₃	6.9	6.9	6.9
Total	100	100	100

*Except for Cl, values in oxide wt%. Lithium and boron could not be accurately measured with EDS, but were accounted for in the compositional analysis (0.65 and 6.95 wt% of Li and B oxides, respectively for the H005-2 samples).

Interior: Measured composition of the unaltered sample area.

Alt. Layer: Measured composition of the alteration layer .

Prec. Layer: Measured composition of the precipitation layer.

The 260 μm thick alteration layer of the 35 d VHT sample segmented into two inter-grown zones of irregular thickness (Fig. 32). One zone preferentially contained Na and Cl, while the other zone had a chemical composition consistent with analcime ($\text{NaAlSi}_2\text{O}_6 \cdot \text{H}_2\text{O}$). An 80 to 120 μm thick precipitate layer at the surface was also present on the 35 d VHT sample (Fig. 32). This precipitate layer had analcime crystals (pyramid-shaped crystals in Fig. 33), areas with calcium-silicate (the lighter fine-grained coral like regions of Figs. 32 and 33), and large bladed crystals of calcium-boron oxide (the large crystal on the right side of Fig. 33) (Table XII). The 35 d, 200°C VHT sample had a mass gain of 4.26%, indicating that approximately 3% of the initial water in the vessel was absorbed into the sample and/or its alteration phases.

IV. DISCUSSION

This work evaluates the utility of standard durability tests as an initial step in developing methods and criteria that can be applied towards the process of qualifying the ceramic waste form for acceptance into the Civilian Radioactive Waste Management System. Since the relatively new ceramic waste form has not been extensively studied for this purpose, a broad range of potential repository conditions are being evaluated to determine the bounding parameters appropriate for the corrosion testing of the ceramic waste form and its behavior under accelerated testing conditions. As part of the test development, extensive characterization of the ceramic waste form before (to provide a baseline for the test results) and after testing was required, as well as an investigation into the usefulness of specific characterization methods and sample preparation techniques.

The characterization of the ceramic waste form for this project has mainly been performed with the use of SEM/EDS, XRD, and AEM. Techniques like XRD and SEM/EDS provide very useful structural and compositional information, respectively. For example, XRD was used in the experiments in which the sodium oxide presence in the glass matrix was

100 μm

Fig. 33. Secondary Electron Images from Scanning Electron Microscopy of the Exposed Surface of a H005-2 35 d VHT Sample. Analcime (pyramid shaped crystals), calcium-silicate material (the light gray fine-grained precipitate), and large B-Ca containing crystals precipitated at the surface of the VHT sample.

TABLE XII. Normalized Energy Dispersive Microscopy of the 35 d VHT H005-2 Sample*

Element	Analcime	Ca-Si	Ca, B-Oxide	Interior	Alt. Lay. Rough	A. L. Smooth	Ppt. L. Rough	Ppt. L. Smooth
Na ₂ O	15.4	3.0	0.0	12.0	21.0	13.4	6.5	11.0
Al ₂ O ₃	26.1	4.3	0.0	21.0	12.0	23.9	5.5	21.0
SiO ₂	56.7	40.9	2.6	43.5	25.0	49.7	40.0	52.0
Cl	0.2	11.8	0.0	5.3	26.0	1.5	8.0	0.3
K ₂ O	0.3	5.2	0.2	3.3	4.0	0.5	3.0	0.8
CaO	0.3	33.3	66.5	6.0	3.0	2.8	27.0	6.0
Y ₂ O ₃	0.2	0.8	0.5	0.1	0.1	0.1	0.4	0.3
SrO	0.0	0.0	0.5	0.0	1.3	0.0	0.2	0.0
Cs ₂ O	0.5	0.2	0.0	0.5	0.4	0.5	0.2	0.8
BaO	0.1	0.3	0.0	0.6	1.0	0.3	0.2	0.2
La ₂ O ₃	0.0	0.3	0.7	0.2	0.0	0.1	0.1	0.0
Ce ₂ O ₃	0.1	0.3	0.4	0.1	0.1	0.0	0.0	0.0
Nd ₂ O ₃	0.1	0.1	0.1	0.1	0.2	0.2	0.0	0.0
LiO ₂				0.6	0.6	0.6	0.6	0.6
B ₂ O ₃			30	6.9	6.9	6.9	6.9	6.9
Total	100	100	100	100	100	100	100	100

*Except for Cl, values in oxide wt%. Lithium and boron could not be accurately measured with EDS, but were accounted for in the compositional analysis (0.65 and 6.95 wt% of Li and B oxides, respectively for the H005-2 samples).

Analclime: Measured composition of the analclime crystals on the surface.

Ca-Si: Measured composition of the Ca-Si areas on the surface.

Ca, B-Oxide: Measured composition; Ca, B-Oxide crystals on the surface.

Interior: Measured composition of the unaltered area.

Alt. Lay Rough: Measured composition of the alteration layer with NaCl.

A. L. Smooth: Measured composition of the alteration layer with analclime.

Ppt. L. Rough: Measured composition of the precipitation layer with calcium silicate.

Ppt. L. Smooth: Measured composition of the precipitation layer with analclime.

identified as the primary mechanism for the accelerated structural transformation of the salt-loaded zeolite 4A to sodalite and to identify the primary crystalline structures of the ceramic waste forms. The higher resolution of the AEM work on the ceramic waste form sample with sodalite (H005-2) was used to investigate the submicrometer structures of the individual components. The zeolite was found to be weakly bonded to the glass and easily separated during the sample sectioning process. Ion milling may be investigated as a method for producing a sample suitable for studying the zeolite-glass interface.

The structural transformations of the zeolite to sodalite and microsommitite, along with the development of feldspar structures in the ceramic waste forms may necessitate a careful identification of the material after processing to comply with the acceptance requirements for crystalline phase identification. While the major phases are easily identified with XRD, electron diffraction techniques may need to be employed to provide the information about the minor phases.

Methods of preparing samples for the durability tests were also investigated to determine the potential effect of water contact on the test samples. The initial leachate results from the 1 h PCTs of the salt-loaded zeolite (Appendix B) and from the DIW used as cutting fluid from the coring process (Table III) both indicated a relatively high release of alkali metal and chloride ions. However, the MCC-1 results (Sect. III.B.2) suggest that the release of salt from the ceramic waste form monoliths is relatively low. These observations indicate that the alkali metals and chloride are released relatively quickly from the zeolites exposed to the leachant, but that the glass matrix impedes the release of the alkali metals and chloride in the ceramic waste form monoliths. However, the use of water for cutting and polishing of monoliths used in the durability tests and for characterization will affect the total salt in the sample. Therefore, when investigating surface effects or samples with high surface areas (as in ground particles for PCTs), the use of water for sample preparation should be limited.

The disproportionately high release of the alkali metals and chloride from the salt-loaded zeolites in the PCT-Bs indicates that preferential ion release (as opposed to bulk dissolution of the entire material; Sect. III.B.1.a) is the primary loss mechanism. The elemental release of a specific component in the salt-loaded zeolite or the ceramic waste form provides meaningful information about the release mechanisms and relative release order between each element. The high surface areas of the PCT-Bs with the 4 μm diameter salt-loaded zeolite accelerates the release rates of the tests such that the use of a NL_i that divides the release by these large surface areas of the zeolites cannot be compared directly to standard PCTs.[10,28] The NL_i is usually associated with the dissolution of the material (primarily a surface effect) and accounts for the external geometric surface area involved. However, if ion exchange or preferential ion release is occurring, then the external geometric surface area is less important and the correlation between the amount released compared to the total amount of an element in the sample (as given with $ER_i\%$) may be more useful. The latter is useful for evaluating ion release from the salt-loaded zeolites. However, geometric effects are important and must be considered for any meaningful evaluation.

With preferential ion release from the salt-loaded zeolite in the PCT-Bs, a correlation between the release order of the individual elements occluded in the zeolite matrix and the ionic radius and ionic charge may exist. However, the order of cation release from the salt-loaded zeolites is affected by the leachant solubility of specific ions. Thus, the presence of specific ions in the leachant has dramatic effects on the ion exchange properties of the zeolite. These effects were most evident with the salt-loaded zeolite and ceramic waste form samples in the Mg-Na-K-Cl and Ca-Na-K-Cl brines. In general, Sr, Ba, and RE ions have very low release rates from the zeolites in DIW and low-ionic-strength leachants. In the high-ionic-strength brines, Mg and Ca were preferentially exchanged in the zeolites for Na and K, and the equilibrium conditions in the zeolites were changed such that significant fractions of the alkaline earth and RE ions were released. Furthermore, Ca, and perhaps Mg, forms precipitates with the RE ions.

A more complete mechanistic understanding of these preferential ion release and ion exchange effects will require calculations that account for the different solubility effects and chemical reactions that are occurring in the system. This work will be performed with the use of programs like Geochemist Workbench[42] and other chemical calculations.

The initial PCT-Bs with salt-loaded zeolites utilized 0.02 μm filters to ensure the separation of very small zeolite particles from the leachates, before analysis. However, the difference between the leachate results that used 0.02 and 0.45 μm filters were sufficiently indistinguishable with regards to zeolite contamination of the leachates. Thus, the use of the 0.02 μm filter will be discontinued and all subsequent leachates will be filtered with only the 0.45 μm filter.

The acceleration of zeolite structural changes when in contact with the 311 glass, the changes in corrosion associated with glass composition, and the different amounts of stress fracturing in several of the samples, all indicate that the ceramic waste form is very sensitive to processing conditions and the composition of the materials used. The formation of stress fractures in the hot isostatically pressed samples adversely affects the evaluation of the VHT samples and may affect the analysis of the data from MCC-1 and PCTs (to be performed), due to increases in the exposed sample surface area during testing. Results from PCTs should be the least affected by fracturing, since the samples are crushed and the surface areas are already large. The presence of fractures in the pretested material suggests that fracture propagation can be decreased with slower cooling rates, appropriate annealing, or crack inhibition materials. The increased time at the elevated temperatures and pressures associated with the hot isostatic pressing process induces increased structural transformation changes from zeolite to microsommitite and sodalite. The initial investigation performed here (Sect. III.A.1.b) suggests that extended processing times may be achievable if the amount of sodium oxide present in the glass is adjusted to minimize solid state diffusion effects that lead to an increased conversion rate of zeolite to sodalite.

While the VHT[32] provided information on corrosion rates, alteration phase formation, material toughness, and structural integrity, the test does have serious difficulties with regards to the relatively detailed sample preparation and handling required before and after the test and the qualitative nature of layer thickness measurements that limits the usefulness of the VHT for acceptance studies. Comparison of the alteration layer thickness between ceramic waste forms that used the 57 and the 311 glass suggests that the reduction of Ca as well as the increase of Si in the glass compositions (Appendix B) greatly enhanced the durability of the ceramic waste form and decreased the rate of alteration and the amount of precipitate formation. The temperatures used to accelerate the reaction product formation are higher than those expected in the repository, and extreme care must be used to ensure that the elevated temperatures do not change the reaction mechanisms. The VHT can provide long-term alteration phase information about the ceramic waste form that may be useful for the performance assessment modeling required for geologic disposal.

The use of other durability tests, like the unsaturated drip test[33] and the accelerated dissolution test[34], with the ceramic waste form may be investigated in future studies. However, the preliminary results discussed here indicates that the PCT-B and MCC-1 tests with different leachants should provide sufficient information for establishing the consistency criteria required for acceptance of the ceramic waste form into the Civilian Radioactive Waste Management System. Additionally, TCLP tests for components of the ceramic waste form, which may be considered hazardous materials, may also have to be performed to meet hazardous waste criteria.

V. CONCLUSIONS AND RECOMMENDATIONS

Scanning electron microscopy, EDS, AEM, and XRD were successfully used to provide information about the morphology, composition, structure, and bonding of the ceramic waste form with zeolite and sodalite, and its precursor materials. The applicability of

the MCC-1, PCT-B, and VHT protocols to the qualification of the ceramic waste form has been investigated. This investigation considered the effects of pH, leachant composition, sample S/V, and test temperature. These initial testing protocols have been used for the testing of high-level borosilicate waste glass.

The salt-loaded zeolite in the ceramic waste form samples was uniformly distributed in the glass matrix for scales larger than 100 μm . The larger glass fragments, with particle diameters as large as 45 μm , concentrated the 4 μm diameter salt-loaded zeolites such that a significant fraction of the zeolites are in contact with each other. The small scale (on the order of 25 μm) conglomeration of the zeolites may form channels that provide water access to the interior zeolites.

Composition analysis from SEM/EDS of ceramic waste form samples agreed reasonably well with the calculated values. Results from SEM/EDS and AEM analyses indicated that regions with anomalously high concentrations of RE elements were present in the samples. However, Sr, Ba, and RE elements appeared to remain in the ceramic waste form structure, even after durability tests in DIW.

Salt-loaded zeolite, sodalite, and nepheline were observed to have a cubic morphology with edge lengths up to 4 μm . The salt-loaded zeolite in open crucible heating tests transformed to nepheline or sodalite within 24 hours at 700°C. An accelerated transformation to sodalite after 4 hours at 700°C was observed when the salt-loaded zeolite was in contact with glass containing sodium oxide. Presumably, the sodium oxide enhances specific ion exchange between the salt-loaded zeolite and the glass that accelerates the transformation to sodalite. Boron, Ca, K, Li, NaCl, and water were eliminated as the primary components associated with the accelerated transformation to sodalite.

As observed in PCT-Bs with salt-loaded zeolite, preferential ion release was the dominant loss mechanism. The univalent ions are preferentially released over the divalent and trivalent ions in DIW and EJ-13 leachant. The order of cation release from the salt-loaded zeolites is affected by the leachant composition of specific ions. Preferential ion exchange between Ca, Mg, and phosphate in the leachant with the Na and K in the salt-loaded zeolite was observed in the PCT-Bs with the 4 μ m zeolite powders. Most of the Li and Cl were released from the salt-loaded zeolite within a few hours in PCT-Bs with DIW. These PCT-Bs, with only salt-loaded zeolite, have extremely high surface areas, creating very accelerated conditions. Therefore, the results from these PCT-Bs provide information about the relative release rates and mechanisms among the different salt-loaded zeolite constituents, but may not be directly comparable with other PCT results from other glass waste forms that were conducted with a coarser grained material.

The utility of the VHT and MCC-1 tests for elucidating the criteria for acceptance of the ceramic waste form into the DOE Civilian Radioactive Waste Management System was investigated. The preliminary results indicate that the MCC-1 tests can provide the relevant information required for acceptance of the ceramic waste form with regards to consistency. Additional PCT-B investigations with salt-loaded zeolite 4A powders were performed to determine the release mechanisms and the effects of leachants on the zeolite. The use of DIW as a leachant provides the upper bound for the corrosion of the ceramic waste form where the system pH is neutral or basic. Acidic leachants increased the dissolution of the zeolite framework. The use of high-ionic-strength brine leachants increased the release of the alkaline earth and RE elements from the salt-loaded zeolite, and Ca, Mg, and P species were preferentially ion exchanged for the zeolite cations Na and K. These results indicate that both DIW and other leachants, which specifically affect the ion exchange behavior of the fission products, are required to fully assess the durability of the ceramic waste form. Furthermore, to avoid premature dissolution of alkali metal and chloride ions, exposure to water should be limited in any of the sample preparation steps.

The effect of the 57 and 311 glass compositions on corrosion rates and alteration phase formation was investigated with VHTs between 120 and 200°C. The reduction of Ca as well as the increase of Si in the glass compositions greatly enhanced the durability of the ceramic waste form, decreasing the rate of alteration and the amount of precipitate formation. The VHT provided information about the toughness and structural integrity of the ceramic waste form. However, the qualitative nature of layer thickness measurements limits the usefulness of VHT corrosion rates for qualification studies. The VHT can provide information about the alteration phases of the ceramic waste form that may be useful for the performance assessment modeling efforts required for geologic disposal.

