EVALUATING LONG-TERM PERFORMANCE
OF IN SITU VITRIFIED WASTE FORMS:
METHODOLOGY AND RESULTS

B. P. McGrail
K. M. Olson

November 1992

Prepared for
the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

Pacific Northwest Laboratory
Richland, Washington 99352

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

for

SUMMARY

In situ vitrification (ISV) is an emerging technology for the remediation of hazardous and radioactive waste sites. The concept relies on the principle of Joule heating to raise the temperature of a soil between an array of electrodes above the melting temperature. After cooling, the melt solidifies into a massive glass and crystalline block similar to naturally occurring obsidian. Determining the long-term performance of ISV products in a changing regulatory environment requires a fundamental understanding of the mechanisms controlling the dissolution behavior of the material. This is true of all vitreous waste forms proposed for hazardous and radioactive waste immobilization and not just ISV products.

A series of experiments was performed to determine the dissolution behavior of samples produced from the ISV processing of typical soils from the Idaho National Engineering Laboratory subsurface disposal area. Dissolution rate constant measurements were completed at 90°C over the pH range 2 to 11 for one sample obtained from a field test of the ISV process. The results showed an intrinsic dissolution rate constant approximately two orders of magnitude smaller than measured on a typical borosilicate waste glass proposed for vitrification of high-level radioactive waste. Depending on pH, this translates to a dissolution rate between 5 to 100 times slower than a waste glass. Extensive static testing of samples from pilot-scale tests and a field test of the ISV process were also completed. Devitrified samples from the field test showed statistically significant differences in dissolution behavior from amorphous samples of equivalent bulk composition taken from different locations in the melt. Depending on the element considered, and the type and length of the test, different conclusions could be drawn as to the effect of devitrification on the durability of the product. These findings illustrate the fallacy of using single point experiments, such as the product consistency test (PCT) test and materials characterization center (MCC) tests developed for comparing high-level waste glass durability, to make reliable comparisons among waste forms. A modeling approach is the only practicable means of predicting or comparing the long-term durability of these materials.

Using a consistent set of constraints on mineral precipitation, computer simulations with the EQ3/6 code reproduced the essential features of the solution chemistry observed in the static tests with a diverse range of ISV product samples of different elemental composition and thermal history. Differences in elemental releases among the different samples were predicted to be the result of different secondary solids that form during the course of the reaction of the ISV product with water. Post-test characterization of the secondary phases formed in the laboratory tests is needed to validate the model predictions.

CONTENTS

SUMMARY	iii
1.0 INTRODUCTION	1
1.1 NEED FOR ISV PRODUCT EVALUATION	1
1.2 PRODUCT EVALUATION APPROACH	3
1.3 REPORT ORGANIZATION	5
2.0 THEORY	7
2.1 REACTION PATH	7
2.2 REACTION KINETICS	8
3.0 EXPERIMENTAL	11
3.1 MATERIALS	11
3.1.1 Materials Selection	11
3.1.2 Materials Characterization	12
3.1.3 Sample Preparation	15
3.2 STATIC TESTS	16
3.2.1 Test Matrix	16
3.2.2 Static Testing Procedure	17
3.3 SINGLE-PASS FLOW-THROUGH TESTS	18
3.3.1 Apparatus	18
3.3.2 Buffer Solutions	19
3.3.3 Procedure	20
4.0 RESULTS	21
4.1 SINGLE-PASS FLOW-THROUGH TEST	21
4.2 STATIC TESTS	21
4.2.1 ES4-1 Glass	23
4.2.2 IFT Samples	24
5.0 DISCUSSION	31
5.1 SINGLE-PASS FLOW-THROUGH TESTS	31
5.2 STATIC TESTS	33
6.0 CONCLUSION	39
7.0 REFERENCES	41
APPENDIX A - CALCULATION OF SPECIFIC SURFACE AREA	A.1
APPENDIX B - ANALYTICAL CHEMISTRY RESULTS FROM STATIC TESTS	B.1

FIGURES

1	Comparison of Radionuclide Concentrations in Water Near an ISV Solid with SDWA Maximum Permissible Concentration.	3
2	Scanning Electron Micrographs and QEDS Analysis of Amorphous Matrix and Dendritic Crystallite in IFT Sample IC038D90IE	14
3	System $\text{CaO-MgO-Al}_2\text{O}_3\text{-SiO}_2$ for Compositions Lying in the 12% Al_2O_3 plane.	15
4	Range of (S/V) x time Values Used to Investigate the Static Dissolution Behavior of INEL ISV Glasses.	17
5	Schematic of Single-Pass Flow-Through Apparatus	19
6	Release of Selected Elements as a Function of Time and pH in Single-Pass Flow-Through Tests with IFT Glass IC007C90IE	22
7	Reaction Progress as a Function of S:t/V For Different S/V Test Ratios.	23
8	Comparison of Reaction Progress Values Calculated From the Release of Different Elements	24
9	Elemental Release From ES4-1 Glass as a Function of Reaction Progress.	25
10	Reaction Progress as a Function of S:t/V as Calculated From Solution Concentration of Several Elements	26
11	Elemental Release From Sample IC007C90IE as a Function of Reaction Progress	27
12	Elemental Release From Sample IC027B90IE as a Function of Reaction Progress	28
13	Results From Student's t-test in Comparing Elemental Releases Among Devitrified IFT Samples	30
14	Dissolution Rate as a Function of In Situ pH From Single-Pass Flow-Through Tests	32
15	Paragenetic Sequence of Minerals Predicted to Form in Static Tests With ES4 Glass	33
16	Paragenetic Sequence of Mineral Phases Predicted to Form in Static Tests With IC007C90IE Glass	36
17	Paragenetic Sequence of Mineral Phases Predicted to Form in Static Tests With IC027B90IE Glass	36

TABLES

1	Bulk Chemical Analysis of Sample ES4-1 and IFT Samples	13
2	Static Test Matrix for ISV Product Evaluation	16
3	Composition of Buffer Solutions Used for Single-Pass Flow-Through Tests	20
4	Regressed Values of Parameters η and $\log \hat{k}$ Obtained From Least-Squares Fit to Equation (16) and Data Shown in Figure 14	32

1.0 INTRODUCTION

In situ vitrification (ISV) is an emerging technology for the remediation of hazardous and radioactive waste sites. The concept relies on the principle of Joule heating to raise the temperature of a soil between an array of electrodes above the melting temperature. After cooling, the melt solidifies into a massive glass and crystalline block. The process is intended to either destroy a contaminant through pyrolysis and combustion or immobilize the waste in the product matrix.

There are a number of engineering issues that must be resolved before the ISV concept may be applied to an actual buried waste site (Nickelson, Luey, and Callow 1992). In addition to these engineering issues, there are also technical issues associated with the long-term effectiveness of ISV in reducing the toxicity and mobility of the waste and in demonstrating compliance with applicable regulatory requirements. In this report, we do not address issues associated with the destruction of organic hazardous wastes by the ISV process but focus on issues associated with the long-term immobilization of the waste materials.

1.1 NEED FOR ISV PRODUCT EVALUATION

The ISV process can produce melting temperatures from approximately 1300°C to 1800°C. These temperatures are 200°C to 700°C higher than the typical processing temperature for borosilicate waste glasses that have been developed for high-level radioactive waste disposal. The high melting temperature allows the process to be used on a wide range of soil compositions and results in a highly stable product similar to naturally occurring obsidian, which is known to have survived for centuries in the environment (Byers, Jercinovic, and Ewing 1987). Unfortunately, the visual similarity of the ISV product to natural obsidian (and waste glass) is often used to infer acceptability of long-term product performance. In fact, the long-term performance of the ISV products cannot be directly inferred from the behavior of naturally-occurring obsidians because ISV products are compositionally and microstructurally dissimilar and the chemical durability of a glass is highly dependent on its composition (Chick and Piepel 1984), homogeneity (Jantzen and Bickford 1985), and the conditions under which it is exposed to water (or water vapor). Acceptability of the ISV process can be derived only by demonstrating acceptable risks to the public from actual releases and transport paths that result in human exposure from the remediated site.

There are sufficient data in the published literature to estimate quantitatively the radiologic dose associated with releases from an ISV product at a low-level waste site. This simple analysis assumes that the dose contribution D_i ($\mu\text{Sv/yr}$) from an individual radionuclide i that has reached the biosphere can be approximated by

$$D_i = \frac{M_i f_i V_i e^{-\lambda_i R_i 0}}{W_i} \quad (1)$$

where

M_i = inventory of radionuclide i in the ISV solid

f_i = fraction of the radionuclide i that dissolves in the groundwater per unit time (yr^{-1})

V_i = dose conversion factor: dose rate from radionuclide i per unit concentration of i in the groundwater ($\mu\text{Sv/yr}$ per pCi/L)

λ_i = decay constant for radionuclide i (yr^{-1})

R_i = retardation factor for species i

θ = groundwater travel time to biosphere (yr)

W_i = dilution factor - a measure of dilution and dispersion of radionuclide i after release from the ISV product (m^3/yr).

To determine risks from long-term releases from an ISV product, we should solve Equation (1) for each radionuclide. However, the water travel time θ and dilution factor W_i involve a complicated analysis of transport that requires data on the site hydrology and geochemical interactions with the strata through which the radionuclide must travel to reach the biosphere. Since this information is not readily available, we simplify the analysis by examining doses or concentrations in the vicinity of the ISV solid. By setting $\theta \approx 0$, Equation (1) simplifies to

$$D_i = \frac{M_i f_i V_i}{W_i} \quad (2)$$

where W_i is now interpreted as the average groundwater flow rate through the LLW site. By dividing both sides of Equation (2) by V_i , we obtain a simple expression for the concentration C_i of radionuclide i in the vicinity of the ISV solid:

$$C_i = \frac{M_i f_i}{W_i} \quad (3)$$

Equation (3) provides a useful means to calculate the concentration of any radionuclide in groundwater contacting the ISV solid. This concentration can then be compared with standards set by the Resource Conservation and Recovery Act and Safe Drinking Water Act (SDWA) (Keller 1990). Figure 1 illustrates the results from such a calculation where the numerator was calculated by conservatively assuming that radionuclide release is controlled by dissolution of the ISV product, a fixed radionuclide inventory of 10^{-6} g/g, a surface area of 100 m^2 , and an average flow rate of 100 L/yr . The shaded area in Figure 1 represents the current range in expected dissolution rates for a typical ISV product as reported by McGrail

Figure 1. Comparison of Radionuclide Concentrations in Water Near an ISV Solid with SDWA Maximum Permissible Concentration. Shaded area represents typical range in dissolution rate of ISV products.

and Bates (1992). The results show that the SDWA standards of 15 pCi/L are exceeded by many orders of magnitude for the intermediate half-life ($25,000 < T_{1/2} < 5,000$ yr) and short half-life ($T_{1/2} < 5,000$ yr) radionuclides and by factors of 10 to 100 for the long-lived radionuclides. These results should not be interpreted as indicating that ISV product performance is unacceptable. The results do indicate, however, that additional data are needed on far-field transport processes and on the dissolution behavior of the ISV product to demonstrate acceptable radionuclide release rates to the biosphere. Additional data are particularly needed on radionuclide release from ISV products since many radionuclides are sparingly soluble in the environment and thus may be released at much lower values than predicted from the highly conservative dissolution rate model used in the calculations shown in Figure 1. By combining defensible release rate predictions from the ISV product with transport in the surrounding geologic media, we expect that the ISV product will indeed meet all regulatory criteria with a safety factor of perhaps several orders of magnitude.

1.2 PRODUCT EVALUATION APPROACH

Nearly all of the past work on ISV product evaluation has been directed at performing short-term laboratory tests to either demonstrate that the product meets "toxicity" criteria established by the Environmental Protection Agency (EPA) or compare the "leaching" behavior of ISV products with natural glasses and high-level nuclear waste glasses. In the

TCLP test established by the EPA, concentration limits are set on a series of heavy metals and organic compounds that are measured under a prescribed experimental format (Federal Register 1988). All ISV products produced to date have passed the TCLP test specifications. Unfortunately, the fact that ISV products passed these regulatory tests does not provide insight into the fundamental mechanisms that determine long-term product performance. The EPA "toxicity" specifications continue to be revised (usually more restrictive) and no TCLP "toxicity limits" have been established for radionuclides. The tests have also neglected the fact that ISV products are complex materials where partitioning of some elements such as Pb and Se into metallic phases has been observed. Release of partitioned elements may have little relationship to the chemical durability of the silicate matrix of the ISV product.

Because of these limitations, extensive comparison tests have been performed using closed-system test methods developed for studies of high-level nuclear waste glass behavior. The conclusion from these studies is that ISV products have "leach" rates 5 to 100 times slower than a typical borosilicate waste glass. However, the comparison of "leach rates" alone cannot be used to demonstrate acceptable product performance as demonstrated by Pigford and coworkers at the University of California-Berkeley (Pigford, Chambré, and Lee 1990). In their studies of high-level waste repository performance, they showed that "leach rate" differences among waste forms had no impact on radionuclide release rates from the disposal system. Exterior field mass transport was shown to overwhelmingly control the release rate. This logic is equally applicable to ISV products and other waste forms for waste immobilization. The slower dissolution rate of an ISV product compared with a high-level waste glass does not necessarily confer an improvement in performance of the immobilization system for a given radionuclide. In addition, comparisons of "leach rates" have the disadvantage of being done under a single set of experimental conditions at a single time. Evidence in the published literature (Van Isegham and Grambow 1988) and data that will be presented in this report show that a modeling approach is needed to extrapolate ISV product behavior in time.

Long-term performance of ISV products must be predicted from a model because direct laboratory observation of the product performance over time scales >1000 yr is impossible. Consequently, the goal of our product evaluation studies is not to provide comparisons between natural or waste glasses, but to develop a source-term model that can reliably predict the behavior of the material far into the future. The source-term model differs from empirical approaches used in past programs (Macedo, Barkatt, and Simmons 1982; Chick and Turcotte 1983) by using models developed from fundamental principles of physics, chemistry, thermodynamics, and mass transport. It is these types of models that the National Research Council has endorsed for assessing the performance of deep geologic repositories (Pigford et al. 1983). Mechanistic models allow performance predictions to be extrapolated in time and, more importantly, over environmental conditions that may not have been studied directly in the laboratory. Consequently, the laboratory testing conducted at Pacific

Northwest Laboratory (PNL)^a and reported here includes the measurement of the necessary rate constants, elemental solubilities, alteration phases, and other factors that are needed to develop and validate a scientifically-defensible model describing the reaction of an ISV product with an aqueous solution.

1.3 REPORT ORGANIZATION

This report includes an introduction to the theoretical basis for modeling glass-water interactions. The theoretical discussion is followed by descriptions of the test materials, characterization methodology, and sample preparation procedures used in the laboratory testing. The experimental section also describes the test matrix and procedures for a series of closed-system isothermal leach tests (static tests) and the apparatus and procedures for single-pass flow-through experiments. The results are presented in Section 4.0 and discussed in Section 5.0. Conclusions are summarized in Section 6.0.

^aOperated for the U.S. Department of Energy by Battelle Memorial Institute under Contract DE-AC06-76RLO 1830.

2.0 THEORY

The theoretical basis for modeling the reaction of silicate minerals and glasses with water has been an area of active research for over 20 years, and internationally accepted models are available that may be applied to the study of ISV glass-water reactions. For the purposes of this discussion, we provide a brief introduction to modeling the irreversible dissolution of silicate glasses and minerals in water followed by a description of reaction kinetics models.

2.1 REACTION PATH

Any chemical reaction can be written as

where A, B, etc. are the reactants and C, D, etc. are the products. For silicate glasses, dissolution reactions are irreversible processes and a master variable called reaction progress (ξ) is used to describe the overall extent of the conversion of the reactants into products. Changes in the quantities of the components A, B, etc. are given by

$$\Delta n_j = \int_{\xi_0}^{\xi_1} b_j d\xi \quad (5)$$

where

n_j = mass of component j in moles

b_j = coefficient of component j in the overall reaction.

The state of an aqueous solution with respect to the progress of some reaction between a solid and solution is commonly given by the saturation index (SI)

$$SI = \log \frac{Q}{K} \quad (6)$$

where Q is the ion activity product for the solid and K is the equilibrium constant. Saturation indices are zero for the case of exact saturation, negative for undersaturation, and positive for supersaturation. By convention, the SI is related to the chemical affinity of the overall reaction by

$$A = 2.303RT \cdot SI \quad (7)$$

where R is the gas constant and T is temperature. The ion activity product for any reaction is

given by

$$\log Q = b_{\text{H}_2\text{O}} \log a_{\text{H}_2\text{O}} + b_{\text{O}_2} \log f_{\text{O}_2} + \sum_{s=1}^M b_s \log \gamma_s m_s \quad (8)$$

where

$a_{\text{H}_2\text{O}}$ = activity of water

f_{O_2} = oxygen fugacity

M = number of aqueous species in reaction

γ_s = activity coefficient of species s

m_s = molal concentration of species s .