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APPENDIX A PREPARATION OF SAMPLES AND SOLUTIONS

Zeolite Dehydration

Zeolite 4A ($\text{Na}_{12}[(\text{AlO}_2)_{12}(\text{SiO}_2)_{12}]$) and zeolite 5A ($[\text{Ca}_4\text{Na}_4][(\text{AlO}_2)_{12}(\text{SiO}_2)_{12}]$) were obtained from UOP (Des Plaines, IL). The powder was loaded into a can and heated stepwise in three stages under flowing nitrogen gas to a maximum of 525°C. The heat was maintained at each stage until no change in relative humidity over time could be detected.

After cooling, the dried zeolite was stored in a sealed container under an *inert* atmosphere.

Simulated Waste Salts

Simulated waste salts represent the LiCl-KCl salt that contains certain fission products and transuranics after the electrometallurgical treatment of spent fuel. A LiCl-KCl salt was blended with anhydrous forms of other salts so that the resulting material would contain approximately 10 wt% fission products comprising SrCl_2 , CsCl , and BaCl_2 and other elements such as cerium, lanthanum, neodymium, yttrium, and possibly plutonium.

Zeolite Ion Exchange and Salt-Loading with the Simulated Waste

Under an argon atmosphere, 21 g of simulated waste salt was mixed with 79 g of zeolite in a rotating blending chamber at 550°C for 24-48 h.

This process achieved 20-22 wt% salt loading, with 4 to 11 chloride ions per unit cell. A pressurized column technique may increase the fission product loading.

Boroaluminosilicate Glasses

A comparison of the glass composition we used here, in wt%, is as follows:

	311	57	K-Si
Al ₂ O ₃	9.7	6.8	9.8
B ₂ O ₃	13.9	19.1	14.0
CaO	13.5	0.8	13.6
K ₂ O	0.8	0.5	5.1
Li ₂ O	—	—	1.8
MgO	0.4	—	—
Na ₂ O	6.5	7.1	—
SiO ₂	55.0	66.5	55.7
SiO	0.1	—	—
ZnO ₂	<0.1	—	—

K-Li Glass Formulation

Essentially, K-Li glass is 311 glass with all of the sodium (~4.8 wt%) replaced by a eutectic K-Li salt mixture (~43 M% K and 57 M% Li).

To prepare a 50-g batch, the components were thoroughly blended in the following properties: 9.3 wt% Al₂O₃, 13.3 wt% B₂O₃, 12.9 wt% CaO, 7.5 wt% K₂CO₃, 52.8 wt% SiO₂, and 4.2 wt% Li₂CO₃.

The mixture was transferred to a platinum crucible, sintered at 1250°C for 30 min, and then heated at 1350°C for 2 h for complete glass formation. Phase separation upon cooling caused some areas of the glass to become milky, while other areas were completely clear. The clear part was pulverized with a hammer for mixing with zeolite.

Glass-Zeolite Blending

Salt-loaded zeolite and glass were ground into fine powders in an industrial grinder; 57 glass was sieved through 325 mesh.

Equal amounts by weight of glass and zeolite were thoroughly mixed and mechanically pressed to 70 MPa in 2.57-cm-diameter 304 ss canisters.

The tops were welded to the canisters in air, then the canisters were evacuated at 500°C for 16 to 48 h, before being placed in the hot isostatic press.

Formation of Glass-Bonded Salt-Loaded Zeolite and Sodalite

Bonding was achieved by hot isostatic pressing of the sample at a maximum isostatic load of 28 MPa, then cooling for 2 h.

Samples H020-1 to -4 comprised 311 glass with salt-loaded zeolite 4A. They were hot isostatically pressed at a maximum of 600°C for 4 h.

Sample H005-2 comprised 311 glass with salt-loaded zeolite 4A that was hot isostatically pressed for 2 h.

Samples H059-1 and -2 and H062-1 to -3 comprised 57 glass with salt-loaded zeolite 5A. The samples were placed in canisters that were loaded into the hot isostatic press and heated in a "bump and run" process as follows: heat at 20°C/min to 500°C; hold at 500°C for 20 min; heat at 20°C/min to 750°C; immediately (without holding) cool at 20°C/min to 600°C; hold at 600° for 1 h; cool at 50°C/min to room temperature.

Samples were stored in a sealed desiccator.

Preparation of Ceramic Waste Form Samples for Durability Tests

Samples were sliced with a diamond blade. They were dry cut (without water or other lubricants) to avoid leaching and contamination. A dry emery cloth was used for polishing. Core samples were obtained with a water-cooled coring bit.

Preparation of Leachant Solutions

Four leachant solutions were chosen to simulate expected conditions that the waste form may encounter: Mg-Na-K-Cl and Ca-Na-K-Cl brines, EJ-13 and 311 glass leachants. The EJ-13 is a potential leachant that may be encountered in the geologic repository at Yucca Mountain, Nevada, and is made of material specific to that location. The 311 glass leachant is a potential leachant that the ceramic waste form may encounter when dissolution of glass materials in the waste form occurs. Both were prepared according to procedures outlined in Document No. DP-05-117 (Equilibration of EJ-13 Water) of the ANL HLW Glass Testing Program. The EJ-13 and 311 glass leachants were prepared by reacting powdered material with J-13 and DIW, respectively, for 30 days at 90°C. The two brines are concentrated leachants that may form when condensed fluid reacts with the glass in the waste form creating thin liquid films. They were prepared in accordance with ASTM standards (ASTM Method C1220-92). Accordingly, either 495.8 g of $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$, for Mg-Na-K-Cl brine, or 358.1 g of $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$, for Ca-Na-K-Cl brine, were added to 96.4 g of KCl and 180 g of NaCl. They were added to sufficient DIW to make 2000 mL of brine. The pH was adjusted to 6.5 with 0.01 M NaOH.

APPENDIX B

LEACHATE ANALYSES FROM DURABILITY TESTING

Normalized release rates (NRR), normalized elemental mass loss (NL_i), and elemental release percent ($ER_i\%$) are defined as:

$$NRR = C_i V_s / f_i A t \text{ (g/m}^2\cdot\text{d)}$$

$$NL_i = C_i V_s / f_i A \text{ (g/m}^2\text{)},$$

$$\& \quad ER_i\% = m_i / M_i = C_i V_s / f_i M$$

where, C_i = concentration of i th element released to the solution ($\mu\text{g/L}$)

V_s = volume of leachate (m^3)

f_i = fraction of i th element in the sample

A = surface area of the sample (m^2)

t = duration of test in days (d)

m_i = mass of i th element in solution (g)

M_i = total mass of the i th element in the sample (g)

M = total mass of the sample (g).

Leachate results from MCC-1 tests and PCT-Bs of the ceramic waste form and the precursor material of salt-loaded zeolite 4A in DIW, EJ-13, and ASTM Mg-Na-K-Cl and Ca-Na-K-Cl brines. The results are reported for each constituent of the sample in parts per billion (ppb), NL_i , and/or $ER_i\%$.

The symbols used in the table are as follows:

Sample Name Column;

- M The first letter in the sample name, indicating a MCC-1 test.
- GZ Letters in the sample name, indicating a GBZ sample.
- D Letter in the sample name, indicating DIW leachate.
- EJ Letters in the sample name, indicating an EJ-13 leachate.
- GB Letters in the sample name, indicating a 311 leachate.
- CB Letters in the sample name, indicating a Ca ASTM brine leachate.
- B Letter in the sample name, indicating a Mg ASTM brine leachate.
- h This additional letter in the sample name indicates a 20,000 m⁻¹ test.
The absence of this letter indicates a 2,000 m⁻¹ test.
- Ca Indicates the Ca-Na salt-loaded zeolite 5A was used in the test.
- ***XX* Number of days the test ran.
- ***-**-X Sample number of the specific test.
- F02 Indicates leachates filtered with a 0.05 µm filter.
- F50 Indicates leachates filtered with a 0.005 µm filter.
- F45 Indicates leachates filtered with a 0.45 µm filter.

TABLE B1. Leachate Analyses Results

Test	#	S/V	Meas.	Filter	days	Li	B	Na	Mg	Al	Si	Cl	K	Ca	Y	Sr	Cs	Ba	La	Ce	Nd	Pr	C(inor)	O	
MW						6.94	10.81	22.99	24.31	26.98	28.09	35.45	39.10	40.08	88.91	87.62	132.91	137.34	138.91	140.12	144.24	141		16.00	
Wt% of Simulated Waste Salt						6.47		0.72				61.50	26.14		0.05	0.30	3.00	1.00	0.60	1.14	1.84				
Cl Wt% of Salt						39.52		1.83					49.84		0.10	0.54	3.80	1.52	1.06	2.01	3.20				
Mole% of Salt						27.73		0.93				51.59	19.88		0.02	0.10	0.67	0.22	0.13	0.24	0.38				
wt% Zeolite 4A								16.18		18.99	19.77													45.05	
wt% Salt Loaded Zeolite 4A						1.36		12.93		15.00	15.62	12.92	5.49		0.01	0.06	0.63	0.21	0.13	0.24	0.39			35.59	
Mole% Salt Loaded Zeolite 4A						4.27		12.26		12.11	12.11	7.94	3.06		0.00	0.02	0.10	0.03	0.02	0.04	0.06			48.46	
						1.34	0.00	11.00		13.00	13.00	11.00	6.00		0.01	0.06	0.63	0.21	0.12	0.08	0.12				
MW						6.94	10.81	22.99	24.31	26.98	28.09	35.45	39.10	40.08	88.91	87.62	132.91	137.34	138.91	140.12	144.24	141		16.00	
Cl Wt% of Salt						40.6							48.6		0.1	0.4	2.3	1	0.6	1.9	3.8	0.8			
Wt% pf Salt						6.65						61.04	25.49		0.05	0.22	1.82	0.66	0.34	1.08	2.19	0.46			
Mole% of Salt						28.42						51.09	19.34		0.02	0.07	0.41	0.14	0.07	0.23	0.45	0.10			
wt% Zeolite 5A								5.47		19.26	20.05			9.54										45.68	
wt% Salt Loaded Zeolite 5A						1.40		4.32		15.22	15.84	12.82	5.35	7.54	0.01	0.05	0.38	0.14	0.07	0.23	0.46	0.10		36.09	
Mole% Salt Loaded Zeolite 5A						4.51		4.21		12.64	12.64	8.11	3.07	4.22	0.00	0.01	0.06	0.02	0.01	0.04	0.07	0.02		50.57	
wt% 57 glass							4.92	4.95		3.83	28.60		0.51	0.84	0.00	0.01		0.00							
wt% H059-1,2; H062-1,2,3						0.70	2.46	4.64		9.52	22.22	6.41	2.93	4.19	0.01	0.03	0.19	0.07	0.04	0.11	0.23	0.05		18.04	
Leachate results from a 28 day MCC-1 test with the H005-2 sample in deionized water.																									
Test	#	S/V	Meas.	Filter	days	Li	B	Na	Mg	Al	Si	Cl	K	Ca	Y	Sr	Cs	Ba	La	Ce	Nd	pH	C(inor)	O	
wt% Salt Loaded Zeolite 4A						1.36	0.00	12.93	0.00	15.00	15.62	12.92	5.49	0.00	0.01	0.06	0.63	0.21	0.13	0.24	0.39				
wt% 311 glass						0.00	4.32	4.83	0.24	5.14	25.70	0.00	0.67	9.64	0.00	0.00	0.00	0.00	0.00	0.00	0.00				
wt% H005-2						0.68	2.16	8.88	0.12	10.07	20.66	6.46	3.08	4.82	0.01	0.03	0.32	0.11	0.07	0.12	0.20				
MCC1A	1	10	µg/L	0.45	28	1.1E+3	1.7E+3	7.4E+3	1.3E+2	7.3E+3	1.2E+4	5.5E+3	2.5E+3	2.9E+3		3.1E+1	2.1E+2	2.2E+1	3.0E+0	5.1E+0	9.0E+0	8.0	2.1E+3		
MCC1A	1	10	ER%	0.45	28	6.4E-1	3.3E-1	3.4E-1	4.4E-1	3.0E-1	2.5E-1	3.5E-1	3.3E-1	2.5E-1		4.3E-1	2.8E-1	8.6E-2	1.9E-2	1.8E-2	1.9E-2				
MCC1A	1	10	NLi	0.45	28	5.5E-1	2.8E-1	3.0E-1	3.8E-1	2.6E-1	2.2E-1	3.0E-1	2.9E-1	2.2E-1		3.7E-1	2.4E-1	7.4E-2	1.6E-2	1.5E-2	1.6E-2				
MCC1ML9428:			ER%		28	5.4E-1	2.8E-1	6.1E-1		2.7E-1	2.3E-1		6.6E-1	1.7E-1		2.1E-1	5.4E-1	1.9E-1							
MCC1ML9428:			NLi		28	6.0E-1	3.1E-1	6.7E-1		3.0E-1	2.5E-1		7.3E-1	1.9E-1		2.3E-1	6.0E-1	2.1E-1							

TABLE B1. (contd.)