Equations 5 through 8 (along with equations for electrical balance) represent the central mass action equations needed to calculate the irreversible thermodynamics of glass dissolution. We have selected the EQ3/6 (Wolery 1983) series of equilibrium/reaction path computer codes to model the aqueous dissolution behavior of the ISV glasses. The EQ3/6 code was selected over other computer models such as PHREEQE (Parkhurst, Thorstenson, and Plummer 1980) because of the superior thermodynamic data base and solid-solution modeling capabilities available with EQ3/6. Although Equations 5-8 provide the essential information for modeling the chemical evolution of an aqueous solution contacting a glass, the model has no capabilities to predict the reaction kinetics. This topic is discussed in the next section.

2.2 REACTION KINETICS

Predicting the time-dependent rate of reaction for a silicate glass or mineral requires a kinetic rate equation describing the dependence of the reaction rate on the appropriate activities of ions in the aqueous phase. The most successful of the kinetic models that have been proposed are based upon transition-state theory (TST) (Aagaard and Helgeson 1982; Murphy and Helgeson 1987). The key assumption in these models is that the dissolution rate is controlled by the decomposition of an activated complex that leads to the detachment of a hydrolyzed surface group. A configuration for the activated complex leading to the dissolution of quartz has recently been calculated from *ab initio* computer simulations of the surface of quartz as a $\text{H}_3\text{SiOH} \cdot \text{H}_2\text{O}^\ddagger$ moiety (Lasaga and Gibbs 1990). Although these studies have significant implications for future model development, the practical application of TST ignores the details of the reaction mechanism to write a semi-empirical expression for the reaction rate derived from the Gibbs free energy change of the overall reaction as a function of reaction progress (Aagaard and Helgeson 1982):

$$\frac{d\xi}{dt} = S\hat{k}(1 - e^{\frac{-A(\xi)}{\sigma RT}}) \prod_i a_i^{-\eta_i} \quad (9)$$

where

S = surface area (m^2)

$\hat{k}$ = intrinsic rate constant [$\text{mol}/(\text{m}^2 \cdot \text{s})$]

η_i = stoichiometric reaction coefficient for the i th reactant species

A = overall chemical affinity (kcal/mol)

σ = stoichiometric factor for mol reactant per mol activated complex (usually 1).

For the hydrolysis of silicate glasses and minerals in dilute aqueous solutions where the $a_{\text{H}_2\text{O}}$ is essentially constant, Equation (9) reduces to

$$\frac{d\xi}{dt} = S\hat{k}a_{\text{H}^+}^{-\eta}(1 - e^{\frac{A(\xi)}{RT}}) \quad (10)$$

where the hydrogen ion activity is assumed to be the only parameter that directly modifies the rate law. Combining Equations (6), (7), and (10) gives the following convenient form for the dissolution rate law:

$$\frac{d\xi}{dt} = S\hat{k}a_{\text{H}^+}^{-\eta}(1 - \frac{Q(\xi)}{K}). \quad (11)$$

The key parameters that are required to apply Equation (11) are the intrinsic rate constant, $\hat{k}$, and the stoichiometric coefficient, η . The experimental methods we are employing to measure these parameters are described in Section 3.3. The parameters Q and K in Equation (11) have been proposed by Grambow (1985) to correspond to the ion activity product and equilibrium constant for a SiO_2 polymorph such as chalcedony or amorphous silica. Bourcier, Knauss, and Merzbacher (1989) proposed that Q and K correspond to a more complex pure phase or solid-solution consisting of an ideal mixture of amorphous SiO_2 and hydrous minerals. This hypothesis is intriguing because it allows the rate of glass reaction to be influenced by the activities of other ions solution rather than just orthosilicic acid as proposed by Grambow (1985). However, Advocat et al. (1990) found that Grambow's simpler approach adequately described the results from their tests with a complex nuclear waste glass. Modeling studies using both methods need to be conducted to determine which approach provides better agreement with ISV products.

3.0 EXPERIMENTAL

In this section, we describe the experimental work that has been conducted at PNL to test and validate the theory in Section 2.0 for application to ISV glass and crystalline waste forms. Two types of test methods were employed; static tests using procedures developed for evaluation of high-level nuclear waste glasses (MCC-1 and PCT tests) and flow-through tests designed to measure dissolution rate constants under controlled chemical conditions. The end goal of performing these tests is to demonstrate sufficient understanding of the reaction of ISV glass with water so that the release of radioactive and hazardous elements from the remediated waste site can be predicted with confidence.

At the present time, our experiments have been limited to samples from a pilot-scale test of the ISV process (Farnsworth et al. 1990) and from a field test (Callow et al. 1991) conducted at the Idaho National Engineering Laboratory (INEL). The field test was conducted at the subsurface disposal area (SDA) at the INEL. Application of the ISV process at the SDA is unique in that the site contains buried steel drums containing Pu-contaminated materials, organic wastes (such as trichloroethylene), and discarded lead shielding materials. The materials, characterization methods, and analytical results from the laboratory tests are given below.

3.1 MATERIALS

A description of the materials used and sample preparation method are given in the following sections.

3.1.1 Materials Selection

The ES-4 ISV test was completed in December 1989 at the PNL Pilot-Scale Test Facility. The melt geometry was in the form of a right circular cylinder approximately 60 cm in diameter and 60 cm in length. The melt materials consisted of approximately 286 kg of soil, 20 kg of stainless steel and 20 kg of low carbon steel, 1.7 kg of paper, and 1.7 kg of wood. A series of EP Tox metals (including Ag, Ba, Pb, and Se) were also distributed vertically along the centerline at 5 cm intervals.

Unfortunately, the large block of glass from this test was broken into pieces shortly after cooling to room temperature. Records were not kept as to the original location of the individual pieces. Consequently, the exact location in the melt of the samples taken for product evaluation is not known. However, the majority of the material consisted of what appeared to be relatively homogeneous gray-colored glass. One large (500 g) piece of glass, typical of this material was selected and designated as ES4-1.

Seven samples from intermediate-scale field test (IFT) 2 were sent to PNL by EG&G Inc. With the exception of a sample designated IC007C90IE, each of the field samples appeared to be partially devitrified with gray colored crystalline material interdispersed

between a green glass phase. Sample IC038C90IE appeared to be completely devitrified with no glass phase visible to the eye. The reader is referred to Callow et al. (1991) for a complete description of the field test and the location where the samples were obtained.

3.1.2 Materials Characterization

Bulk chemical analysis of the ISV samples was performed by NaOH/Na₂O₂ and KOH/KNO₃ fusions of samples ground to -200 mesh. The fusions were dissolved in deionized water and analyzed by inductively coupled argon plasma emission spectroscopy (ICP-AES). In addition, the Fe²⁺/Fe ratio was determined by standard colorimetric techniques. The results from these analyses are given in Table 1.

X-ray diffraction (XRD) analyses were also performed on each of the ISV samples. These analyses showed that the primary crystalline phases in the ES4 sample were quartz, cordierite (Mg₂Al₄Si₅O₁₈), and α -Fe. However, percent crystallinity was less than 1% by volume. In contrast, the only crystalline phase identified by XRD for sample IC007C90IE was α -Fe. For all the other IFT samples, both α -Fe and diopside (CaMgSi₂O₆) were identified. Figures 2a-2c are scanning electron micrographs (SEM) that show typical areas of the IFT samples that appeared to be completely crystalline to the eye. The figure clearly shows, however, that the crystals are embedded in an amorphous matrix. Figure 2d shows a typical quantitative energy dispersive x-ray spectroscopy (QEDS) analysis localized to a spot on the glassy matrix and a crystallite. The comparison shows that the crystals are enhanced in Mg, Ca, and Fe compared with the matrix. Diopside is a pyroxene with a crystal structure (XYZ₂O₆) consisting of alternating tetrahedral- and octahedral-coordinated layers. Both Fe²⁺ and Mg²⁺ readily substitute in the octahedral Y-site and in the distorted 6-8 coordinated X site. The exact stoichiometry could not be determined from the XRD analyses because of the high amorphous background and small differences in the d-spacings and intensities that occur as Fe²⁺ substitutes for Mg²⁺ in the structure. However, the XRD analysis and QEDS analysis are consistent with a general class of clinopyroxene minerals known as augite. Augite is a Ca-Mg-Fe rich silicate that is commonly found in volcanic rocks. Since no suitable standards for augite were available for comparison, %crystallinity in each of the IFT samples was estimated (see Table 1) by comparison of peak intensities with diopside. The peak heights were assumed to be linearly related to the mass fraction present in the ISV samples, which is not strictly accurate because of matrix absorption effects. However, the values should be good indicators of relative %crystallinity between samples.