Leachate results from MCC-1 tests with the H059-1,2 samples in deionized water, EJ-13 leachant, and Mg-Na-K-Cl brine.																								
Test	#	S/V	Meas.	Filter	days	Li	B	Na	Mg	Al	Si	Cl	K	Ca	Y	Sr	Cs	Ba	La	Ce	Nd	pH	C(inor)	O
wt% H059-1,2; H062-1,2,3						0.70	2.46	4.64		9.52	22.22	6.41	2.93	4.19	0.01	0.03	0.19	0.07	0.04	0.11	0.23			1.8E+1
MGZD	1	10	µg/L	0.02	3	2.6E+2	1.1E+2	3.7E+3	4.1E+0	2.9E+3	5.3E+3	5.0E+3	9.7E+2	2.6E+3		2.2E+1	7.4E+1	1.0E+1	1.2E-2	1.7E-2	3.8E-2	8.7	4.7E+3	
MGZD	1	10	µg/L	0.45	3	2.9E+2	1.3E+2	5.1E+3	5.4E+0	3.7E+3	6.0E+3	5.0E+3	1.1E+3	2.6E+3	5.1E-2	2.6E+1	8.0E+1	3.5E+1	7.0E-2	8.4E-2	1.3E-1	8.7	4.7E+3	
MGZD	2	10	µg/L	0.02	3	3.6E+2	1.5E+2	3.3E+3	5.5E+0	2.0E+3	4.1E+3	5.4E+3	1.6E+3	1.3E+3	1.9E-2	2.9E+0	1.0E+2	2.3E+0	3.0E-2	4.9E-2	6.8E-2	8.7	2.0E+3	
MGZD	2	10	µg/L	0.45	3	3.7E+2	4.0E+2	3.9E+3	4.6E+0	2.3E+3	4.2E+3	5.4E+3	1.8E+3	1.2E+3		2.7E+0	9.6E+1	5.6E+0	3.0E-2	3.5E-2	6.8E-2	8.7	2.0E+3	
MGZD	3	10	µg/L	0.02	3	3.7E+2	3.7E+2	3.2E+3	3.8E+0	2.1E+3	4.0E+3	5.0E+3	1.6E+3	1.3E+3		2.6E+0	9.9E+1	2.3E+0	1.1E-2	2.2E-2	3.0E-3	8.7	2.1E+3	
MGZD	3	10	µg/L	0.45	3	3.7E+2	1.4E+2	3.1E+3	3.4E+0	2.5E+3	4.2E+3	5.0E+3	1.7E+3	1.3E+3		2.7E+0	1.0E+2	3.9E+0	2.7E-2	3.6E-2	2.3E-2	8.7	2.1E+3	
MGZD	1	10	µg/L	0.02	7	3.0E+2	1.1E+2	3.2E+3	3.9E+0	4.0E+3	6.3E+3	5.9E+3	1.2E+3	3.0E+3	2.0E-2	2.5E+1	6.9E+1	2.0E+1	4.0E-2	2.2E-2	1.0E-2	8.7	3.9E+3	
MGZD	1	10	µg/L	0.45	7	2.9E+2	1.1E+2	3.1E+3	4.1E+0	4.3E+3	6.4E+3	5.9E+3	1.2E+3	2.9E+3		2.4E+1	6.7E+1	2.6E+1	5.8E-2	9.9E-2	1.3E-1	8.7	3.9E+3	
MGZD	2	10	µg/L	0.02	7	5.7E+2	5.7E+2	3.5E+3	8.6E+0	3.7E+3	5.9E+3	7.6E+3	2.4E+3	1.8E+3		9.4E+1	1.5E+2	2.7E+0	6.0E-2	1.2E-1	7.2E-2	9.2	4.1E+3	
MGZD	2	10	µg/L	0.45	7	5.7E+2	5.9E+2	3.8E+3	5.0E+0	3.8E+3	6.1E+3	7.6E+3	2.3E+3	1.7E+3	7.1E-3	2.7E+0	1.4E+2	4.3E+0	1.2E-1	4.0E-1	6.7E-1	9.2	4.1E+3	
MGZD	1	10	µg/L	0.02	28	5.9E+2	6.2E+2	5.4E+3	5.5E+0	4.4E+3	8.9E+3	1.8E+4	1.6E+3	4.0E+3		2.7E+1	1.2E+2	1.7E+1	1.3E-2	3.6E-2	4.3E-2	9.2	1.2E+1	
MGZD	1	10	µg/L	0.45	28	4.1E+2	4.5E+2	3.8E+3	4.7E+0	3.5E+3	6.4E+3	1.8E+4	1.2E+3	2.8E+3		2.0E+1	8.2E+1	1.8E+1	2.1E-2	7.2E-2	1.1E-1	9.2	1.2E+1	
MGZD	2	10	µg/L	0.02	28	9.7E+2	1.1E+3	6.1E+3	7.8E+0	5.6E+3	9.6E+3	2.0E+4	3.7E+3	2.1E+3		2.3E+0	4.7E+2	4.2E-1	1.1E-2	3.5E-2	8.6E-2	9.2	5.0E+2	
MGZD	2	10	µg/L	0.45	28	9.8E+2	1.1E+3	6.0E+3	7.8E+0	6.1E+3	1.0E+4	2.0E+4	3.6E+3	2.3E+3	1.6E-2	3.2E+0	4.9E+2	4.8E+0	3.4E-1	1.3E+0	2.1E+0	9.2	5.0E+2	
MGZD	1	10	µg/L	0.02	91																			
MGZD	1	10	µg/L	0.45	91																			
MGZD	2	10	µg/L	0.02	91																			
MGZD	2	10	µg/L	0.45	91																			
MGZEJ	1	10	µg/L	0.02	3	1.4E+2	2.1E+2	5.2E+3	2.5E+1	1.5E+3	1.2E+4	1.1E+4	5.1E+3	3.1E+3	4.0E-2	2.2E+1	2.7E+1	9.3E-2	3.0E-2	5.3E-2	7.3E-2	9.8	1.8E+4	
MGZEJ	1	10	µg/L	0.45	3	1.4E+2	1.9E+2	5.2E+3	3.6E+1	8.1E+2	1.2E+4	1.1E+4	5.0E+3	3.8E+3	1.3E-2	2.8E+1	2.7E+1	9.6E+0	1.1E-1	1.1E-1	2.1E-1	9.8	1.8E+4	
MGZEJ	2	10	µg/L	0.02	3	2.1E+2	1.9E+2	5.1E+3	2.5E+1	1.2E+3	1.2E+4	1.3E+4	5.5E+3	3.5E+3	1.0E-2	1.4E+1	8.1E+1	3.8E-1	2.1E-2	2.0E-2	1.0E-2	9.8	1.8E+4	
MGZEJ	2	10	µg/L	0.45	3	2.1E+2	1.9E+2	5.3E+3	3.3E+1	8.0E+2	1.3E+4	1.3E+4	5.8E+3	3.7E+3	6.0E-3	1.6E+1	7.6E+1	4.5E+0	2.2E-1	1.2E+0	3.1E-1	9.8	1.8E+4	
MGZEJ	1	10	µg/L	0.02	28	5.6E+2	3.0E+2	5.2E+3	8.6E+0	2.0E+3	1.2E+4	1.3E+4	4.1E+3	2.6E+3	1.2E-2	1.9E+1	1.2E+2	5.0E-1	7.2E-2	1.3E-1	1.4E-1	10.0	3.0E+3	
MGZEJ	1	10	µg/L	0.45	28	5.9E+2	3.1E+2	5.5E+3	2.6E+1	1.0E+3	1.3E+4	1.3E+4	4.4E+3	3.6E+3	1.8E-2	2.5E+1	1.2E+2	1.4E+1	9.1E-2	1.6E-1	2.5E-1	10.0	3.0E+3	
MGZEJ	2	10	µg/L	0.02	28	6.9E+2	4.3E+2	5.3E+3	3.0E+1	1.8E+3	1.3E+4	1.5E+4	6.5E+3	3.3E+3	5.1E-3	1.0E+1	1.8E+2	6.7E-1	8.1E-1	1.6E+0	1.8E+0	10.0	3.0E+3	
MGZEJ	2	10	µg/L	0.45	28	6.7E+2	3.6E+2	5.4E+3	2.8E+1	8.9E+2	1.3E+4	1.5E+4	6.7E+3	3.7E+3	1.3E-2	7.8E+0	1.8E+2	4.3E+0	5.8E-2	9.3E-2	2.4E-1	10.0	3.0E+3	
MGZB	1	10	µg/L	0.02	3	1.8E+2	3.5E+4	2.4E+7	2.0E+7	7.3E+3	1.0E+4	1.7E+8	1.7E+7	7.1E+3	1.0E+0	7.4E+1	8.9E+1	7.8E+1	3.8E+1	6.7E+1	9.7E+1	5.9		
MGZB	1	10	µg/L	0.45	3	1.8E+2	8.8E+3	3.3E+7	2.8E+7	1.3E+4	1.0E+4	1.7E+8	2.2E+7	4.1E+3	2.7E+0	6.2E+1	1.1E+2	7.2E+1	6.5E+1	8.8E+1	9.7E+1	5.9		
MGZB	2	10	µg/L	0.02	3	2.0E+2	1.3E+4	3.2E+7	2.7E+7	3.9E+3	7.4E+3	1.7E+8	2.2E+7	5.2E+3	1.4E+0	6.0E+1	1.4E+2	1.0E+2	6.0E+1	6.9E+1	8.5E+1	5.9		
MGZB	2	10	µg/L	0.45	3	2.0E+2	6.0E+3	3.4E+7	2.9E+7	4.5E+3	7.8E+3	1.7E+8	2.3E+7	2.0E+3	7.8E-1	5.9E+1	1.1E+2	4.6E+1	4.8E+1	4.4E+1	1.5E+1	5.9		
MGZB	1	10	µg/L	0.02	28	4.1E+2	4.5E+3	3.4E+7	2.7E+7	2.9E+2	1.4E+4	1.7E+8	2.2E+7	7.4E+3	1.9E+0	9.1E+1	2.1E+2	1.2E+2	7.5E+1	1.4E+2	8.9E+1	5.5		
MGZB	1	10	µg/L	0.45	28	4.3E+2	2.6E+3	3.1E+7	2.6E+7	2.3E+3	1.4E+4	1.7E+8	1.9E+7	6.6E+3	2.0E+0	9.6E+1	1.6E+2	1.2E+2	8.1E+1	1.2E+2	1.1E+2	5.5		
MGZB	2	10	µg/L	0.02	28	5.7E+2	3.4E+3	3.3E+7	2.8E+7	1.6E+2	1.2E+4	1.7E+8	2.2E+7	5.9E+3	3.6E+0	9.1E+1	3.5E+2	1.0E+2	6.6E+1	8.7E+1	1.6E+2	5.5		
MGZB	2	10	µg/L	0.45	28	5.6E+2	2.1E+3	3.3E+7	2.7E+7	1.9E+2	1.7E+4	1.7E+8	2.1E+7	7.0E+3	4.3E+0	9.6E+1	3.4E+2	1.0E+2	9.4E+1	1.9E+2	1.3E+2	5.5		

TABLE B1. (contd.)

Leachate results from MCC-1 tests with the H059-1,2 samples in deionized water, EJ-13 leachant, and Mg-Na-K-Cl brine (contd.).																						
Test	#	S/V	Meas.	Filter	days	Li	B	Na	Mg	Al	Si	Cl	K	Ca	Y	Sr	Cs	Ba	La	Ce	Nd	
MGZD	1	10	ER%	0.02	3	0.2844	0.0351	0.6138		0.2347	0.1833	0.6026	0.2567	0.4722		0.6235	0.3001	0.1133	0.0003	0.0001	0.0001	
MGZD	1	10	ER%	0.45	3	0.3248	0.0395	0.8497		0.3007	0.2097	0.6026	0.3005	0.4833	0.0078	0.7604	0.3234	0.3869	0.0015	0.0006	0.0004	
MGZD	2	10	ER%	0.02	3	0.876	0.1057	1.2318		0.3642	0.3117	1.4406	0.9516	0.5332	0.0065	0.1876	0.9122	0.0557	0.0014	0.0007	0.0005	
MGZD	2	10	ER%	0.45	3	0.9178	0.2787	1.4437		0.4141	0.3226	1.4406	1.0511	0.5018		0.173	0.8617	0.1346	0.0015	0.0005	0.0005	
MGZD	3	10	ER%	0.02	3	0.5359	0.1524	0.7106		0.223	0.1831	0.7961	0.5667	0.3238		0.1012	0.5287	0.0333	0.0003	0.0002	1E-05	
MGZD	3	10	ER%	0.45	3	0.5397	0.0571	0.6763		0.2678	0.1914	0.7961	0.601	0.3212		0.1033	0.549	0.0569	0.0008	0.0003	0.0001	
MGZD	1	10	ER%	0.02	7	0.3373	0.0357	0.5418		0.3344	0.2243	0.7302	0.3233	0.564	0.0032	0.7526	0.2887	0.2244	0.0009	0.0002	3E-05	
MGZD	1	10	ER%	0.45	7	0.3297	0.0341	0.5242		0.3565	0.2279	0.7302	0.3191	0.5495		0.7177	0.2782	0.2868	0.0013	0.0007	0.0004	
MGZD	2	10	ER%	0.02	7	0.538	0.1513	0.4988		0.2543	0.173	0.779	0.5385	0.2749		2.3011	0.5131	0.0252	0.0011	0.0007	0.0002	
MGZD	2	10	ER%	0.45	7	0.5367	0.1563	0.5349		0.2612	0.1806	0.779	0.5122	0.2624	0.0009	0.0664	0.4859	0.0399	0.0022	0.0023	0.0019	
MGZD	1	10	ER%	0.02	28	0.8576	0.2557	1.175		0.4661	0.4039	2.8386	0.5505	0.9574		1.0208	0.6429	0.2367	0.0004	0.0003	0.0002	
MGZD	1	10	ER%	0.45	28	0.597	0.1835	0.8348		0.3669	0.2921	2.8386	0.4162	0.6667		0.7682	0.4365	0.2583	0.0006	0.0006	0.0005	
MGZD	2	10	ER%	0.02	28	1.4639	0.48	1.3721		0.6138	0.4529	3.2787	1.3363	0.5347		0.0914	2.6067	0.0062	0.0003	0.0003	0.0004	
MGZD	2	10	ER%	0.45	28	1.4707	0.4725	1.3559		0.6777	0.481	3.2787	1.2955	0.5802	0.0034	0.125	2.7278	0.0711	0.0099	0.0116	0.0096	
MGZD	1	10	ER%	0.02	91																	
MGZD	1	10	ER%	0.45	91																	
MGZD	2	10	ER%	0.02	91																	
MGZD	2	10	ER%	0.45	91																	
MGZEJ	1	10	ER%	0.02	3	0.2448	0.0448	-13.58		0.1046	-2.225	0.792	-0.018	-0.316	-3.75	-0.078	0.1982	-0.012	-5E-04	-9E-04	0.0004	
MGZEJ	1	10	ER%	0.45	3	0.2284	0.0342	-13.6		0.002	-2.232	0.792	-0.06	-0.055	-3.758	0.2763	0.1981	0.1889	0.0027	-2E-04	0.0013	
MGZEJ	2	10	ER%	0.02	3	0.1442	0.0126	-5.12		0.0262	-0.844	0.4551	0.0743	-0.064	-1.412	-0.191	0.235	-0.002	-3E-04	-5E-04	-5E-06	
MGZEJ	2	10	ER%	0.45	3	0.1411	0.0128	-5.096		9E-05	-0.833	0.4551	0.1435	-0.031	-1.413	-0.155	0.22	0.0303	0.0029	0.0052	0.0007	
MGZEJ	1	10	ER%	0.02	28	0.5788	0.0532	-6.93		0.0988	-1.134	0.6777	-0.264	-0.245	-1.918	-0.139	0.4832	-0.002	0.0006	4E-05	0.0004	
MGZEJ	1	10	ER%	0.45	28	0.6111	0.0547	-6.89		0.0186	-1.115	0.6777	-0.18	-0.065	-1.918	0.0445	0.4761	0.1428	0.001	0.0002	0.0008	
MGZEJ	2	10	ER%	0.02	28	0.4423	0.0576	-4.28		0.0508	-0.699	0.5661	0.2305	-0.074	-1.187	-0.232	0.4318	0.0002	0.0101	0.0061	0.0036	
MGZEJ	2	10	ER%	0.45	28	0.4321	0.0442	-4.269		0.0047	-0.694	0.5661	0.2543	-0.031	-1.186	-0.276	0.4293	0.0245	0.0002	-1E-04	0.0005	
MGZB	1	10	ER%	0.02	3	0.1707	11.129			0.6104	0.3084			0.5035	0.16	0.611	0.3504	0.5274	0.4754	0.464	0.3255	
MGZB	1	10	ER%	0.45	3	0.1713	2.4486			1.087	0.2989			-0.075	0.4263	0.2778	0.4446	0.4624	1.0957	0.6126	0.3269	
MGZB	2	10	ER%	0.02	3	0.154	3.0853			0.2537	0.1588			0.1112	0.1772	0.1635	0.4666	0.6314	0.7718	0.3783	0.2268	
MGZB	2	10	ER%	0.45	3	0.1541	1.2186			0.2943	0.1695			-0.38	0.0999	0.133	0.3591	0.1262	0.5563	0.2358	0.03	
MGZB	1	10	ER%	0.02	28	0.6858	1.6286			0.0247	0.6593			0.8727	0.4818	1.792	1.3738	1.5532	2.066	1.4905	0.4639	
MGZB	1	10	ER%	0.45	28	0.7174	0.6944			0.2924	0.7079			0.6246	0.5075	2.0238	1.0385	1.5495	2.275	1.3448	0.6007	
MGZB	2	10	ER%	0.02	28	0.4871	0.5506			0.0041	0.2934			0.2074	0.4575	0.9054	1.158	0.6441	0.8771	0.4826	0.4244	
MGZB	2	10	ER%	0.45	28	0.4834	0.2189			0.0063	0.4263			0.3814	0.5489	1.0215	1.1127	0.6194	1.3887	1.0567	0.3572	

TABLE B1. (contd.)