Figure 3 shows part of the phase diagram for the system CaO-MgO-Al₂O₃-SiO₂ interpolated in the 12 wt% Al₂O₃ plane (Levin and McMurdie 1975). The analyzed composition for both ES4-1 and the IFT samples are shown. For ES4-1, both cordierite and quartz were identified from our XRD analyses consistent with two stable phases identified in Figure 3. The major compositional difference between the ES4 samples and the IFT samples is in the CaO content. The IFT samples have over three times more CaO by weight, which

Table 1. Bulk Chemical Analysis of Sample ES4-1 and IFT Samples in Wt%

Oxide	SAMPLE IDENTIFICATION NUMBER							
	<u>ES4-1</u>	<u>IC007C90IE</u>	<u>IC027B90IE</u>	<u>IC038B90IE</u>	<u>IC038C90IE</u>	<u>IC038D90IE</u>	<u>IC048B90IW</u>	<u>IC048C90IW</u>
Ag ₂ O	0.08	0.00	0.03	0.00	0.00	0.00	0.00	0.00
Al ₂ O ₃	12.77	11.30	12.20	12.00	12.00	11.90	12.50	12.50
BaO	0.11	0.40	0.09	0.09	0.09	0.09	0.09	0.09
CaO	4.63	11.50	10.70	10.90	10.50	10.70	8.77	8.48
Cr ₂ O ₃	0.00	0.00	0.26	0.27	0.26	0.26	0.13	0.14
FeO	4.44	3.68	3.94	3.36	3.96	3.90	4.42	4.21
Fe ₂ O ₃	0.03	0.47	0.17	0.79	0.22	0.24	0.19	0.39
K ₂ O	4.88	2.90	3.20	3.10	2.90	3.00	3.10	3.20
MgO	2.17	2.70	2.60	2.70	2.70	2.60	2.60	2.70
MnO	0.18	0.08	0.19	0.19	0.19	0.19	0.13	0.13
Na ₂ O	1.50	2.15	2.30	2.40	2.70	3.18	2.00	2.70
SeO ₂	0.04	0.00	0.00	0.00	0.00	0.00	0.00	0.00
SiO ₂	59.40	62.20	64.30	63.60	64.40	63.10	65.60	65.30
SrO	0.00	0.03	0.03	0.03	0.03	0.03	0.03	0.03
TiO ₂	0.90	0.54	0.58	0.58	0.57	0.58	0.61	0.62
TOTAL	91.05	97.95	100.56	100.01	100.52	99.77	100.17	100.49
Fe ²⁺ /Fe	0.923	0.897	0.963	0.827	0.952	0.947	0.963	0.924
%cryst	<1%	<1%	20%	4%	18%	21%	3%	4%

Figure 2. Scanning Electron Micrographs and QEDS Analysis of Amorphous Matrix and Dendritic Crystallite in IFT Sample IC038D90IE

Figure 3. System CaO-MgO-Al₂O₃-SiO₂ for Compositions Lying in the 12% Al₂O₃ plane. Symbols shown correspond to: ○ ES4 Glass, ● IFT Glasses.

places them in the stability region for diopside crystallization (Figure 3). The solidus temperature for this phase is between 1135 and 1200°C, so the location where the IFT samples were obtained must have been held near this temperature range for a period of time to promote crystallization of the diopside or augite.

3.1.3 Sample Preparation

Samples of glass to be used for MCC-1 type static tests (see Section 3.2.1) were cut into coupons of approximately 4 cm² surface area using a diamond impregnated saw. The samples were ultrasonically cleaned in acetone, absolute ethanol, and deionized water, and dried at room temperature for 2 h before being placed into a desiccator until needed. Samples to be used for modified PCT tests (described later) were prepared by breaking the glass into small pieces with a steel anvil and crushing the pieces with a mortar and pestle. The crushed material was sieved through a 100 mesh screen and collected on a 200 mesh screen. After sieving this material, the granules that did not pass through the 200 mesh screen were collected into 30 g batches. These batches were ultrasonically cleaned in deionized water by decanting stirred supernate three times and replacing with fresh water. The deionized water was decanted after the final rinse and replaced with 30 mL of absolute ethanol. After decanting the ethanol, the cleaned glass was dried for 15 min at 50°C in a

vacuum oven. The separate batches of glass were then combined into a single jar and stored in a desiccator until needed. An estimate of the specific surface area of the cleaned glass was calculated as indicated in Appendix A, assuming a mean spherical size particle of 0.112-mm diameter and a measured glass density of 2.7 g/m³ to give 0.02 m²/g. Surface area was also measured by N₂ BET. The measured area was 0.0138 ± 0.005 m²/g. The N₂ BET measurement exhibited distinct nonlinear behavior with relative pressure. Consequently, the accuracy of the measurement is uncertain. We have assumed a specific surface area of 0.02 m²/g for all data reported here.

3.2 STATIC TESTS

3.2.1 Test Matrix

A series of closed-system, isothermal experiments was designed to elucidate the dissolution behavior, alteration phase formation, and elemental solubilities for the INEL ISV glasses. The test matrix is given in Table 2. Three different surface area-to-volume (S/V) ratios were investigated with overlapping (S/V) X time as illustrated in Figure 4. This range was selected so that sufficient glass reaction occurs to saturate the leachate with respect to the major elements of concern. The 10 m⁻¹ experiments were performed with the MCC-1 method (Strachan, Turcotte, and Barnes 1981) and the higher S/V tests used a procedure called the Product Consistency Test (PCT) (Jantzen and Bibler 1987). The test matrix shown in Table 2 was performed in its entirety for samples ES4-1, IC007C90IE, IC027B90IE, IC038C90IE, and IC048B90IW. Samples IC038B90IE, IC038D90IE, and IC048C90IW were only tested in triplicate for 28 d duration using both MCC-1 and PCT test methods.

At the present time, only experiments at 90°C in deionized water have been conducted. Although 90°C is far above the expected temperature range where the ISV waste form may be contacted by water, the elevated temperature permits the more rapid formation of crystalline alteration products that may be identified by surface analysis techniques and gives a greater extent of alteration in a short period of time. Both of these factors simplify comparison of the experimental results with model predictions. It has also been recently

Table 2. Static Test Matrix for ISV Product Evaluation

<u>Material</u>	<u>Test</u>	<u>S/V, m⁻¹</u>	<u>Temperature</u>	<u>No. Tests</u>				
				<u>7d</u>	<u>14d</u>	<u>28d</u>	<u>56d</u>	<u>91d</u>
ISV Glass	MCC-1	10	90°C	1	3	1	1	3
Blank	MCC-1			1	1	1	1	1
ISV Glass	PCT	100	90°C	3	1	3	1	3
	PCT	1000	90°C	3	1	3	1	3
Blank	PCT			1	1	1	1	1

Figure 4. Range of (S/V) x time Values Used to Investigate the Static Dissolution Behavior of INEL ISV Glasses.

demonstrated that the basic mechanism of the reaction of a complex waste glass with water does not change up to 200°C (Delage and Dussossoy 1991). Consequently, assuming that the appropriate temperature dependencies of the rate constants are known, extrapolating to a lower temperature is straightforward using the mechanistic model described in Section 2.0. However, low temperature experiments should be planned in future work to validate the model at more realistic service temperatures.

3.2.2 Static Testing Procedure

A single coupon or appropriate mass of crushed glass was added to a Teflon PFA^a jar containing ≈40 ml of deionized water to achieve the desired S/V ratio. The jar was then sealed and placed in an oven that was kept at 90 ±2°C. Because Teflon PFA is a relatively porous polymer, atmospheric gases readily diffuse through the container wall, keeping the leachate in contact with air during the test (McGrail 1986). At the end of the planned test duration for the ES4-1 samples, the containers were removed from the oven and allowed to cool. The containers were then weighed and an aliquot of unfiltered leachate was removed to determine pH and discarded. The remaining leachate was filtered, using a 0.45 μm filter, into

^aPerfluoroalkoxy polymer, E. I. DuPont de Nemours, Wilmington, Delaware

three polyethylene vials. Two of the leachates were acidified to 1 wt% HNO₃; one was analyzed by inductively coupled plasma emission spectroscopy (ICP-AES) and the other placed in storage. The third leachate sample was analyzed by ion chromatography (IC).

Because the ES4-1 leachates were allowed to cool before filtering and acidification, precipitation of secondary phases may have occurred. To prevent this, the IFT samples were treated in a different manner as follows: The container was removed from the oven and weighed, then placed in a pre-heated metal block machined to fit the knurled containers. Immediately, three 10 mL aliquots of leachate were removed and filtered using a 0.45 μ m filter, and placed in three separate polyethylene vials. The pH of the leachate remaining in the container was then measured. The temperature was measured immediately after the pH measurement and was found to be 75 $\pm$ 5°C. The pH was also remeasured at room temperature. As before, two of the leachates were acidified; one was analyzed by ICP-AES and the other placed in storage. The unacidified sample was analyzed by IC.

3.3 SINGLE-PASS FLOW-THROUGH TESTS

As discussed in Section 2.2, modeling the dissolution kinetics of a silicate glass or mineral requires that the key parameters $\bar{k}$, the intrinsic rate constant, and η , the stoichiometric coefficient for a_{H^+} be known. In traditional static leach tests, these parameters cannot be determined because the chemical affinity term (1-Q/K) and pH change over the course of the test. Instead, the conditions of the experiment must be controlled so that these parameters are fixed. In a recent study by Knauss et al. (1990), the dissolution behavior of a simple 5-component borosilicate glass was studied over a pH range of 2 to 13 and temperatures from 25 to 70°C. Their results clearly showed that for their simple glass, the forward rate ($\bar{k}$)^a depended strongly on solution pH in addition to temperature. The functional dependency of $\bar{k}$ on H⁺ activity has been well documented for a variety of silicate minerals (Knauss and Wolery 1986). However, the study by Knauss et al. (1990) was the first to identify this dependence for a simple simulated waste glass. It remains to be seen whether similar behavior is observed for much more compositionally complex waste glasses and ISV glasses.

3.3.1 Apparatus

A single-pass flow-through cell was developed based on a modified design described in Knauss et al. (1990). The apparatus of Knauss et al. utilized a plug flow type of reaction cell. Testing conducted at PNL utilizing this design clearly showed that at 90°C, gas and/or water vapor bubbles formed in the inlet tubes leading to the cell. All efforts to minimize bubble formation by thoroughly degassing the supply water were unsuccessful in controlling the bubble formation. The bubbles are of concern in a plug-flow cell because they may exclude water contact over portions of the sample. Consequently, we modified the design of

^aThe forward rate is defined here as the reaction rate at constant temperature and a_{H^+} .

Knauss et al. from a plug-flow type of cell to a well-mixed tank type of cell that does not have a flow path directly through the sample (Figure 5). Our design also permits the maintenance of a controlled atmosphere during the test. With the present configuration, we are able to simultaneously run 24 separate experiments; 12 tests can be run with 12 different buffer solutions and the other 12 cells share a common buffer solution reservoir.

3.3.2 Buffer Solutions

Ten separate buffer solutions were prepared covering pH range from 2 to 12. The composition of the buffers was selected so that at most only 1 element (potassium) was present in both the buffer solutions and the glasses. The EQ3NR code was used to calculate the 25 and 90°C pH of the solutions. Electrical balance was achieved by adjusting a_{H^+} . The composition of the buffers as well as the measured and calculated pH values are given in Table 3. The room temperature pH of the effluent from the cells was checked periodically over the course of the test. No pH changes could be detected over the 60 d run duration within the experimental error of the pH measurement of approximately ± 0.1 pH unit.

Figure 5. Schematic of Single-Pass Flow-Through Apparatus

Table 3. Composition of Buffer Solutions Used for Single-Pass Flow-Through Tests

Buffer	Composition	I ^a	pH 25°C (meas)	pH 25°C (calc)	pH 90°C (calc)
1	0.005 m KHPht + 0.0123 m HCl	0.0131	2.38	2.13	2.14
2	0.005 m KHPht + 0.0033 m HCl	0.0062	3.07	3.00	2.96
3	0.005 m KHPht + 0.0003 m HCl	0.0051	3.97	3.97	4.04
4	0.005 m KHPht + 0.0018 m LiOH	0.0086	5.04	5.05	5.54
5	0.005 m KHPht + 0.0041 m LiOH	0.0131	5.89	5.92	6.41
6	0.005 m H ₃ BO ₃ + 0.0003 m LiOH	0.0003	8.00	8.09	7.74
7	0.005 m H ₃ BO ₃ + 0.0020 m LiOH	0.0019	9.00	9.07	8.68
8	0.005 m H ₃ BO ₃ + 0.0044 m LiOH	0.0044	9.93	10.02	9.30
9	0.005 m LiCl + 0.0107 m LiOH	0.0214	11.94	12.14	10.56
10	0.005 m LiCl + 0.1265 m LiOH	0.1142	12.65	12.91	11.33

^aStoichiometric ionic strength.

3.3.3 Procedure

Approximately 1 g of -100 +200 mesh glass from sample IC007C90IE was placed in each of 10 flow-through cells that was pre-filled with a buffer solution given in Table 2. The peristaltic pump was then turned on, giving a flow-rate of approximately 30 ml/d. The oven was then brought up to the test temperature of 90 ±2°C. Approximately every 3 to 4 days, each sample bottle was weighed and an aliquot of the effluent in each sample bottle was removed, placed in separate container, and acidified to 1% HNO₃. These samples were analyzed by ICP-AES. The remaining effluent solution was discarded and a clean sample bottle replaced in-line. The flow rate was calculated for each cell from the weight change and sample interval. Variability in the measured flow rates was small over the 60 d test duration averaging between 2 and 5%.

4.0 RESULTS

In this section, we present the test results from both flow-through and static experiments that have been completed on INEL ISV samples. See Section 5.0, Discussion, for an interpretation of these results.

4.1 SINGLE-PASS FLOW-THROUGH TEST

At the present time, single-pass flow-through experiments have been completed only on sample IC007C90IE at a single temperature of 90°C. Time-dependent concentrations measured for selected elements released from this sample are given in Figure 6. The data in Figure 6 have been normalized by the measured bulk composition of the sample so that equivalent concentrations of each element represent stoichiometric dissolution of the solid. As expected, dissolution of the glass appears to be essentially stoichiometric over the entire pH range with the exception of Na release which is consistently lower. The Na concentrations were near the ICP-AES detection limit for most of the tests so these data are probably unreliable. Note that steady-state concentrations are achieved after only a few days. These results contrast with those of Knauss et al. (1990) where the solution concentrations continued to decrease for several weeks. The rapid attainment of steady-state concentrations with our apparatus suggests that bubble formation with the plug-flow design used by Knauss et al. (1990) most likely accounts for the long times to achieve steady-state that they reported.

4.2 STATIC TESTS

A complete listing of the analytical chemistry results from both MCC-1 and modified PCT static tests with the INEL ISV glasses we have studied to date is given in Appendix B. We will present here selected data sets from these data to illustrate key points about the dissolution behavior of the materials. All of the analytical chemistry data were entered into an electronic format using the *Excel*^a software package which is a widely supported spreadsheet package for both IBM and Macintosh computer systems. However, the spreadsheet format is poorly suited for extracting data sets that meet specific criteria. To facilitate such queries, the *Excel* spreadsheet file was imported to an electronic database, *Superbase 4*^b. The *Superbase 4* software package was used to selectively extract the data sets used for the analyses reported in this Section. Currently, the ISV product evaluation database contains over 10,000 entries and is available from the authors in either *Excel* or *dBase* file format on request.

^a*Excel*[™] is a registered trademark of Microsoft Corporation, Redmond, Washington.

^b*Superbase 4*[™] is a registered trademark of Precision Software Inc., Irving, Texas.

Figure 6. Release of Selected Elements as a Function of Time and pH in Single-Pass Flow-Through Tests with IFT Glass IC007C90IE

4.2.1 ES4-1 Glass

Calculated values of the parameter reaction progress (ξ) are plotted in Figure 7 as a function of the combined variable $S \cdot t/V$. As long as the S/V values are not too large where the initial ion exchange can rapidly raise the solution pH, $S \cdot t/V$ provides a useful means of scaling static test data. The scaling law is clearly obeyed for ES4-1 glass as demonstrated by the excellent reproducibility of the data at different values of overlapping S/V . The line indicated in Figure 7 was calculated from a non-linear least-squares fit to a function of the form:

$$\xi = A(1 - e^{-B\tau}) + C\tau \quad (12)$$

where A, B, C are fitting constants and τ is $S \cdot t/V$ (d/m). Although a phenomenological basis for the form of Equation (12) can be derived, we simply use the equation as a smoothing function for calculating ξ from our test data. The reader should not construe Equation (12) as a mechanistic model describing the glass dissolution process as it is only valid for the conditions of this experiment.

Sodium was used for the fit shown in Figure 7 because of its high solubility and limited incorporation in secondary solids under the conditions of the experiment. Other elements, such as Al and Si become sequestered in secondary phases which makes them unsuitable for monitoring reaction progress as is clearly illustrated in Figure 8.

Figure 7. Reaction Progress (ξ) as a Function of $S \cdot t/V$ For Different S/V Test Ratios. Sodium concentrations were used to calculate ξ .

Figure 8. Comparison of Reaction Progress Values Calculated From the Release of Different Elements

Using Equation (12), a reaction progress value was calculated for each value of $S \cdot t/V$ in the static tests. This permits us to plot concentrations for each element released from the ISV product as a function of reaction progress (ξ) as illustrated in Figure 9. By converting the data to this format, it is then possible to directly compare the solution concentration values measured in the static tests with predictions from a geochemical reaction path code such as EQ3/6.