Leachate results from MCC-1 tests with the H059-1,2 samples in deionized water, EJ-13 leachant, and Mg-Na-K-Cl brine (contd.).																							
Test	#	S/V	Meas.	Filter	days	Li	B	Na	Mg	Al	Si	Cl	K	Ca	Y	Sr	Cs	Ba	La	Ce	Nd		
MGZD	1	10	NLi	0.02	3	3.6817	0.4546	7.9464		3.0388	2.3725	7.8013	3.3236	6.1128		8.0711	3.8844	1.4668	0.0034	0.0015	0.0017		
MGZD	1	10	NLi	0.45	3	4.2048	0.5118	11		3.8928	2.7148	7.8013	3.8896	6.2569	0.1015	9.8436	4.186	5.0085	0.0196	0.0074	0.0057		
MGZD	2	10	NLi	0.02	3	5.1231	0.6182	7.2038		2.1299	1.8229	8.4254	5.5653	3.1186	0.0383	1.097	5.3351	0.326	0.0085	0.0044	0.003		
MGZD	2	10	NLi	0.45	3	5.3677	1.6297	8.4431		2.4218	1.8868	8.4254	6.1472	2.9345		1.0118	5.0394	0.787	0.0085	0.0031	0.003		
MGZD	3	10	NLi	0.02	3	5.2515	1.4938	6.963		2.1854	1.7939	7.8013	5.5535	3.1731		0.9912	5.1806	0.3259	0.0031	0.002	0.0001		
MGZD	3	10	NLi	0.45	3	5.288	0.56	6.6266		2.6239	1.8753	7.8013	5.8894	3.1474		1.0125	5.3796	0.5571	0.0076	0.0032	0.001		
MGZD	1	10	NLi	0.02	7	4.2517	0.4497	6.8301		4.2153	2.8273	9.2055	4.076	7.1105	0.0402	9.4874	3.6401	2.829	0.0113	0.002	0.0004		
MGZD	1	10	NLi	0.45	7	4.1564	0.4299	6.6089		4.4937	2.8277	9.2055	4.023	6.9277		9.0477	3.5076	3.6156	0.0164	0.0087	0.0055		
MGZD	2	10	NLi	0.02	7	8.1897	2.3028	7.5926		3.8715	2.6328	11.858	8.1965	4.185		35.027	7.811	0.384	0.0169	0.0105	0.0032		
MGZD	2	10	NLi	0.45	7	8.1699	2.379	8.142		3.9752	2.749	11.858	7.7966	3.9948	0.0141	1.0105	7.397	0.6079	0.0327	0.035	0.0293		
MGZD	1	10	NLi	0.02	28	8.4853	2.5303	11.625		4.6117	3.9965	28.085	5.4463	9.472		10.099	6.3603	2.3423	0.0037	0.0032	0.0019		
MGZD	1	10	NLi	0.45	28	5.9061	1.815	8.259		3.6301	2.8896	28.085	4.1179	6.5966		7.6004	4.3184	2.5558	0.0059	0.0063	0.0049		
MGZD	2	10	NLi	0.02	28	13.932	4.5685	13.059		5.8414	4.3101	31.205	12.718	5.0891		0.8701	24.809	0.0593	0.0031	0.0031	0.0037		
MGZD	2	10	NLi	0.45	28	13.997	4.4973	12.904		6.4499	4.5782	31.205	12.33	5.5222	0.0324	1.1897	25.962	0.6767	0.0942	0.1107	0.0912		
MGZD	1	10	NLi	0.02	91																		
MGZD	1	10	NLi	0.45	91																		
MGZD	2	10	NLi	0.02	91																		
MGZD	2	10	NLi	0.45	91																		
MGZEJ	1	10	NLi	0.02	3	2.068	0.8369	11.238		1.5388	5.5729	16.695	17.271	7.3515	0.0807	8.2352	1.4133	0.0131	0.0085	0.0047	0.0032		
MGZEJ	1	10	NLi	0.45	3	1.9584	0.7654	11.166		0.8515	5.5263	16.695	16.99	9.1025	0.0263	10.61	1.4124	1.3564	0.0301	0.0096	0.009		
MGZEJ	2	10	NLi	0.02	3	2.9988	0.7607	10.939		1.3048	5.4291	19.503	18.715	8.3308	0.0201	5.3479	4.2746	0.0536	0.0059	0.0018	0.0004		
MGZEJ	2	10	NLi	0.45	3	2.9434	0.7654	11.379		0.8395	5.6326	19.503	19.949	8.9142	0.0121	5.9927	4.0085	0.6319	0.0629	0.1039	0.0134		
MGZEJ	1	10	NLi	0.02	28	8.0241	1.2349	11.284		2.1352	5.5916	20.283	13.92	6.2496	0.0243	6.9363	6.4266	0.0701	0.0201	0.0115	0.0063		
MGZEJ	1	10	NLi	0.45	28	8.4473	1.2538	11.817		1.0815	5.8449	20.283	15.025	8.6169	0.0365	9.3429	6.3331	1.9647	0.0256	0.0141	0.011		
MGZEJ	2	10	NLi	0.02	28	9.816	1.7589	11.389		1.9165	5.6367	23.404	22.282	7.8969	0.0101	3.831	9.2482	0.0951	0.2266	0.1394	0.0768		
MGZEJ	2	10	NLi	0.45	28	9.5988	1.4748	11.63		0.9375	5.7465	23.404	22.788	8.8024	0.0263	2.9095	9.1957	0.6116	0.0162	0.0082	0.0105		
MGZB	1	10	NLi	0.02	3	2.5941	143.7			7.7002	4.6801			17.054	1.9913	27.519	4.6475	11.031	10.598	5.9234	4.217		
MGZB	1	10	NLi	0.45	3	2.6017	35.66			13.632	4.5622			9.8548	5.3056	23.371	5.8198	10.222	18.318	7.7729	4.2338		
MGZB	2	10	NLi	0.02	3	2.8823	53.515			4.0766	3.3299			12.53	2.7758	22.475	7.5963	14.357	16.77	6.0741	3.7178		
MGZB	2	10	NLi	0.45	3	2.8839	24.273			4.7139	3.497			4.8411	1.5657	21.997	5.9113	6.4428	13.395	3.8416	0.6353		
MGZB	1	10	NLi	0.02	28	5.9265	18.141			0.2998	6.0878			17.731	3.8331	34.172	11.217	16.824	21.118	12.007	3.8563		
MGZB	1	10	NLi	0.45	28	6.1781	10.708			2.4294	6.474			15.757	4.0379	36.016	8.5492	16.795	22.78	10.848	4.9449		
MGZB	2	10	NLi	0.02	28	8.1059	13.813			0.1665	5.4416			14.039	7.1718	34.105	18.438	14.563	18.429	7.7128	6.8173		
MGZB	2	10	NLi	0.45	28	8.0477	8.614			0.2011	7.5238			16.766	8.6045	35.925	17.727	14.175	26.447	16.712	5.7639		

TABLE B1. (contd.)

Leachate results from PCT-Bs with salt-loaded zeolite 4A samples in deionized water.																																		
Test	#	S/V	Meas.	Filter	days	Li	B	Na	Mg	Al	Si	Cl	K	Ca	Y	Sr	Cs	Ba	La	Ce	Nd	pH	C(inor)	O										
wt% Salt Loaded Zeolite 4A						1.36	0.00	12.93	0.00	15.00	15.62	12.92	5.49	0.00	0.01	0.06	0.63	0.21	0.13	0.24	0.39													
PZD	1	2K	µg/L	0.02	0	6.6E+3	1.1E+1	1.5E+4	2.8E+1	7.8E+3	3.9E+3	5.5E+4	2.9E+3	1.0E+0		3.4E-1	6.7E+0	6.0E-1	2.4E-1	8.4E-1	1.6E+0	7.8	4.3E+3											
PZD	1	2K	µg/L	0.01	0	7.9E+3	3.7E+1	1.7E+4	1.6E+1	7.3E+3	4.3E+3	5.5E+4	2.8E+3	2.3E+2		5.7E-1	5.5E+0	5.5E-2	4.4E-1	8.5E+0	1.0E+0	7.8	4.3E+3											
PZD	1	2K	µg/L	0.02	0.04	1.6E+4	1.0E+1	3.3E+4	6.4E+0	1.6E+4	9.9E+3	1.0E+5	9.4E+3	1.1E+2		1.2E-1	3.5E+1	3.2E-1	2.4E+0	1.4E+1	2.1E+1	8.4	7.6E+3											
PZD	1	2K	µg/L	0.01	0.04	1.7E+4	1.0E+1	4.1E+4	6.4E+0	1.5E+4	1.0E+4	1.0E+5	9.4E+3	1.1E+2		3.4E-1	2.4E+1	1.2E-1	5.3E-2	2.5E-2	4.7E-1	8.4	7.6E+3											
PZD	1	2K	µg/L	0.02	3	3.4E+4	4.7E+0	9.9E+4	5.7E-1	3.3E+4	2.7E+4	2.4E+5	4.8E+4	3.6E+0		1.5E-1	2.1E+3	3.0E-2	5.1E+0	2.9E+1	4.7E+1	9.6	1.2E+4											
PZD	2	2K	µg/L	0.02	3	3.3E+4	1.1E+1	9.6E+4	5.7E-1	3.5E+4	2.9E+4	2.4E+5	4.6E+4	2.6E+1		6.0E-2	2.0E+3	2.8E-2	4.5E+0	2.5E+1	4.0E+1	9.5	1.2E+4											
PZD	1	2K	µg/L	0.02	7	2.8E+4	1.7E+2	1.3E+5	1.6E+1	3.6E+4	3.1E+4	2.2E+5	6.3E+4	3.6E+2		8.4E-1	2.7E+3		6.3E+0	3.3E+1	6.4E+1	10.3	2.6E+3											
PZD	2	2K	µg/L	0.02	7	2.6E+4	6.4E+1	1.2E+5	1.3E+2	3.6E+4	3.1E+4	2.1E+5	5.8E+4	7.1E+2		8.3E-1	2.4E+3		4.8E+0	2.1E+1	4.5E+1	10.2	2.8E+3											
PZD	3	2K	µg/L	0.02	28	2.9E+4	3.6E+1	1.3E+5	3.1E+1	3.6E+4	2.9E+4	2.8E+5	6.6E+4	2.2E+2		7.5E-1	2.7E+3	1.6E-1	1.0E+1	2.9E+1	8.4E+1	8.6	9.5E+3											
PZD	4	2K	µg/L	0.02	28	2.8E+4	2.7E+2	1.3E+5	3.0E+1	3.6E+4	2.9E+4	2.9E+5	6.3E+4	2.4E+2		7.5E-1	2.7E+3	1.5E+0	6.9E+0	3.0E+1	5.7E+1	8.6	8.0E+3											
PZD	1	2K	µg/L	0.02	91	2.8E+4	1.9E+2	1.3E+5	8.8E+1	3.2E+4	2.7E+4	3.4E+5	6.2E+4	1.8E+2	2.7E+0	1.3E-1	3.0E+3	2.6E-1	3.0E+0	1.5E+1	1.6E+1	10.0	3.6E+3											
PZD	2	2K	µg/L	0.02	91	2.7E+4	1.3E+2	1.3E+5	2.9E+2	3.2E+4	2.8E+4	3.2E+5	5.8E+4	1.8E+2	2.5E+0	4.5E-1	2.7E+3	3.3E-1	4.1E+0	1.6E+1	2.2E+1	10.0	1.8E+3											
PZD	1	2K	µg/L	0.02	365																													
PZD	2	2K	µg/L	0.02	365																													
PZDh	1	20K	µg/L	0.02	3	2.6E+5	9.3E+0	7.4E+5	7.4E+0	3.2E+4	1.1E+4	2.9E+6	3.6E+5	1.2E+2		2.1E+0	1.2E+4	1.7E-1	1.9E-1	1.1E+0	4.5E+0	10.6	1.0E+4											
PZDh	1	20K	µg/L	0.01	3	2.6E+5	4.1E+0	7.3E+5	2.8E+1	3.3E+4	1.2E+4	2.9E+6	3.2E+5	4.7E+2		2.7E+0	1.2E+4	1.9E+0	1.6E-1	2.3E-1	4.0E+0	10.6	1.0E+4											
PZDh	1	20K	µg/L	0.02	7	2.9E+5	9.7E+2	1.2E+6	2.7E+1	4.7E+4	1.4E+4	3.3E+6	4.9E+5	6.2E+1		2.9E+0	1.7E+4	9.6E+0	2.2E-1	2.2E+0	1.7E+0	10.9	4.7E+3											
PZDh	2	20K	µg/L	0.02	7	2.9E+5	6.6E+2	1.2E+6	1.1E+2	5.0E+4	1.4E+4	3.3E+6	5.0E+5	1.2E+2		2.9E+0	1.7E+4	9.6E+0	2.2E-1	3.2E+0	5.0E+0	10.9	4.5E+3											
PZDh	1	20K	µg/L	0.02	28	2.9E+5	4.2E+2	1.3E+6	7.5E+1	5.0E+4	1.4E+4	3.3E+6	4.9E+5	7.5E+1		2.9E+0	1.8E+4	4.6E+0	1.0E+0	1.0E+0	1.6E+0	10.6	8.4E+3											
PZDh	2	20K	µg/L	0.02	28	2.9E+5	2.6E+2	1.2E+6	2.7E+1	5.3E+4	1.5E+4	3.0E+6	5.2E+5	7.3E+1		4.7E+0	1.8E+4	4.3E+0	1.5E-1	1.6E+0	3.2E+0	10.6	8.1E+3											
PZDh	1	20K	µg/L	0.02	91	3.3E+5	5.1E+2	9.9E+5	2.7E+1	5.7E+4	3.4E+4	3.4E+6	4.8E+5		8.8E-2	1.8E+0	2.1E+4	6.2E+0	1.1E+0	2.1E+0	1.7E+0	10.1	2.0E+4											
PZDh	2	20K	µg/L	0.02	91	3.3E+5	3.5E+2	9.9E+5	3.3E+1	5.3E+4	2.5E+4	3.4E+6	4.8E+5		8.8E-2	7.6E-1	2.1E+4	5.3E+0	1.5E+0	1.4E+0	1.7E+0	10.2	1.9E+4											
PZDh	2	20K	µg/L	0.45	91	3.2E+5	2.0E+2	9.9E+5	2.4E+2	5.3E+4	3.2E+4	3.4E+6	4.7E+5		5.8E-1	3.8E+0	2.1E+4	1.2E+1	3.3E+0	5.4E+0	9.1E+0	10.1	2.0E+4											

TABLE B1. (contd.)

Leachate results from PCT-Bs with salt-loaded zeolite 4A samples in deionized water (contd.).																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																																									</
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TABLE B1. (contd.)