4.2.2 IFT Samples

As was done for the ES-4 samples, smoothing functions were fit to the static test data for the IFT samples as illustrated in Figure 10. However, Equation (12) provided a poor fit to the data so a smoothing function of the form

$$\xi = A \log \tau + B \quad (13)$$

was used instead. Sodium values were again used to track reaction progress. The slopes of the lines (A in Equation [13]) were found to vary from sample to sample from between 0.58 and 0.68. The proximity of the fit to a $\sqrt{t}$ dependence has led many researchers in the past to the incorrect conclusion that the glass dissolution proceeds by a diffusion-controlled process. As the data for Al, Ca, and Si clearly indicate, solution chemical effects control the release rate, and the proximity to a $\sqrt{t}$ behavior is an artifact of the complex changes in solution chemistry that occur as the ISV product dissolves in water.

Figure 9. Elemental Release From ES4-1 Glass as a Function of Reaction Progress. Lines were calculated from the EQ3/6 code.

Figure 10. Reaction Progress as a Function of $S \cdot t/V$ as Calculated From Solution Concentration of Several Elements

Releases of selected elements as a function of ξ are plotted in Figures 11 and 12 for samples IC007C90IE and IC027B90IE. Sample IC007C90IE was analyzed as x-ray amorphous whereas sample IC027B90IE was analyzed as 20% crystalline. Otherwise, the two samples are essentially identical in composition (see Table 1). A close examination of these two figures shows that there are distinct differences in the solution concentrations between the two samples. To obtain a more detailed understanding of the differences in the dissolution behavior among the IFT samples, a statistical comparison of the mean concentrations between the amorphous sample IC007C90IE and IFT samples IC048B90IW, IC038C90IE, and IC027B90IE was performed. The latter three samples are listed in the order of increasing %crystallinity. A two-sample approximate t-statistic was computed from the formula (DuMouchel 1987)

$$t_R = \frac{M_x - M_y}{\sqrt{\Delta^2 \left(\frac{1}{N_x} + \frac{1}{N_y} \right) e^{\frac{6}{N_x + N_y} - 0.1}}} \quad (14)$$

where

Figure 11. Elemental Release From Sample IC007C90IE as a Function of Reaction Progress. Lines were calculated from the EQ3/6 code.

Figure 12. Elemental Release From Sample IC027B90IE as a Function of Reaction Progress. Lines were calculated from the EQ3/6 code.

M_x = median of sample x

M_y = median of sample y

Δ = pooled interquartile range

N_x = number of observations in sample x

N_y = number of observations in sample y .

The computed approximate t-statistic is compared against a student's t-distribution with $N_x + N_y - 2$ degrees of freedom. Since the subject experiments were performed in triplicate, we selected a confidence level of 99% with 4 degrees of freedom. Thus, a computed t-statistic of greater than 3.75 or less than -3.75 indicates a statistically significant variance in the means of the two samples at the 99% confidence level.

The results from the computed t-statistic analysis are given in Figure 13. The shaded area represents the 99% confidence interval. Values falling above the shaded area have higher mean concentrations compared with sample IC007C90IE and values below the shaded area have smaller mean concentrations. Also drawn on Figure 13 are two vertical lines that represent the S·t/V values of two test methods that have been commonly used to compare between borosilicate waste glasses and between waste glasses and ISV products. The statistical analysis clearly shows that dissolution behavior of the devitrified ISV products is different from the amorphous sample IC007C90IE. However, there is no clear conclusion as to the relative effect of devitrification on the dissolution behavior. Release of Al is clearly smaller or unaffected by devitrification at S·t/V values less than 5000 d/m. However, at higher values of S·t/V, Al releases become significantly larger. Si release is lower throughout the entire S·t/V range we studied. Sodium, as does Al, shows initially lower releases at small S·t/V that finally exceed the mean concentration values for IC007C90IE at the largest S·t/V value studied. In Section 5.0, we provide a phenomenological explanation as to why this behavior is observed with the devitrified samples. The key point to be made here is that single point experiments, such as the PCT test and MCC-1 test, cannot be used to make reliable comparisons among waste forms. Based on an MCC-1 test alone, a conclusion could be drawn from the data in Figure 13 that the devitrified samples have better overall durability than the amorphous sample. This conclusion is clearly in error. Single point experiments only provide information at that single time and should never be used to extrapolate. Consequently, comparing the durability of an ISV glass with a natural obsidian or waste glass using a single point experiment, such as the PCT, is neither an accurate nor defensible basis for comparing the long-term durability of the materials.

Figure 13. Results From Student's t-test in Comparing Elemental Releases Among Devitrified IFT Samples

5.0 DISCUSSION

In this section, we discuss the results from both static and flow-through testing of ISV products to develop a consistent understanding of the dissolution behavior of the materials.

5.1 SINGLE-PASS FLOW-THROUGH TESTS

Dissolution rates in the single-pass flow-through experiments were calculated at each pH value from the steady-state concentrations of Si measured in the effluent. The measured Si concentrations were converted to dissolution rate by

$$\bar{k} = \frac{q\hat{C}_{Si}}{Sf_{Si}} \quad (15)$$

where

$\bar{k}$ = dissolution rate (g/m²)

$\hat{C}_{Si}$ = blank corrected steady-state concentration of Si (g/m³)

q = flow rate (m³/d)

f_{Si} = mass fraction of Si in the sample

S = surface area (m²).

The steady-state Si concentration was calculated from a fit to a function of the form $A \cdot (1 - e^{-Bt})$ as shown in Figure 6. A plot of the measured dissolution rate of sample IC007C90IE as a function of pH is shown in Figure 14. These data are compared with the dissolution rate measured for a borosilicate waste glass, designated P0798 (McGrail 1992).

To interpret the dissolution rate data obtained in the single-pass flow-through test, we divide both sides of Equation (11) by S and set $Q \approx 0$ to write

$$\log \bar{k} = \log \hat{k} + \eta[\text{pH}]. \quad (16)$$

From a plot of $\log \bar{k}$ versus pH, $\log \hat{k}$ and η can be determined from the intercept and slope, respectively, of a least-squares fit to Equation (16). The results from the fit to the data shown in Figure 14 are given in Table 4. In the neutral-to-alkaline pH range, where a typical groundwater contacting an ISV solid would be expected, the intrinsic rate constant ($\hat{k}$) for the ISV product is about two orders of magnitude smaller than the intrinsic rate for the

Figure 14. Dissolution Rate as a Function of In Situ pH From Single-Pass Flow-Through Tests

Table 4. Regressed Values of Parameters η and $\log \hat{k}$ Obtained From Least-Squares Fit to Equation (16) and Data Shown in Figure 14

Glass	η	$\log \hat{k}$ [g/(m ² ·d)]	η	$\log \hat{k}$ [g/(m ² ·d)]
	Acidic to Neutral		Neutral to Basic	
IC007C90IE	-0.197 ±0.031	-0.81 ±0.12	0.321 ±0.041	-3.80 ±0.41
P0798	-0.278 ±0.041	1.33 ±0.19	0.214 ±0.022	-1.91 ±0.20

borosilicate waste glass. The intrinsic rate constant is a fundamental materials property that depends only on glass composition and temperature. The difference in the intrinsic rate is consistent with the higher alumina and silica content and melting temperature of the ISV product compared with the waste glass. This does not necessarily mean that releases of encapsulated radionuclides will be two orders of magnitude lower with the ISV product (assuming equivalent waste loading). The radionuclide release rate is determined by many other factors including the chemistry of the groundwater contacting the solid.

5.2 STATIC TESTS

The ultimate goal of conducting static experiments with ISV products is to determine how well or how poorly a predictive model reproduces the data. This goal presents a considerable challenge with the ISV products since we have studied eight glasses with different composition and thermal history and have over 10,000 pieces of analytical data! It would be highly undesirable to have a unique set of thermodynamic data and secondary minerals associated with each material. In essence this would amount to a fitting exercise that defeats the purpose of having a mechanistic model. Consequently, the approach we have taken in attempting to model this large data set is to establish a consistent set of thermodynamic data and secondary minerals that reproduces the solution chemistry in each data set. To simplify the analysis further, the computer simulations were performed on three individual glasses. The ES4 composition was selected as representative of the composition from the engineering-scale tests; sample IC007C90IE was selected as representative of an amorphous solid from IFT 2 and sample IC027B90IE as representative of the devitrified product. Sample IC027B90IE was selected for the devitrified product because it was one of the most highly crystallized samples and also had the most complete static testing data set.