Leachate results from PCT-Bs with salt-loaded zeolite 4A samples in deionized water (contd.).																						
Test	#	S/V	Meas.	Filter	days	Li	B	Na	Mg	Al	Si	Cl	K	Ca	Y	Sr	Cs	Ba	La	Ce	Nd	
PZD	1	2K	NLi	0.02	0	0.2414		0.0571		0.026	0.0126	0.2128	0.0267			0.0003	0.0005	0.0001	9E-05	0.0002	0.0002	
PZD	1	2K	NLi	0.01	0	0.2894		0.0676		0.0242	0.0138	0.2128	0.0255			0.0005	0.0004	1E-05	0.0002	0.0018	0.0001	
PZD	1	2K	NLi	0.02	0.04	0.5844		0.1264		0.054	0.0315	0.4025	0.0858			0.0001	0.0028	8E-05	0.0009	0.003	0.0027	
PZD	1	2K	NLi	0.01	0.04	0.6239		0.1577		0.0507	0.0328	0.4025	0.0856			0.0003	0.0019	3E-05	2E-05	5E-06	6E-05	
PZD	1	2K	NLi	0.02	3	1.2667		0.3829		0.1101	0.0867	0.9288	0.4339			0.0001	0.1642	7E-06	0.002	0.0061	0.006	
PZD	2	2K	NLi	0.02	3	1.2182		0.3702		0.1161	0.0934	0.9288	0.4206			5E-05	0.1554	7E-06	0.0017	0.0052	0.0051	
PZD	1	2K	NLi	0.02	7	1.0404		0.515		0.1194	0.0984	0.8553	0.573			0.0007	0.217		0.0024	0.0069	0.0082	
PZD	2	2K	NLi	0.02	7	0.9632		0.4809		0.1192	0.0978	0.8204	0.5243			0.0007	0.1886		0.0019	0.0044	0.0058	
PZD	1	2K	NLi	0.02	28	1.0695		0.5217		0.12	0.0941	1.0681	0.5977			0.0006	0.2144	4E-05	0.0039	0.006	0.0108	
PZD	2	2K	NLi	0.02	28	1.0469		0.5072		0.1184	0.0923	1.1184	0.5719			0.0006	0.217	0.0004	0.0026	0.0063	0.0073	
PZD	1	2K	NLi	0.02	91	1.0229		0.5189		0.1063	0.0862	1.3274	0.5647		0.0133	0.0001	0.2344	6E-05	0.0011	0.003	0.0021	
PZD	2	2K	NLi	0.02	91	0.9753		0.4872		0.1055	0.0893	1.2384	0.5285		0.0127	0.0004	0.2106	8E-05	0.0016	0.0032	0.0028	
PZDh	1	20K	NLi	0.02	3	0.9559		0.2872		0.0108	0.0037	1.1029	0.3298			0.0002	0.0981	4E-06	7E-06	2E-05	6E-05	
PZDh	1	20K	NLi	0.01	3	0.9513		0.2823		0.0109	0.0037	1.1029	0.2897			0.0002	0.098	5E-05	6E-06	5E-06	5E-05	
PZDh	1	20K	NLi	0.02	7	1.0655		0.4528		0.0157	0.0045	1.2577	0.4477			0.0002	0.1339	0.0002	8E-06	5E-05	2E-05	
PZDh	2	20K	NLi	0.02	7	1.0589		0.4615		0.0168	0.0046	1.2577	0.4556			0.0002	0.1327	0.0002	8E-06	7E-05	6E-05	
PZDh	1	20K	NLi	0.02	28	1.0755		0.4854		0.0166	0.0043	1.2926	0.4504			0.0002	0.1403	0.0001	4E-05	2E-05	2E-05	
PZDh	2	20K	NLi	0.02	28	1.0777		0.4734		0.0178	0.0047	1.1533	0.4749			0.0004	0.1456	0.0001	6E-06	3E-05	4E-05	
PZDh	1	20K	NLi	0.02	91	1.2192		0.3831		0.0191	0.0109	1.3158	0.4416		4E-05	0.0002	0.1697	0.0001	4E-05	4E-05	2E-05	
PZDh	2	20K	NLi	0.02	91	1.2124		0.3809		0.0176	0.008	1.3158	0.4379		4E-05	6E-05	0.1701	0.0001	6E-05	3E-05	2E-05	
PZDML93-7			NLi		7	0.13		0.049		0.01	0.009		0.048				0.0054					
PZDML93-28			NLi		28	0.03		0.0067		0.0013	0.0088		0.008				0.0018					

TABLE B1. (contd.)

Leachate results from PCT-Bs with salt-loaded zeolite 4A samples in phosphate buffers (contd.).																								
Test	#	S/V	Meas.	Filter	days	Li	B	Na	Mg	Al	Si	Cl	K	Ca	Y	Sr	Cs	Ba	La	Ce	Nd	pH	C(inor)	O
Zeolite						1.47	0.00	12.93	0.00	15.00	15.62	12.72	5.49	0.00	0.01	0.06	0.63	0.21	0.13	0.24	0.39			
PO43			µg/L			4.9E+1	1.4E+2	8.7E+5	5.8E+0	0.0E+0	0.0E+0	43000	0.0E+0	0.0E+0		2.2E-1	1.9E+0	3.5E-1	1.1E-1	9.0E-2	3.4E-2	3.0	4.6E+1	
PZ3P	1	2K	µg/L	0.02	3	3.9E+4	8.2E+2	1.2E+6	3.0E+1	6.8E+2	1.3E+5	256000	1.4E+5	1.1E+2		4.0E+0	4.8E+3	2.6E-1	3.3E-2	1.1E-1	1.2E-1	6.5	1.7E+2	
PZ3P	1	2K	µg/L	0.45	3	3.8E+4	8.2E+2	1.2E+6	3.9E+1	4.0E+4	1.7E+5	256000	1.3E+5	1.5E+2		1.2E+2	5.0E+3	4.9E+2	2.1E+2	8.7E+2	1.5E+3	6.5	1.7E+2	
PO47			µg/L			4.9E+1	1.2E+2	1.4E+6	1.6E+1	4.8E+0	0.0E+0	260	0.0E+0	0.0E+0		5.2E-1	2.4E+0	6.7E-1	1.5E-1	4.6E-2	3.4E-2	7.0	8.9E+2	
PZ7P	1	2K	µg/L	0.02	3	3.4E+4	4.6E+2	1.5E+6	2.1E+1	6.1E+3	5.9E+4	222000	1.2E+5	1.1E+3		2.0E+0	5.2E+3	2.1E+0	2.0E-1	3.8E-1	2.5E-1	7.3	1.2E+3	
PZ7P	1	2K	µg/L	0.45	3	3.4E+4	3.0E+2	1.5E+6	1.2E+1	7.9E+3	5.9E+4	222000	1.2E+5	3.9E+2		1.2E+1	5.2E+3	5.5E+1	4.5E+0	2.1E+1	4.0E+1	7.3	1.2E+3	
PO95			µg/L			2.4E+1	8.5E+1	1.7E+6	3.8E+1	0.0E+0	0.0E+0	0	0.0E+0	0.0E+0		3.8E-1	4.1E-1	1.2E+0	7.5E-2	1.6E-1	5.6E+0	9.5	7.7E+3	
PZ95P	1	2K	µg/L	0.02	3	3.6E+4	2.1E+2	1.8E+6	1.2E+1	5.5E+3	5.4E+3	219000	1.3E+5	1.0E+3		4.1E-1	4.8E+3	4.2E-1	1.1E-1	1.3E-1	1.1E-1	10.0	7.0E+3	
PZ95P	1	2K	µg/L	0.45	3	3.5E+4	1.8E+2	1.8E+6	1.6E+1	1.2E+4	5.3E+3	219000	1.3E+5	6.0E+2		3.5E+0	4.6E+3	5.0E+0	2.2E+0	8.6E+0	1.6E+1	10.0	7.0E+3	
PZ3P	1	2K	ER%	0.02	3	99.592		97.491		0.1695	32.378	62.795	92.533			0.2389	28.466	-0.001	-0.002	0.0004	0.0008			
PZ3P	1	2K	ER%	0.45	3	96.629		89.883		10.114	39.79	62.795	91.371			7.6618	29.687	8.7838	6.0277	13.526	14.589			
PZ7P	1	2K	ER%	0.02	3	87.436		19.23		1.5157	14.074	65.371	83.614			0.0927	31.129	0.0254	0.0016	0.0053	0.0021			
PZ7P	1	2K	ER%	0.45	3	86.696		29.02		1.984	14.055	65.371	83.783			0.6926	30.884	0.9787	0.1249	0.324	0.3828			
PZ95P	1	2K	ER%	0.02	3	91.005		22.207		1.3781	1.2965	64.564	87.227			0.0018	28.313	-0.014	0.0009	-4E-04	-0.053			
PZ95P	1	2K	ER%	0.45	3	90.327		33.024		2.9339	1.2667	64.564	86.531			0.1921	27.28	0.0677	0.0623	0.1322	0.0998			
PZ3P	1	2K	NLi	0.02	3	1.3279		1.2999		0.0023	0.4317	0.8373	1.2338			0.0032	0.3795	-2E-05	-3E-05	5E-06	1E-05			
PZ3P	1	2K	NLi	0.45	3	1.2884		1.1984		0.1348	0.5305	0.8373	1.2183			0.1022	0.3958	0.1171	0.0804	0.1803	0.1945			
PZ7P	1	2K	NLi	0.02	3	1.1658		0.2564		0.0202	0.1877	0.8716	1.1149			0.0012	0.415	0.0003	2E-05	7E-05	3E-05			
PZ7P	1	2K	NLi	0.45	3	1.1559		0.3869		0.0265	0.1874	0.8716	1.1171			0.0092	0.4118	0.013	0.0017	0.0043	0.0051			
PZ95P	1	2K	NLi	0.02	3	1.2134		0.2961		0.0184	0.0173	0.8608	1.163			2E-05	0.3775	-2E-04	1E-05	-6E-06	-7E-04			
PZ95P	1	2K	NLi	0.45	3	1.2044		0.4403		0.0391	0.0169	0.8608	1.1537			0.0026	0.3637	0.0009	0.0008	0.0018	0.0013			

TABLE B1. (contd.)

Leachate results from PCT-Bs with salt-loaded zeolite 4A samples in Mg-Na-K-Cl Brine and Ca-Na-K-Cl Brine.																								
Test	#	S/V	Meas.	Filter	days	Li	B	Na	Mg	Al	Si	Cl	K	Ca	Y	Sr	Cs	Ba	La	Ce	Nd	pH	C(inor)	O
Mg Brine			μg/L			3.3E+1	1.3E+3	3.5E+7	2.9E+7	9.8E+1	1.9E+3	1.3E+8	2.3E+7	4.5E+3	0.0E+0	5.3E+1	5.5E+0	3.2E+1	1.7E+1	1.7E+0	3.8E+0	6.5	9.3E+2	
PZAB	1	2K	μg/L	0.02	7	3.5E+4	3.7E+2	3.3E+7	2.8E+7	3.3E+2	2.5E+4	1.3E+8	2.3E+7	6.6E+3		1.4E+3	9.7E+3	3.4E+3	1.4E+2	6.0E+2	1.7E+3	5.6	1.5E+2	
PZAB	2	2K	μg/L	0.02	7	3.5E+4	9.4E+1	3.5E+7	2.9E+7	2.6E+2	2.2E+4	1.5E+8	2.4E+7	5.2E+3		1.4E+3	9.2E+3	3.2E+3	1.5E+2	5.6E+2	1.9E+3	5.6	3.8E+2	
PZAB	1	2K	μg/L	0.02	28	3.9E+4	1.1E+2	3.4E+7	2.8E+7	2.4E+2	2.5E+4	1.3E+8	2.3E+7	5.3E+3		1.5E+3	1.0E+4	3.8E+3	4.0E+2	1.3E+3	2.9E+3	5.4	5.6E+2	
PZAB	2	2K	μg/L	0.02	28	3.9E+4	1.1E+2	3.4E+7	2.8E+7	1.9E+2	2.1E+4	1.5E+8	2.3E+7	5.0E+3		1.6E+3	1.1E+4	4.1E+3	4.0E+2	1.4E+3	3.4E+3	5.4	3.9E+2	
PZAB	1	2K	μg/L	0.02	91	4.2E+4	3.6E+3	3.3E+7	2.2E+7	2.3E+2	2.3E+4	1.9E+8	2.2E+7	1.6E+4	1.2E+2	1.8E+3	1.3E+4	4.8E+3	4.9E+2	1.3E+3	2.0E+3	5.6	4.1E+1	
PZAB	2	2K	μg/L	0.02	91	3.9E+4	1.5E+3	3.5E+7	2.8E+7	3.1E+2	1.9E+4	1.8E+8	2.2E+7	2.5E+3	8.6E+1	1.7E+3	1.4E+4	4.9E+3	4.8E+2	1.3E+3	1.3E+3	5.6	6.4E+1	
Ca Brine			μg/L			2.7E+1	1.0E+3	3.4E+7	2.8E+3	5.4E+0	4.8E+3	1.3E+8	2.3E+7	4.4E+7		2.9E+4	4.3E+0	1.3E+3	4.6E-1	4.3E+0	0.0E+0	6.5	1.2E+3	
PZCB	1	2K	μg/L	0.02	7	3.8E+4	3.7E+2	3.7E+7	2.8E+3	3.3E+3	6.5E+3	1.2E+8	2.5E+7	4.9E+7		3.1E+4	1.1E+4	5.4E+3	5.1E+1	3.7E+0	2.4E+1	6.5	2.2E+3	
PZCB	2	2K	μg/L	0.02	7	3.8E+4	1.9E+2	3.5E+7	2.9E+3	3.5E+3	1.0E+4	1.2E+8	2.3E+7	4.7E+7		3.2E+4	1.1E+4	5.7E+3	4.2E+1	3.0E+0	1.3E+1	6.5	2.1E+3	
PZCB	1	2K	μg/L	0.02	28	3.8E+4	1.1E+2	3.4E+7	2.7E+3	2.2E+3	9.2E+3	1.4E+8	2.3E+7	4.5E+7		3.0E+4	1.1E+4	5.4E+3	7.0E+1	3.9E+0	8.4E+1	5.8	1.8E+3	
PZCB	2	2K	μg/L	0.02	28	3.8E+4	6.4E+1	3.4E+7	2.7E+3	2.1E+3	5.1E+3	1.4E+8	2.3E+7	4.7E+7		3.0E+4	1.1E+4	5.2E+3	8.4E+1	2.5E+1	1.2E+2	5.8	1.0E+3	
PZCB	1	2K	μg/L	0.02	91	4.3E+4	9.7E+2	3.4E+7	1.2E+4	9.8E+2	9.4E+3	1.8E+8	2.2E+7	4.9E+7	1.1E+1	3.4E+4	1.3E+4	7.0E+3	1.6E+2	1.4E+2	6.4E+2	5.8	4.4E+2	
PZCB	2	2K	μg/L	0.02	91	3.9E+4	6.9E+2	3.5E+7	3.7E+3	9.4E+2	1.1E+4	1.8E+8	2.2E+7	4.9E+7	3.2E+1	3.4E+4	1.2E+4	6.6E+3	2.3E+2	1.3E+2	3.5E+2	5.8	1.8E+2	
Ca Brine	2		μg/L				3.7E+2	3.8E+7	1.8E+3	2.6E+2	3.5E+2		2.4E+7	2.9E+7	6.5E+1	1.5E+3	3.4E+1	4.0E+1	2.9E+0	1.5E+2	2.8E+0			
PZCB	3	2K	μg/L	0.02	7	2.5E+4	4.6E+2	3.6E+7	1.2E+3	3.8E+3	2.1E+3	1.2E+8	2.3E+7	2.8E+7	1.9E+0	2.8E+3	1.2E+4	4.1E+3	3.1E+0	1.2E+1	4.5E+0			
PZCB	4	2K	μg/L	0.02	7	2.6E+4	4.1E+2	3.7E+7	3.3E+3	3.8E+3	2.2E+3	1.3E+8	2.4E+7	2.8E+7	7.7E-1	2.7E+3	1.1E+4	4.2E+3	2.6E+0	1.6E+1				
PZCB	3	2K	μg/L	0.45	7	24880	667	4E+07	34780	3968	2448	1.2E+8	2E+07	3E+07	0.731	2731	11680	4513	15.6	42.7	25.8			
PZCB	4	2K	μg/L	0.45	7	24520	431	4E+07	1490	3123	2159	1.3E+8	2E+07	3E+07	1.84	2816	11510	4111	12.9	28	39.1			

TABLE B1. (contd.)