Figure 15. Paragenetic Sequence of Mineral Phases Predicted to Form in Static Tests With ES4 Glass

The first step that was performed was to enter the final solution composition from one of the static tests, in this case from ES-4, and run the EQ3NR code. The EQ3NR code calculates the charge imbalance and saturation indices for the minerals in its thermodynamic database based upon the input solution composition. The results from this calculation indicated that approximately 30 minerals and 7 solid solutions^a were highly supersaturated, and 7 minerals were near saturation. In most instances, the supersaturated phases were high temperature phases that could not form under the conditions of our experiments, or the thermodynamic data were acknowledged to be of poor quality. The one exception was a dioctahedral smectite "solid solution" that was found to be nearly two orders of magnitude supersaturated. Smectites are common alteration minerals in glass/water systems and would be expected to form in these experiments. The smectite solid solution in the EQ3NR database consists of a mixture of Ca, K, Na, and Mg beidellite, nontronite, and montmorillonite.

Reaction path simulations consist of "dissolving" a solid in 1 kg of water in discrete steps. At each step in reaction progress, saturation indices are checked with respect to a set of minerals in the thermodynamic database. Once a saturation index of 0 or greater is achieved for a mineral, precipitation is assumed to occur instantaneously. In this case, the supersaturated mineral phases identified from the EQ3NR run were not allowed to precipitate in the reaction path simulations performed with the EQ6 code. We also set equilibrium constraints with atmospheric pressure of CO₂ and O₂ because the experiments were performed in Teflon PFA containers that are permeable to atmospheric gases. The simulations were run to a maximum reaction progress of 3×10^{-3} mol/kg, consistent with maximum reaction progress achieved in the static tests. Initial comparisons of the predicted solution composition with the measurements indicated good agreement with the test data. However, the orthopyroxene solid solution was also suppressed although the phase was indicated approximately saturated from the EQ3NR run. The ferrosilite end-member is not stable under the oxidizing conditions of the experiments and the thermodynamic data was acknowledged to be of poor quality. In this manner then, a set of mineral phases were identified that achieved good agreement with the data from the static tests with ES4 glass. The predicted sequence of precipitation and relative amount of these phases is shown in Figure 15. The clinoptilolite solid solution is a mixture of Ca, K, Mg, and Na clinoptilolite end members. The epidote solid solution is a mixture of clinozoisite [Ca₂Al₃Si₃O₁₂(OH)] and epidote [Ca₂FeAl₂Si₃O₁₂(OH)]. As shown in Figure 9, the predicted concentrations of the major cations released from the glass as a function of reaction progress is in excellent agreement with the analytical data with the exception of the Ca concentrations, which are higher by about a factor of two. It should be stated here that none of the predicted mineral phases has been identified in our experiments. Programmatic funding was terminated before the surface analyses were completed.

^aA solid solution has a precise thermodynamic and structural meaning that is not consistent with the EQ3/6 documentation. A "solid solution" in the EQ3/6 package is implemented as a ideal mixing model of a set of end member phases.

The primary difference in chemical composition of samples ES4 and IC007C90IE is in the Ca content; the IFT sample having nearly three times more Ca by weight. A comparison of the element concentrations in Figures 9 and 11 shows that the differences in composition have a significant effect on the solution concentrations of some elements. In particular, the Ca concentrations are approximately three times higher with IC007C90IE. The difference is not directly related to higher Ca content of the IC007C90IE sample since solubility constraints are clearly affecting the solution concentrations in both experiments. In fact, the Al concentrations are lower by about a factor of two despite the similar Al content of the samples.

To understand the differences in the solution composition that occur with these glasses, the EQ6 code was run with the same set of mineral phase constraints as was done for the simulations with the ES4 glass, but the input glass composition was changed to correspond with the bulk composition of sample IC007C90IE. The results are illustrated in Figure 11. Calcium concentrations were predicted to increase relative to the ES4 glass simulations, although about 30% lower than the test data. The Al concentrations fell by approximately a factor of two in accordance with the experimental measurements. For this series of samples, at temperature pH values were available for direct comparison with the code predictions; these measurements agree with the simulations within about 0.5 pH unit at the highest values of reaction progress.

The differences in the solution composition between these two glasses can be understood from the changes in secondary minerals that form during the course of the dissolution process. The predicted sequence of secondary minerals to precipitate and relative mol% of each mineral are shown in Figure 16 for sample IC007C90IE. In the case of IC007C90IE, the Mg and Ca nontronites dissolve in favor of the more soluble epidote-ss which forms at three times the mol% of the ES4 simulation. This results in higher Ca concentrations but a decrease in Al concentration as this element is consumed in the precipitation of the epidote.

The final test of the simulation capabilities of the reaction path model was to test whether the effects of devitrification of the IFT samples could be simulated. As was discussed in Section 4.0 and shown in Figure 13, crystallization of IFT samples had a statistically significant effect on the solution concentrations of the major cations. To simulate this effect, we established the same constraints on mineral phase precipitation that were used for modeling the ES4 and IC007C90IE static test data with one exception. For sample IC027B90IE ($\approx 20\%$ crystallized by volume), we allowed diopside to precipitate. Diopside is the closest mineral present in the EQ3/6 database to simulate augite, which was identified by XRD and QEDS analysis. By allowing the diopside to precipitate, the resulting sequence of secondary minerals was changed from that of the compositionally-similar IC007C90IE sample as illustrated in Figure 17. In this case, the presence of the diopside stabilizes both Ca-nontronite and Ca-beidellite at the expense of the enstatite and epidote-ss.

Figure 16. Paragenetic Sequence of Mineral Phases Predicted to Form in Static Tests With IC007C90IE Glass

Figure 17. Paragenetic Sequence of Mineral Phases Predicted to Form in Static Tests With IC027B90IE Glass

Because the diopside is less soluble, peak Ca concentrations are reduced by over 1 mg/kg in the simulations (see Figure 12) which is smaller than the 3 mg/kg difference observed in the static tests but at least the trend is in the correct direction. Peak Al concentrations increase from about 3.5 to 4.5 mg/kg which is excellent agreement with the experimental data. Silicon concentrations are predicted to be smaller in the test with the devitrified sample relative to the amorphous sample by about 20%, again in excellent agreement with the trends observed in the experimental data.

In summary, a mineral phase assemblage has been established that reproduces the essential features of the solution chemistry observed in closed-system tests with a diverse range of ISV product samples of different elemental composition and thermal history. This is a significant accomplishment considering the uncertainties in the thermodynamic data and in the selection of appropriate mineral assemblages to control the solution chemistry. Improvements to the model can be obtained only by a thorough study of the alteration phases that form on the surfaces of these ISV products and by obtaining good thermodynamic data for these phases where they do not exist or are known to be unreliable. However, the present study establishes considerable confidence in the overall approach of using a geochemical model such as EQ3/6 to predict the long-term behavior of ISV products.

Although we have not attempted it here, there is sufficient information in this report to test the kinetic theory (Equation [11]) in predicting the reaction kinetics for sample IC007C90IE. Unfortunately, this is the only sample for which we have completed the necessary rate constant measurements, and then only at 90°C. The single-pass flow-through apparatus is well-suited for obtaining the necessary rate constants for a homogeneous solid. Interpretation of the data from this test for a devitrified ISV product is considerably more difficult. This is because the residual glass matrix and crystalline phase(s) are likely to have considerably different dissolution behavior compared with the amorphous solid. It may be necessary, for example, to study the dissolution behavior of selected clinopyroxene minerals separately to unambiguously determine the intrinsic rate constants that are needed to apply the kinetic theory.

6.0 CONCLUSION

A series of experiments was performed to determine the dissolution behavior of samples produced from the ISV processing of typical soils from the INEL subsurface disposal area. Dissolution rate constant measurements were completed at 90°C over the pH range 2 to 11 for one sample obtained from a field test of the ISV process. The results showed an intrinsic dissolution rate constant approximately two orders of magnitude smaller than measured on a typical borosilicate waste glass. Depending on pH, this translates to a dissolution rate between 5 to 100 times slower than a waste glass.

Extensive static testing of samples from pilot-scale tests and a field test of the ISV process were also completed. Devitrified samples from the field test showed marked differences in dissolution behavior from amorphous samples of equivalent bulk composition taken from different locations in the melt. Depending on the type and length of the test, different conclusions could be drawn as to the effect of devitrification on the durability of the product. However, using a consistent set of constraints on mineral precipitation, excellent agreement was obtained between the solution composition observed in the laboratory and the composition predicted with the EQ3/6 geochemical code. Differences in elemental releases among different samples were predicted to be the result of different secondary solids that form during the course of the reaction of the ISV product with water. Post-test characterization of the secondary phases formed in the laboratory tests is needed to validate the model predictions. Additional thermodynamic and kinetic data on the mineral augite will be required to adequately explain the differences in the dissolution kinetics of the partially-devitrified ISV products.