Leachate results from PCT-Bs with salt-loaded zeolite 4A samples in Mg-Na-K-Cl Brine and Ca-Na-K-Cl Brine (contd.).																						
Test	#	S/V	Meas.	Filter	days	Li	B	Na	Mg	Al	Si	Cl	K	Ca	Y	Sr	Cs	Ba	La	Ce	Nd	
PZAB	1	2K	ER%	0.02	7	88.279				0.0591	5.5097					84.466	57.74	60.462	3.7009	9.3664	16.239	
PZAB	2	2K	ER%	0.02	7	88.589				0.0407	4.8756					81.995	54.917	56.815	3.8321	8.7047	18.511	
PZAB	1	2K	ER%	0.02	28	99.693				0.035	5.4341					90.424	61.951	67.516	11.179	19.962	28.131	
PZAB	2	2K	ER%	0.02	28	100.03				0.0233	4.5144					99.721	67.28	73.148	10.985	21.405	32.808	
PZAB	1	2K	ER%	0.02	91	106.92				0.034	5.1398				43.274	109.69	75.227	86.002	13.586	19.823	18.749	
PZAB	2	2K	ER%	0.02	91	99.766				0.0518	4.1275				32.074	102.9	81.114	87.077	13.244	19.8	12.126	
PZCB	1	2K	ER%	0.02	7	96.927				0.8277	0.4158					124.01	65.29	73.252	1.4617	-0.009	0.2269	
PZCB	2	2K	ER%	0.02	7	96.654				0.8661	1.3241					164.41	68.279	78.546	1.1993	-0.02	0.1247	
PZCB	1	2K	ER%	0.02	28	97.024				0.5404	1.0766					47.177	63.744	72.121	2.0111	-0.006	0.8074	
PZCB	2	2K	ER%	0.02	28	97.028				0.5273	0.0698					19.556	62.548	69.446	2.4207	0.3173	1.2006	
PZCB	1	2K	ER%	0.02	91	108.5				0.2434	1.1247					282.53	78.174	101.1	4.6984	2.1412	6.1956	
PZCB	2	2K	ER%	0.02	91	98.617				0.233	1.454					325.19	73.149	93.801	6.5236	1.8919	3.3456	
Test	#	S/V	Meas.	Filter	days	Li	B	Na	Mg	Al	Si	Cl	K	Ca	Y	Sr	Cs	Ba	La	Ce	Nd	
PZAB	1	2K	NLi	0.02	7	1.1771				0.0008	0.0735					1.1262	0.7699	0.8062	0.0493	0.1249	0.2165	
PZAB	2	2K	NLi	0.02	7	1.1812				0.0005	0.065					1.0933	0.7322	0.7575	0.0511	0.1161	0.2468	
PZAB	1	2K	NLi	0.02	28	1.3292				0.0005	0.0725					1.2057	0.826	0.9002	0.1491	0.2662	0.3751	
PZAB	2	2K	NLi	0.02	28	1.3338				0.0003	0.0602				0	1.3296	0.8971	0.9753	0.1465	0.2854	0.4374	
PZAB	1	2K	NLi	0.02	91	1.4256				0.0005	0.0685				0.577	1.4625	1.003	1.1467	0.1811	0.2643	0.25	
PZAB	2	2K	NLi	0.02	91	1.3302				0.0007	0.055				0.4276	1.372	1.0815	1.161	0.1766	0.264	0.1617	
PZCB	1	2K	NLi	0.02	7	1.2924				0.011	0.0055					1.6534	0.8705	0.9767	0.0195	-1E-04	0.003	
PZCB	2	2K	NLi	0.02	7	1.2887				0.0115	0.0177					2.1921	0.9104	1.0473	0.016	-3E-04	0.0017	
PZCB	1	2K	NLi	0.02	28	1.2937				0.0072	0.0144					0.629	0.8499	0.9616	0.0268	-8E-05	0.0108	
PZCB	2	2K	NLi	0.02	28	1.2937				0.007	0.0009					0.2607	0.834	0.9259	0.0323	0.0042	0.016	
PZCB	1	2K	NLi	0.02	91	1.4467				0.0032	0.015					3.767	1.0423	1.348	0.0626	0.0285	0.0826	
PZCB	2	2K	NLi	0.02	91	1.3149				0.0031	0.0194					4.3359	0.9753	1.2507	0.087	0.0252	0.0446	
											</											

TABLE B1. (contd.)

Leachate results from PCT-Bs with salt-loaded zeolite 4A samples in 311 glass leachant (GB) and EJ-13 ground water (EJ).																								
Test	#	S/V	Meas.	Filter	days	Li	B	Na	Mg	Al	Si	Cl	K	Ca	Y	Sr	Cs	Ba	La	Ce	Nd	pH	C(inor)	O
Zeolite A						1.36	0.00	12.93	0.00	15.00	15.62	12.92	5.49	0.00	0.01	0.01	0.63	0.21	0.13	0.24	0.39			
GB Leachant			µg/L	0.02		1.6E+1	3.3E+4	9.3E+4	2.1E+1	5.4E+2	5.8E+4	6.6E+3	3.3E+3	8.8E+3	0.0E+0	3.4E+2	1.7E+0	8.7E+0	1.3E-2	1.7E-2	0.0E+0	9.3	1.9E+4	
PZGB	1	2K	µg/L	0.02	7	3.1E+4	3.3E+4	1.5E+5	5.9E+0	7.4E+3	6.8E+4	2.8E+5	6.9E+4	8.8E+2	1.3E+0	6.1E-1	2.5E+3	2.6E-1	6.2E-1	4.9E+0	5.4E+0	9.8	1.7E+4	
PZGB	2	2K	µg/L	0.02	7	3.2E+4	3.2E+4	1.7E+5	1.1E+1	6.2E+3	6.2E+4	3.4E+5	6.6E+4	7.4E+2		4.4E-2	2.4E+3	3.1E-1	7.0E-1	4.9E+0	7.9E+0	9.8	1.8E+4	
PZGB	1	2K	µg/L	0.02	28	3.2E+4	3.2E+4	1.5E+5	7.1E+0	9.5E+3	6.7E+4	2.2E+5	7.3E+4	8.5E+2		7.7E-1	3.1E+3	1.5E+0	6.4E-1	5.1E+0	6.3E+0	10.1	1.4E+4	
PZGB	2	2K	µg/L	0.02	28	2.9E+4	3.3E+4	1.6E+5	6.3E+0	7.9E+3	7.0E+4	2.0E+5	6.6E+4	7.1E+2	3.1E+1	9.3E-2	2.8E+3	9.3E-1	6.2E-1	5.7E+0	4.9E+0	10.1	1.4E+4	
PZGB	1	2K	µg/L	0.02	91	2.8E+4	3.3E+4	2.1E+5	1.5E+2	1.2E+4	6.2E+4	3.6E+5	7.6E+4	4.6E+2	5.2E-1	1.5E+0	3.9E+3	1.0E+0	4.5E-1	6.8E+0	4.3E+0	9.7		
PZGB	2	2K	µg/L	0.02	91	2.7E+4	3.3E+4	1.9E+5	4.2E+1	9.5E+3	6.2E+4	3.4E+5	6.3E+4	4.2E+2	3.5E-1	8.1E-2	3.2E+3	3.4E-1	3.7E-1	2.8E+0	1.1E+0	9.7		
PZGB	1	2K	µg/L	0.45	91	2.7E+4	3.4E+4	2.1E+5	1.8E+2	5.5E+3	6.3E+4	3.6E+5	7.3E+4	3.7E+2	3.2E+0	7.4E-1	3.6E+3	2.0E+0	1.2E+1	2.2E+1	9.7E+1	9.7		
PZGB	2	2K	µg/L	0.45	91	2.8E+4	3.5E+4	2.0E+5	1.2E+2	5.9E+3	6.5E+4	3.4E+5	7.3E+4	5.8E+2	1.9E+0	8.0E-1	3.4E+3	4.1E+0	5.3E+0	2.3E+1	3.4E+1	9.7		
PZGBh	1	20K	µg/L	0.02	7	2.4E+5	2.9E+4	9.2E+5	1.3E+1	1.2E+4	4.0E+4	3.3E+6	3.9E+5	4.2E+2	5.3E+1	9.9E-2	1.7E+4	4.7E-1	9.3E-2	3.8E-1	6.4E-1	10.6	1.6E+4	
PZGBh	2	20K	µg/L	0.02	7	2.4E+5	2.9E+4	9.3E+5	4.5E+0	1.1E+4	3.2E+4	3.3E+6	3.6E+5	4.0E+2		2.6E-1	1.7E+4	4.7E-1	4.1E-2	5.6E-1	6.5E-1	10.6	1.6E+4	
PZGBh	1	20K	µg/L	0.02	28	2.6E+5	3.3E+4	1.0E+6	3.8E+2	1.3E+4	3.1E+4	3.1E+6	4.8E+5	3.6E+3		1.2E+1	1.9E+4	6.6E+0	9.0E-2	6.5E-2	3.7E-1	10.2	1.9E+4	
PZGBh	2	20K	µg/L	0.02	28	2.7E+5	3.3E+4	1.0E+6	2.9E+1	1.2E+4	3.0E+4	3.1E+6	4.4E+5	4.4E+2	2.4E+1	2.4E-1	2.0E+4	2.0E-1	1.1E-1	2.2E-1	6.4E-1	10.2	1.9E+4	
PZGBh	1	20K	µg/L	0.02	91	3.3E+5	3.6E+4	1.1E+6	7.5E+2	2.0E+4	3.3E+4	3.5E+6	5.0E+5		8.2E-2	2.3E+0	2.2E+4	2.5E+1	1.3E-1	8.4E-1	5.8E-1	10.1		
PZGBh	2	20K	µg/L	0.02	91	3.4E+5	3.9E+4	1.1E+6	1.5E+2	1.6E+4	3.2E+4	3.6E+6	4.9E+5		8.2E-2	7.3E-1	2.2E+4	5.2E+0	2.6E-1	8.3E-1	5.6E-1	10.1		
PZGBh	1	20K	µg/L	0.45	91	3.5E+5	3.9E+4	1.1E+6	7.0E+1	1.2E+4	3.3E+4	3.5E+6	4.9E+5		8.2E-2	1.9E+0	2.2E+4	9.3E+0	3.1E+0	9.9E+0	1.0E+1	10.1		
EJ Leachant			µg/L	0.02		3.0E+1	1.3E+2	4.7E+4	4.3E+1	8.0E+2	4.6E+4	7.3E+3	5.1E+3	4.0E+3	1.3E+1	2.3E+1	1.6E+0	6.4E-1	4.3E-2	1.2E-1	1.2E-2	8.7	1.8E+4	
PZEJ	1	2K	µg/L	0.02	7	2.9E+4	9.6E+2	1.6E+5	3.5E+1	9.9E+3	4.3E+4	2.9E+5	5.5E+4	8.3E+2	4.9E+1	2.7E+0	3.0E+3	1.6E+0	2.0E+0	8.3E+0	1.5E+1	9.3	2.1E+4	
PZEJ	2	2K	µg/L	0.02	7	2.8E+4	3.4E+2	1.5E+5	3.4E+0	8.2E+3	4.0E+4	2.9E+5	5.0E+4	4.8E+2	3.5E+1	3.9E-1	2.6E+3	2.7E-1	3.8E-1	3.1E+0	4.9E+0	9.4	2.1E+4	
PZEJ	1	2K	µg/L	0.02	28	2.6E+4	2.1E+2	1.4E+5	6.8E+0	8.4E+3	4.0E+4	2.1E+5	4.9E+4	4.5E+2	2.5E+1	5.4E-1	2.5E+3	1.0E+0	1.5E-1	3.1E+0	3.2E+0	9.6	1.9E+4	
PZEJ	2	2K	µg/L	0.02	28	2.9E+4	1.4E+2	1.5E+5	5.9E+1	8.0E+3	3.8E+4	2.2E+5	4.8E+4	1.1E+3	9.2E+0	2.8E+0	2.3E+3	1.6E+0	4.6E-1	3.4E+0	3.5E+0	9.6	1.9E+4	
PZEJ	1	2K	µg/L	0.02	91	2.7E+4	3.6E+2	1.6E+5	7.2E+1	1.2E+4	4.4E+4	3.5E+5	5.9E+4	2.6E+2	4.1E-1	1.8E-1	2.9E+3	1.1E-1	5.3E-1	3.4E+0	2.8E+0	9.7	2.2E+4	
PZEJ	2	2K	µg/L	0.02	91	2.8E+4	2.8E+2	1.5E+5	3.0E+1	1.0E+4	4.5E+4	3.1E+5	5.7E+4	1.0E+2	5.0E-1	6.9E-1	2.7E+3	3.3E-1	1.4E+0	3.3E+0	1.9E+0	9.7	2.2E+4	
PZEJ	1	2K	µg/L	0.45	91	2.8E+4	3.5E+2	1.5E+5	7.8E+1	8.0E+3	4.6E+4	3.5E+5	5.8E+4	1.3E+2	3.2E+0	6.6E-1	2.5E+3	1.7E+0	8.5E+0	3.7E+1	6.7E+1	9.7	2.2E+4	
PZEJ	2	2K	µg/L	0.45	91	2.7E+4	2.2E+2	1.5E+5	5.1E+1	7.7E+3	4.5E+4	3.1E+5	6.4E+4	9.2E+1	3.5E+0	6.0E-1	2.9E+3	1.5E+0	9.2E+0	3.6E+1	5.9E+1	9.7	2.2E+4	
PZEJh	1	20K	µg/L	0.02	7	2.5E+5	1.1E+3	9.7E+5	2.9E+1	2.4E+4	2.9E+4	3.2E+6	4.5E+5	5.7E+2	3.6E-1	4.7E-1	1.8E+4	2.5E+0	1.7E-1	8.1E-1	8.7E-1	9.3	2.0E+4	
PZEJh	2	20K	µg/L	0.02	7	2.5E+5	5.0E+2	1.0E+6	1.7E+1	2.4E+4	2.9E+4	3.2E+6	5.0E+5	4.1E+2	2.5E+1	1.6E-1	1.7E+4	1.7E-1	6.0E-3	5.6E-1	1.2E+0	9.4	2.0E+4	
PZEJh	1	20K	µg/L	0.02	28	2.6E+5	3.5E+2	1.1E+6	2.5E+0	1.9E+4	2.6E+4	3.1E+6	4.9E+5	4.6E+2	3.9E+1	9.2E-1	1.7E+4	3.2E-1	6.0E-2	5.6E-1	5.4E-1	9.6	2.3E+4	
PZEJh	2	20K	µg/L	0.02	28	2.6E+5	2.3E+2	1.1E+6	0.0E+0	1.8E+4	2.3E+4	3.1E+6	5.2E+5	3.7E+2	4.7E+1	2.9E-1	2.0E+4	1.4E-1	5.4E-2	2.7E-1	8.3E-1	9.6	2.4E+4	
PZEJh	1	20K	µg/L	0.02	91	3.4E+5	5.3E+2	1.1E+6	2.0E+2	3.3E+4	2.3E+4	3.3E+6	5.6E+5		6.1E-1	1.8E+0	2.5E+4	4.9E+0	9.5E-1	1.0E+0	6.1E+0	10.2	3.3E+4	
PZEJh	2	20K	µg/L	0.02	91	3.5E+5	3.6E+2	1.1E+6	2.8E+1	3.1E+4	2.3E+4	3.6E+6	5.3E+5		6.1E-1	1.5E+0	2.4E+4	4.1E+0	1.7E+0	2.3E+0	6.1E+0	10.2	3.3E+4	
PZEJh	1	20K	µg/L	0.45	91	3.5E+5	3.0E+2	1.1E+6	2.7E+1	1.8E+4	2.4E+4	3.3E+6	5.5E+5		6.1E-1	9.4E-1	2.5E+4	1.2E+1	2.4E+0	1.9E+0	6.2E+0	10.2	3.3E+4	

TABLE B1. (contd.)

Leachate results from PCT-Bs with salt-loaded zeolite 4A samples in 311 glass leachant (GB) and EJ-13 ground water (EJ) (contd.).																							
Test	#	S/V	Meas.	Filter	days	Li	B	Na	Mg	Al	Si	Cl	K	Ca	Y	Sr	Cs	Ba	La	Ce	Nd		
PZGB	1	2K	ER%	0.02	7	84.532	2.3173	15.989	-72.18	1.7047	2.2333	80.515	44.794	-90.01	0.4896	-210.2	14.66	-0.151	0.0176	0.0765	0.0517		
PZGB	2	2K	ER%	0.02	7	88.746	-0.577	21.429	-49.87	1.4212	0.9039	96.188	42.955	-91.66		-210.6	14.564	-0.15	0.0199	0.0756	0.0764		
PZGB	1	2K	ER%	0.02	28	88.907	-0.522	17.898	-66.72	2.2418	2.1511	61.649	47.633	-90.31		-210.1	18.271	-0.128	0.018	0.0788	0.0605		
PZGB	2	2K	ER%	0.02	28	80.394	1.9535	19.412	-70.25	1.8369	2.8747	56.714	42.505	-91.91	11.619	-210.6	16.887	-0.139	0.0174	0.0885	0.0467		
PZGB	1	2K	ER%	0.02	91	78.377	-0.117	34.195	603.25	2.9762	0.9987	102.57	49.365	-94.74	0.1953	-209.7	23.01	-0.137	0.0125	0.1058	0.0418		
PZGB	2	2K	ER%	0.02	91	74.819	0.9998	29.248	98.531	2.2448	0.788	96.769	40.977	-95.2	0.1296	-210.6	18.787	-0.149	0.0103	0.0441	0.0107		
PZGB	1	2K	ER%	0.45	91	75.488	4.8092	35.308	722.08	1.2331	1.1034	102.57	47.335	-95.81	1.2114	-210.2	21.231	-0.12	0.3538	0.3409	0.9318		
PZGB	2	2K	ER%	0.45	91	76.567	6.5455	31.884	443.51	1.3514	1.6739	96.769	47.386	-93.41	0.7248	-210.1	20.135	-0.082	0.1524	0.3584	0.3233		
PZGBh	1	20K	ER%	0.02	7	65.363	-11.16	23.98	-39.4	0.2774	-0.45	95.59	26.651	-95.2	1.989	-21.06	9.9565	-0.015	0.0002	0.0006	0.0006		
PZGBh	2	20K	ER%	0.02	7	64.805	-10.92	24.154	-78.67	0.2608	-0.618	95.59	24.106	-95.43		-21.05	9.9991	-0.015	8E-05	0.0008	0.0006		
PZGBh	1	20K	ER%	0.02	28	71.144	2.1514	26.966	1704.6	0.3029	-0.643	90.075	32.263	-59.21		-20.28	11.355	-0.004	0.0002	8E-05	0.0004		
PZGBh	2	20K	ER%	0.02	28	73.784	0.8166	27.011	34.83	0.2845	-0.68	90.946	29.653	-95.02	0.915	-21.05	11.804	-0.015	0.0003	0.0003	0.0006		
PZGBh	1	20K	ER%	0.02	91	92.067	11.579	28.675	3409	0.4751	-0.604	100.23	33.659		0.0031	-20.92	13.322	0.0297	0.0003	0.0013	0.0006		
PZGBh	2	20K	ER%	0.02	91	94.117	18.502	29.045	616.36	0.398	-0.619	103.51	33.182		0.0031	-21.02	12.967	-0.006	0.0007	0.0013	0.0005		
PZGBh	2	20K	ER%	0.45	91	96.559	19.872	29.775	230.83	0.2933	-0.604	100.23	33.17		0.0031	-20.94	12.831	0.0011	0.0089	0.0155	0.0099		
PZEJ	1	2K	ER%	0.02	7	80.953	59.353	33.962	-84.42	2.2853	-0.531	81.182	34.31	-88.53	13.807	-12.96	17.574	0.0177	0.0558	0.1278	0.1405		
PZEJ	2	2K	ER%	0.02	7	78.351	5.8667	30.3	35.746	1.8393	-1.413	82.053	30.353	-72.8	8.5567	-14.38	15.192	-0.007	0.0098	0.0471	0.0467		
PZEJ	1	2K	ER%	0.02	28	71.016	765.36	26.916	-33.62	1.8959	-1.242	57.672	30.031	-85.68	4.5216	-14.29	14.907	0.0065	0.003	0.0471	0.0309		
PZEJ	2	2K	ER%	0.02	28	79.838	280.82	28.534	-60.77	1.7939	-1.853	62.897	29.583	-89.59	-1.272	-12.86	13.808	0.0163	0.012	0.0505	0.0331		
PZEJ	1	2K	ER%	0.02	91	73.975	163.25	31.263	-94.28	2.7083	-0.35	99.468	37.141	-88.45	-4.572	-14.51	17.077	-0.009	0.014	0.0506	0.0265		
PZEJ	2	2K	ER%	0.02	91	77.053	77.302	30.902	-100	2.3804	-0.01	87.858	35.544	-90.55	-4.539	-14.19	16.253	-0.006	0.0399	0.0496	0.0186		
PZEJ	2	2K	ER%	0.45	91	75.759	303	29.935	349.74	1.807	0.0491	99.468	35.921		-3.54	-14.22	15.135	0.0185	0.2437	0.5687	0.648		
PZEJ	2	2K	ER%	0.45	91	74.352	174.82	30.869	-35.04	1.7211	-0.211	87.858	40.16		-3.402	-14.25	17.285	0.0149	0.2638	0.5542	0.5664		
PZEJh	1	20K	ER%	0.02	7	69.706	163.25	26.633	-94.28	0.588	-0.407	92.667	30.566	-88.45	-0.459	-1.433	10.432	0.0033	0.0004	0.0011	0.0008		
PZEJh	2	20K	ER%	0.02	7	69.35	77.302	28.514	-100	0.5905	-0.398	92.667	33.761	-90.55	0.4719	-1.452	10.301	-8E-04	-1E-04	0.0007	0.0012		
PZEJh	1	20K	ER%	0.02	28	72.102	303	31.19	349.74	0.4636	-0.477	89.184	33.005		1.0038	-1.405	10.215	-6E-04	5E-05	0.0007	0.0005		
PZEJh	2	20K	ER%	0.02	28	70.867	174.82	29.155	-35.04	0.428	-0.551	88.314	35.315		1.2866	-1.444	11.981	-9E-04	3E-05	0.0002	0.0008		
PZEJh	2	20K	ER%	0.02	91	94.526	124.35	30.245	-38.12	0.8063	-0.53	95.57	37.75		-0.45	-1.352	15.109	0.0076	0.0026	0.0014	0.0059		
PZEJh	1	20K	ER%	0.02	91	95.922	-100	29.778	-100	0.7442	-0.536	104.28	36.019	-100	-0.45	-1.371	14.567	0.0062	0.0048	0.0034	0.0058		
PZEJh	2	20K	ER%	0.45	91	96.602	-100	30.819	-100	0.4327	-0.527	95.57	37.528	-100	-0.449	-1.404	14.673	0.0211	0.0068	0.0028	0.0059		

TABLE B1. (contd.)

Leachate results from PCT-Bs with salt-loaded zeolite 4A samples in 311 glass leachant (GB) and EJ-13 ground water (EJ) (contd.)																					
Test	#	S/V	Meas.	Filter	days	Li	B	Na	Mg	Al	Si	Cl	K	Ca	Y	Sr	Cs	Ba	La	Ce	Nd
PZGB	1	2K	NLi	0.02	7	1.1271		0.2132		0.0227	0.0298	1.0735	0.5973		0.0065	-2.803	0.1955	-0.002	0.0002	0.001	0.0007
PZGB	2	2K	NLi	0.02	7	1.1833		0.2857		0.0189	0.0121	1.2825	0.5727			-2.808	0.1942	-0.002	0.0003	0.001	0.001
PZGB	1	2K	NLi	0.02	28	1.1854		0.2386		0.0299	0.0287	0.822	0.6351			-2.802	0.2436	-0.002	0.0002	0.0011	0.0008
PZGB	2	2K	NLi	0.02	28	1.0719		0.2588		0.0245	0.0383	0.7562	0.5667		0.1549	-2.808	0.2252	-0.002	0.0002	0.0012	0.0006
PZGB	1	2K	NLi	0.02	91	1.045		0.4559		0.0397	0.0133	1.3676	0.6582		0.0026	-2.796	0.3068	-0.002	0.0002	0.0014	0.0006
PZGB	2	2K	NLi	0.02	91	0.9976		0.39		0.0299	0.0105	1.2902	0.5464		0.0017	-2.808	0.2505	-0.002	0.0001	0.0006	0.0001
PZGB	1	2K	NLi	0.45	91	1.0065		0.4708		0.0164	0.0147	1.3676	0.6311		0.0162	-2.802	0.2831	-0.002	0.0047	0.0045	0.0124
PZGB	2	2K	NLi	0.45	91	1.0209		0.4251		0.018	0.0223	1.2902	0.6318		0.0097	-2.802	0.2685	-0.001	0.002	0.0048	0.0043
PZGBh	1	20K	NLi	0.02	7	0.8715		0.3197		0.0037	-0.006	1.2745	0.3553		0.0265	-0.281	0.1328	-2E-04	3E-06	8E-06	8E-06
PZGBh	2	20K	NLi	0.02	7	0.8641		0.322		0.0035	-0.008	1.2745	0.3214			-0.281	0.1333	-2E-04	1E-06	1E-05	8E-06
PZGBh	1	20K	NLi	0.02	28	0.9486		0.3595		0.004	-0.009	1.201	0.4302			-0.27	0.1514	-5E-05	3E-06	1E-06	5E-06
PZGBh	2	20K	NLi	0.02	28	0.9838		0.3601		0.0038	-0.009	1.2126	0.3954		0.0122	-0.281	0.1574	-2E-04	4E-06	4E-06	8E-06
PZGBh	1	20K	NLi	0.02	91	1.2276		0.3823		0.0063	-0.008	1.3365	0.4488		4E-05	-0.279	0.1776	0.0004	5E-06	2E-05	7E-06
PZGBh	2	20K	NLi	0.02	91	1.2549		0.3873		0.0053	-0.008	1.3802	0.4424		4E-05	-0.28	0.1729	-8E-05	1E-05	2E-05	7E-06
PZGBh	1	20K	NLi	0.45	91	1.2874		0.397		0.0039	-0.008	1.3365	0.4423		4E-05	-0.279	0.1711	1E-05	0.0001	0.0002	0.0001
PZEJ	1	2K	NLi	0.02	7	1.0794		0.4528		0.0305	-0.007	1.0824	0.4575		0.1841	-0.173	0.2343	0.0002	0.0007	0.0017	0.0019
PZEJ	2	2K	NLi	0.02	7	1.0447		0.404		0.0245	-0.019	1.094	0.4047		0.1141	-0.192	0.2026	-9E-05	0.0001	0.0006	0.0006
PZEJ	1	2K	NLi	0.02	28	0.9469		0.3589		0.0253	-0.017	0.769	0.4004		0.0603	-0.191	0.1988	9E-05	4E-05	0.0006	0.0004
PZEJ	2	2K	NLi	0.02	28	1.0645		0.3805		0.0239	-0.025	0.8386	0.3944		-0.017	-0.171	0.1841	0.0002	0.0002	0.0007	0.0004
PZEJ	1	2K	NLi	0.02	91	0.9863		0.4168		0.0361	-0.005	1.3262	0.4952		-0.061	-0.193	0.2277	-1E-04	0.0002	0.0007	0.0004
PZEJ	2	2K	NLi	0.02	91	1.0274		0.412		0.0317	-1E-04	1.1714	0.4739		-0.061	-0.189	0.2167	-7E-05	0.0005	0.0007	0.0002
PZEJ	1	2K	NLi	0.45	91	1.0101		0.3991		0.0241	0.0007	1.3262	0.4789		-0.047	-0.19	0.2018	0.0002	0.0032	0.0076	0.0086
PZEJ	2	2K	NLi	0.45	91	0.9914		0.4116		0.0229	-0.003	1.1714	0.5355		-0.045	-0.19	0.2305	0.0002	0.0035	0.0074	0.0076
PZEJh	1	20K	NLi	0.02	7	0.9294		0.3551		0.0078	-0.005	1.2356	0.4075		-0.006	-0.019	0.1391	4E-05	5E-06	1E-05	1E-05
PZEJh	2	20K	NLi	0.02	7	0.9247		0.3802		0.0079	-0.005	1.2356	0.4501		0.0063	-0.019	0.1373	-1E-05	-1E-06	9E-06	2E-05
PZEJh	1	20K	NLi	0.02	28	0.9614		0.4159		0.0062	-0.006	1.1891	0.4401		0.0134	-0.019	0.1362	-8E-06	7E-07	9E-06	7E-06
PZEJh	2	20K	NLi	0.02	28	0.9449		0.3887		0.0057	-0.007	1.1775	0.4709		0.0172	-0.019	0.1597	-1E-05	4E-07	3E-06	1E-05
PZEJh	1	20K	NLi	0.02	91	1.2603		0.4033		0.0108	-0.007	1.2743	0.5033		-0.006	-0.018	0.2015	0.0001	3E-05	2E-05	8E-05
PZEJh	2	20K	NLi	0.02	91	1.279		0.397		0.0099	-0.007	1.3904	0.4803		-0.006	-0.018	0.1942	8E-05	6E-05	4E-05	8E-05
PZEJh	1	20K	NLi	0.45	91	1.288		0.4109		0.0058	-0.007	1.2743	0.5004		-0.006	-0.019	0.1956	0.0003	9E-05	4E-05	8E-05

APPENDIX C
MATRIX FOR CERAMIC WASTE FORM TESTING

1. Product Consistency Tests of Salt-Loaded Zeolite in DIW, EJ-13, 311 Leachant, and Ca and Mg — ASTM Brines

The symbols used in the table are as followed:

Sample Name Column;

- PZ Letters in the sample names, indicating a salt-loaded zeolite sample in a PCT test.
- D The third letter in the sample names, indicating DIW leachate.
- EJ Letters in the sample name, indicating an EJ-13 leachate.
- GB Letters in the sample name, indicating a 311 leachate.
- CB Letters in the sample name, indicating a Ca ASTM brine leachate.
- AB Letters in the sample name, indicating a Mg ASTM brine leachate.
- h This additional letter in the sample name indicates a 20,000 m⁻¹ test.
The absence of this letter indicates a 2,000 m⁻¹ test.
- Ca Indicates the Ca-Na salt-loaded zeolite 5A was used in the test.
- ***-XX-* Number of days the test ran.
- ***-**-X Sample number of the specific test.

Analysis Completed Column;

- x Indicates the test was terminated.
- C Solution carbon analysis was performed and the results reported.
- Cl Solution cation analysis for Cl was performed and the results reported.
- Cation Solution cation analysis was performed and the results reported.

TABLE C1. Matrix of PCT-Bs with Salt-Loaded Zeolite 4A

Sample Name	Date In	Date Out	Analysis Completed
20,000 m ⁻¹			
PZDh-3-1,2	2-5-96	2-8-96	x;C,Cl,Cation
PZDh-7-1,2	4-4-96	4-11-96	x;C,Cl,Cation
PZDh-28-1,2	4-4-96	5-2-96	x;C,Cl,Cation
PZDh-91-1,2	4-4-96	7-3-96	x;C,Cl,Cation
PZDh-365-1,2	4-4-96	4-4-97	
PZEJh-7-1,2	4-4-96	4-11-96	x;C,Cl,Cation
PZEJh-28-1,2	4-4-96	5-2-96	x;C,Cl,Cation
PZEJh-91-1,2	4-4-96	7-3-96	x;C,Cl,Cation
PZEJh-365-1,2	4-4-96	4-4-97	
PZGBh-7-1,2	4-4-96	4-11-96	x;C,Cl,Cation
PZGBh-28-1,2	4-4-96	5-2-96	x;C,Cl,Cation
PZGBh-91-1,2	4-4-96	7-3-96	x;C,Cl,Cation
PZGBh-365-1,2	4-4-96	4-4-97	
CaPZDh-7-3,4	5-2-96	5-9-96	x;C,Cl,Cation
CaPZDh-28-3,4	5-2-96	5-30-96	x;C,Cl,Cation
CaPZEJh-7-3,4	5-2-96	5-9-96	x;C,Cl,Cation
CaPZEJh-28-3,4	5-2-96	5-30-96	x;C,Cl,Cation
2,000 m ⁻¹			
PZ3P-3-1	2-20-96	2-23-96	x;C,Cl,Cation
PZ7P-3-1	2-20-96	2-23-96	x;C,Cl,Cation
PZ95P-3-1	2-20-96	2-23-96	x;C,Cl,Cation
PZD-2-1	2-5-96	2-5-96	x;C,Cl,Cation
PZD-1-1	2-5-96	2-5-96	x;C,Cl,Cation
PZD-3-1,2	2-5-96	2-8-96	x;C,Cl,Cation
PZD-7-1,2	4-8-96	4-15-96	x;C,Cl,Cation
PZD-28-3,4	2-14-96	3-13-96	x;C,Cl,Cation
PZD-91-1,2	4-8-96	7-8-96	x;C,Cl,Cation
PZD-365-1,2	4-8-96	4-8-97	
PZEJ-7-1,2	3-8-96	3-15-96	x;C,Cl,Cation
PZEJ-28-1,2	3-8-96	4-5-96	x;C,Cl,Cation
PZEJ-91-1,2	3-8-96	6-7-96	x;C,Cl,Cation
PZEJ-365-1,2	3-8-96	3-8-97	
PZGB-7-1,2	3-8-96	3-15-96	x;C,Cl,Cation
PZGB-28-1,2	3-8-96	4-5-96	x;C,Cl,Cation
PZGB-91-1,2	3-8-96	6-7-96	x;C,Cl,Cation
PZGB-365-1,2	3-8-96	3-8-97	

TABLE C1. (contd.)

PZAB-7-1,2	2-16-96	2-23-96	x;C,Cl,Cation
PZAB-28-1,2	2-16-96	3-15-96	x;C,Cl,Cation
PZAB-91-1,2	2-16-96	5-17-96	x;C,Cl,Cation
PZAB-365-1,2	2-16-96	2-16-97	
PZCB-7-1,2	2-16-96	2-23-96	x;C,Cl,Cation
PZCB-28-1,2	2-16-96	3-15-96	x;C,Cl,Cation
PZCB-91-1,2	2-16-96	5-17-96	x;C,Cl,Cation
PZCB-365-1,2	2-16-96	2-16-97	
CaPZD-7-3,4	5-2-96	5-9-96	x;C,Cl,Cation
CaPZEJ-7-3,4	5-2-96	5-9-96	x;C,Cl,Cation

2. Product Consistency Tests, Materials Characterization Center Test #1, and Vapor Hydration Tests of the Ceramic Waste Form in DIW, EJ-13, and Mg — ASTM Brines

The symbols used in the table are as follows:

Sample Name Column;

- M The first letter in the sample name, indicating a MCC-1 test.
- GZ Letters in the sample name, indicating a ceramic waste form sample.
- D Letter in the sample name, indicating DIW leachate.
- EJ Letters in the sample name, indicating an EJ-13 leachate.
- GB Letters in the sample name, indicating a 311 leachate.
- CB Letters in the sample name, indicating an ASTM Ca-Na-K-Cl brine.
- B Letter in the sample name, indicating an ASTM Mg-Na-K-Cl brine.
- h This additional letter in the sample name indicates a 20,000 m⁻¹ test.
The absence of this letter indicates a 2,000 m⁻¹ test.
- Ca Indicates the Ca-Na salt-loaded zeolite 5A was used in the test.
- ***-XX-* Number of days the test ran.
- ***-**-X Sample number of the specific test.

Analysis Completed Column;

- x Indicates the test was terminated.
- C Solution carbon analysis was performed and the results reported.
- Cl Solution cation analysis for Cl was performed and the results reported.
- Cation Solution cation analysis was performed and the results reported.

TABLE C2. Matrix of VHT and MCC-1 Tests with Ceramic Waste Form Samples

Sample Name	Date In	Date Out	Analysis Completed
MGZD-3-1,2,3	6/24/96	6/27/96	x;C,Cl,Cation
MGZD-7-1,2	6/24/96	7/1/96	x;C,Cl,Cation
MGZD-28-1,2	6/26/96	7/24/96	x;C,Cl,Cation
MGZD-91-1,2	6/26/96	9/25/96	x;
MGZB-3-1,2	6/24/96	6/27/96	x;C,Cl,Cation
MGZB-28-1,2	6/26/96	7/24/96	x;C,Cl,Cation
MGZEJ-3-1,2	6/24/96	6/27/96	x;C,Cl,Cation
MGZEJ-28-1,2	6/26/96	7/24/96	x;C,Cl,Cation
VGZ-200-3-1	6/24/96	6/27/96	x;
VGZ-200-7-1	6/24/96	7/1/96	x;
VGZ-200-21-1	6/26/96	7/17/96	x;
VGZ-200-35-1	6/26/96	7/31/96	x;
VGZ-200-56-1	6/26/96	8/21/96	x;
VGZ-200-TBD-1,2	7/10/96	TBD	
VGZ-150-7-1	6/24/96	7/1/96	x;
VGZ-150-21-1	6/26/96	7/17/96	x;
VGZ-150-35-1	6/26/96	7/31/96	x;
VGZ-150-56-1	6/26/96	8/21/96	x;
VGZ-150-91-1	6/26/96	9/25/96	
VGZ-150-TBD-1,2	7/10/96	TBD	
VGZ-120-21-1	6/26/96	7/17/96	x;
VGZ-120-35-1	6/26/96	7/31/96	x;
VGZ-120-56-1	6/26/96	8/21/96	x;
VGZ-120-91-1	6/26/96	9/25/96	x;
VGZ-120-182-1	7/10/96	1/8/97	
VGZ-120-TBD-1	7/10/96	TBD	

TABLE C3. Matrix of PCTs with Ceramic Waste Form Samples

Sample Name	Date In	Date Out	Analysis Completed
PCT, 20,000 m ⁻¹			
PGZDh-7-1,2			
PGZDh-28-1,2			
PGZDh-91-1			
PGZDh-182-1			
PGZDh-TBD-1			
PCT, 2,000 m ⁻¹			
PGZD-7-1 to 11			x
PGZD-28-1,2			
PGZD-91-1,2			
PGZD-182-1,2			
PGZD-365-1,2			
PGZD-TBD-1,2			
PGZEJ-7-1,2			
PGZEJ-28-1,2			
PGZEJ-91-1,2			
PGZEJ-365-1,2			
PGZEJ-TBD-1,2			
PGZGB-7-1,2			
PGZGB-28-1,2			
PGZGB-91-1,2			
PGZGB-365-1,2			
PGZGB-TBD-1,2			

APPENDIX D
MASS LOSS OF SALT-LOADED ZEOLITE AND GLASS MATERIAL AT
ELEVATED TEMPERATURES

1. 311 Glass Mass Dependence on Temperature

TABLE D1. Measurement of Volatiles (probably all water)
Adsorbed in the 311 Glass Powder

	Crucible A (g)	Crucible B (g)
Crucible weight: 1 hr., 150°C	6.1737	6.2360
Crucible weight: 1 hr., 20°C	6.1739	6.2363
311 Glass	1.0000	1.0000
Crucible + 311 glass	7.1739	7.2363
A in desiccator, B open: 24 hr.	7.1731	7.2363
wt% change	-0.06	0.00
24 hr, 150°C	7.1720	7.2344
wt% change	-0.17	-0.14
24 hr, 500°C	7.1705	7.2327
wt% change	-0.32	-0.33
2 hr, 850°C, hot	7.1672	7.2300
wt% change	-0.65	-0.60
2 hr, 20°C, cool	7.1694	7.2321
wt% change	-0.43	-0.39
Final 311 Glass Weight:	0.9955	0.9958
wt% change	-0.43	-0.39
Final Crucible Weight:	6.1739	7.2363

2. Mass Change of Salt-Loaded Zeolite, K-Li Glass, and 311 Glass at 700°C

TABLE D2. Measurement of Mass Change Due to Volatile Elements in the Salt-Loaded Zeolite 4A and Glass Samples

Sample A: Salt-Loaded Zeolite 4A.

Sample B: K-Li Glass.

Sample C: 311 Glass.

	Sample A (g)	Sample B (g)	Sample C (g)
Crucible weight: 1 hour, 150°C	5.4938	6.4147	6.2362
Crucible + sample	5.8893	6.7003	6.8552
Sample mass	0.3955	0.2856	0.6190
1 hr, 700°C	5.8814	6.6979	6.8521
wt% change	-2.00	-0.84	-0.50
4 hr, 700°C	5.8806	6.6979	6.8521
wt% change	-2.20	-0.84	-0.50
8 hr, 700°C	5.8792	6.6979	6.8521
wt% change	-2.55	-0.84	-0.50
24 hr, 700°C	5.8726	6.6979	6.8521
wt% change	-4.22	-0.84	-0.50
48 hr, 700°C	5.8634	6.6979	6.8521
wt% change	-6.55	-0.84	-0.50

3. Salt-Loaded Zeolite 4A Conversion to Sodalite and Nepheline

TABLE D3. Measurement of the Effect of Time and Na Dependence
on Zeolite 4A Conversion at 700°C

	Sample A (g)	Sample B (g)	Sample C (g)	Sample D (g)
Crucible wt.: 1 hr., 150°C	5.4936	6.4142	6.1732	6.2357
Crucible wt.: 1 hr., 20°C	5.4939	6.4148	6.1738	6.2362
Crucible + Sample wt.	6.3664	7.4148	7.1747	7.1613
Starting Sample wt.	0.8725	1.0000	1.0009	0.9251
Total wt., 1 hr, 700°C:	6.3507	7.3936	7.1559	7.1413
wt% change	-1.80	-2.12	-1.88	-2.16
	Zeolite	Zeolite	Zeolite	Zeolite
Crucible + Sample wt.	6.2331	7.2182	7.0100	6.9508
Sample wt.	0.7395	0.8040	0.8368	0.7151
Total wt., 2 hr, 700°C:	6.2289	7.2145	7.0058	6.9481
wt% change	-0.57	-0.46	-0.50	-0.38
	Zeolite	Zeolite	Zeolite	Z w/ peak spl.
Crucible + Sample wt.	6.1124	7.0882	6.9011	6.8454
Sample wt.	0.6168	0.6717	0.7238	0.6051
Total wt., 4 hr, 700°C:	6.1104	7.0859	6.8970	6.8408
wt% change	-0.32	-0.34	-0.57	-0.76
	Zeolite	Zeolite	Zeolite/NaCl	Sodalite/vwZ
Crucible + Sample wt.	5.9547	6.9048	6.7408	6.6864
Sample wt.	0.4582	0.4866	0.5642	0.4438
Total wt., 8 hr, 700°C:	5.9518	6.9008	6.7374	6.6795
wt% change	-0.63	-0.82	-0.60	-1.55
Crucible + Sample wt.	5.8100	6.7187	6.5900	6.5720
Sample wt.	0.3084	0.2962	0.4090	0.3280
Total wt., 24 hr, 700°C:	5.8020	6.7104	6.5822	6.5638
wt% change	-2.59	-2.80	-1.90	-2.50
	Neph/Sod.	Sod./Neph.	Ne./So./NaCl	Sod/Neph.
Crucible + Sample wt.	5.6731	6.6266	6.4841	6.4191
Sample wt.	0.1716	.2097	0.3038	0.1812
Total wt., 48 hr, 700°C:	5.6652	6.6239	6.4770	6.4169
wt% change	-4.60	-1.29	-2.34	-1.21
Sample A (TZO7):	Salt-loaded zeolite 4A.			
Sample B (TZK7):	1:1 Salt-loaded zeolite 4A : K-Li glass. (Appendix A)			
	(K-Li eutectic salt ratio substituted for Na in 311 glass).			
Sample C (TZNa7):	1:1 Salt-loaded zeolite 4A : NaCl			
Sample D (TZ37):	1:1 Salt-loaded zeolite 4A : 311 glass			

APPENDIX E XRD SPECTRA OF THE CERAMIC WASTE FORM AND SALT-LOADED ZEOLITE SAMPLES

CMT/ACL

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Fig. E1. The XRD Spectrum from a H005-2 Sample Corresponds Primarily to Sodalite.

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<cc>lgakudatFeb_96< Friday, Apr 19, 1996 @01:45p

Fig. E2. The XRD (TZO7-1-1) Spectrum from Sample A of Appendix D (salt-loaded zeolite) after 1 h at 700°C is Consistent with Salt-Loaded Zeolite and NaCl.

CMT/ACL

Fig. E3. The XRD (TZK7-1-1) Spectrum from Sample B of Appendix D (K-Li glass with salt-loaded zeolite) after 1 h at 700°C is Consistent with Salt-Loaded Zeolite and NaCl.

CMT/ACL

<c:\igakudat\feb_96> Friday, Apr 19, 1996 00:15:57p

Fig. E4. The XRD (TZ37-1-1) Spectrum from Sample D of Appendix D (311 glass with salt-loaded zeolite) after 1 h at 700°C is Consistent with Salt-Loaded Zeolite and NaCl.

CMT/ACL

<c:\vigakudat\ab_96> Friday, Apr 19, 1998 @01:59p

Fig. E5. The XRD (TZ37-2-1) Spectrum from Sample D of Appendix D (311 glass with salt-loaded zeolite) after 2 h at 700°C is Consistent with Salt-Loaded Zeolite and NaCl. Peak splitting occurred.

Fig. E6. The XRD (TZ37-4-1) Spectrum from Sample D of Appendix D (311 glass with salt-loaded zeolite) after 4 h at 700°C is Consistent with Sodalite and NaCl.

Fig. E7. The XRD (TZ07-4-1) Spectrum from Sample A of Appendix D (salt-loaded zeolite) after 4 h at 700°C is Consistent with Salt-Loaded Zeolite and NaCl. Only the salt-loaded zeolite with 311 glass sample changed structure after 4 hours.

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<viga\data\feb_96> Friday, Apr 19, 1996 00:15:59

Fig. E8. The XRD (TZO7-24-1) Spectrum from Sample A of Appendix D (NaCl with salt-loaded zeolite) after 24 h at 700°C is Consistent with Nepheline and NaCl.

Fig. E9. The XRD (TZNa7-24-1) Spectrum from Sample C of Appendix D (NaCl with salt-loaded zeolite) after 24 h at 700°C is consistent with Nepheline, Sodalite, and NaCl.

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<vlgakudata\lab_96> Friday, Apr 19, 1996 00:07:19

Fig. E10. The XRD (TZK7-24-1) Spectrum from Sample B of Appendix D (K-Li glass with salt-loaded zeolite) after 24 h at 700°C is Consistent with Sodalite and NaCl.

CMT/ACL

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Fig. E11. The XRD (TZ37-24-1) Spectrum from Sample D (salt-loaded zeolite with 311 glass) after 24 h at 700°C is Consistent with Sodalite and NaCl.

Fig. E12. The XRD Spectra from the Top (H0623-T), Middle (H0623-M), and Bottom (H0623-B) Third of the Ceramic Waste Form Designated H062-3 (monolith samples).

Fig. E13. Powdered XRD Spectra from the Ceramic Waste Form Samples Designated H059-1 and H062-2.

Fig. E14. Powdered XRD Spectra from the Ceramic Waste Form Samples Designated H0592-2, H0621-1, and H0623-3.

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