7.0 REFERENCES

- Aagaard, P., and H. C. Helgeson. 1982. "Thermodynamic and Kinetic Constraints on Reaction Rates Among Minerals and Aqueous Solutions. I. Theoretical Considerations." *Am. J. Sci.* 282:237-285.
- Advocat, T., J. L. Crovisier, B. Fritz, and E. Vernaz. 1990. "Thermokinetic Model of Borosilicate Glass Dissolution: Contextual Affinity." In Scientific Basis for Nuclear Waste Management XIII, Eds. V. M. Oversby and P. W. Brown, Materials Research Society, Pittsburgh, Pennsylvania.
- Arnorsson, S. 1975. "Application of the Silica Geothermometer in Low Temperature Hydrothermal Areas in Iceland." *Am. J. Sci.* 275:763-784.
- Bourcier, W. L., K. G. Knauss, and C. I. Merzbacher. 1989. "A Kinetic Model for the Dissolution of Borosilicate Glass." *Water-Rock Interaction* 107-110, Rotterdam.
- Bunker, B. C., G. W. Arnold, E. K. Beauchamp, and D. E. Day. 1983. "Mechanisms for Alkali Leaching in Mixed Na-K Silicate Glasses." *J. Non-Cryst. Solids* 58:295-322.
- Byers, C. D., M. J. Jercinovic, and R. C. Ewing. 1987. A Study of Natural Glass Analogues as Applied to Alteration of Nuclear Waste Glass. ANL-86-46, Argonne National Laboratory, Argonne, Illinois.
- Callow, R. A., L. E. Thompson, J. R. Weidner, C. A. Loehr, B. P. McGrail, and S. O. Bates. 1991. In Situ Vitrification Application to Buried Waste: Final Report of Intermediate Field Tests at Idaho National Engineering Laboratory. EGG-WTD-9807, Idaho National Engineering Laboratory, EG&G Idaho, Inc., Idaho Falls, Idaho.
- Chick, L. A., and G. F. Piepel. 1984. "Statistically Designed Optimization of a Glass Composition." *J. Am. Cer. Soc.* 67(11):763-768.
- Chick, L. A., and R. P. Turcotte. 1983. Glass Leaching Performance. PNL-4576, Pacific Northwest Laboratory, Richland, Washington.
- Deer, W. A., R. A. Howie, and J. Zussman. 1978. Rock-Forming Minerals: Single-Chain Silicates. John Wiley & Sons, New York, New York.
- Delage, F., and J. L. Dussossoy. 1991. "R7T7 Glass Initial Dissolution Rate Measurements Using a High-Temperature Soxhlet Device." In Scientific Basis for Nuclear Waste Management XIV, Materials Research Society, Pittsburgh, Pennsylvania.
- DuMouchel, W. 1987. A Data Analysis Advisor. Technical Report, BBN Software Products Corporation, Cambridge, Massachusetts

Eggleston, C. M., M. F. Hochella, and G. A. Parks. 1989. "Sample Preparation and Aging Effects on the Dissolution Rate and Surface Composition of Diopside." *Geochim. Cosmochim. Acta* 53:797-804.

Farnsworth, R. K., J. S. Tixier, J. W. Shade, and B. L. Charboneau. 1990. Engineering-Scale Test #4, PNL-7641, Pacific Northwest Laboratory, Richland, Washington.

Federal Register. 1988. Rules and Regulations. 51(216):40643-40652.

Grambow, B. 1985. "A General Rate Equation for Nuclear Waste Glass Corrosion." In Scientific Basis for Nuclear Waste Management VIII, eds. C. M. Jantzen, J. A. Stone, and R. C. Ewing, Materials Research Society, Pittsburgh, Pennsylvania.

Ishiguro, K., F. Kamei, N. Sasaki, and H. Nagaki. 1983. Long-Term Soxhlet Leach Tests of Simulated Waste Glasses(I). PNCT N831-83-01, Power Reactor and Nuclear Fuels Corporation, Japan.

Jantzen, C. M., and D. F. Bickford. 1985. "Leaching of Devitrified Glass Containing Simulated SRP Nuclear Waste." In Scientific Basis for Nuclear Waste Management VIII, Eds. C. M. Jantzen, J. A. Stone, and R. C. Ewing, Materials Research Society, Pittsburgh, Pennsylvania.

Jantzen, C. M., and N. E. Bibler. 1987. Product Consistency Test (PCT) for DWPF Glass: Part I. Test Development and Protocol, DPST-87-575, Savannah River Laboratory, Aiken, South Carolina.

Keller, J. F. 1990. "Regulatory Requirements for Ground-Water Monitoring Networks at Hazardous Waste Sites." In Environmental Monitoring, Restoration, and Assessment: What Have We Learned?, Proceedings of Twenty-Eighth Hanford Symposium on Health and the Environment, ed. R. H. Gray, Richland, Washington.

Knauss, K. G. et al. 1990. "Dissolution Kinetics of a Simple Analogue Nuclear Waste Glass as a Function of pH, Time and Temperature." In Scientific Basis for Nuclear Waste Management XIII, eds. V. M. Oversby and P. W. Brown, Materials Research Society, Pittsburgh, Pennsylvania.

Knauss, K. G., and T. J. Wolery. 1986. "Dependence of Albite Dissolution Kinetics on pH and Time at 25°C and 70°C." *Geochim. Cosmochim. Acta* 50:2481-2497.

Lasaga, A. C., and G. V. Gibbs. 1990. "Ab-Initio Quantum Mechanical Calculations of Water-Rock Interactions: Adsorption and Hydrolysis Reactions." *Am. J. Sci.* 290:263-295.

Levin, E. M., and H. F. McMurdie. 1975. Phase Diagrams for Ceramists - 1975 Supplement. American Ceramic Society, Columbus, Ohio.

- Macedo, P. B., A. Barkatt, and J. H. Simmons. 1982. "A Flow Model for the Kinetics of Dissolution of Nuclear Waste Glass." *Nucl. Chem. Waste Manage.* 3:13-21.
- McGrail, B. P. 1986. "Waste Package Component Interactions with Savannah River Defense Waste Glass in a Low-Magnesium Salt Brine." *Nuc. Tech.* 75(2):168-186.
- McGrail, B. P., and S. O. Bates. 1992. "Aqueous Dissolution of Laboratory and Field Samples From the In Situ Vitrification Process." In Proceedings of the VII International Conference on the Physics of Non-Crystalline Solids, Taylor & Francis Ltd., London, UK.
- Murphy, W. M., and H. C. Helgeson. 1987. "Thermodynamic and Kinetic Constraints on Reaction Rates Among Minerals and Aqueous Solutions. III. Activated Complexes and the pH-dependence of Rates of Feldspar, Pyroxene, Wollastonite, and Olivine Hydrolysis." *Geochim. Cosmochim. Acta* 51:3137-3153.
- Nickelson, D. F., J. Luey, and R. A. Callow. 1992. ISV Technology Development Plan for Buried Waste. EGG-WTD-10325. Idaho National Engineering Laboratory, EG&G Inc., Idaho Falls, Idaho.
- Oversby, V.M. 1984. The Nevada Nuclear Waste Storage Investigations Project Interim Acceptance Specifications for Defense Waste Processing Facility and West Valley Demonstration Project Waste Forms and Canisterized Waste. UCID-20165, Lawrence Livermore National Laboratory, Livermore, California.
- Parkhurst, D. L., D. C. Thorstenson, and L. N. Plummer. 1980. PHREEQE - A Computer Program for Geochemical Calculations. USGS/WRI-80-96, U.S. Geological Survey, Reston, Virginia.
- Pigford, T. H., J. O. Blomeke, T. L. Brekke, G. A. Cowan, W. E. Falconer, N. J. Grant, J. R. Johnson, J. M. Matusek, R. R. Parizek, R. L. Pigford, and D. E. White. 1983. A Study of the Isolation System for Geologic Disposal of Radioactive Wastes, National Academy Press, Washington, D.C.
- Pigford, T. H., P. L. Chambré, and W. W.-L. Lee. 1990. A Review of Near-Field Mass Transfer in Geologic Disposal Systems. LBL-27045, Lawrence Berkeley Laboratory, Berkeley, California.
- Pederson, L. R. 1987. "Comparison of Sodium Leaching Rates From a $\text{Na}_2\text{O} \cdot 3\text{SiO}_2$ Glass in H_2O and D_2O ." *Phys. Chem. Glasses* 28(1):17-21.
- Schott, J., R. A. Berner, and E. L. Sjöberg. 1981. "Mechanism of Pyroxene and Amphibole Weathering - I. Experimental Studies of Iron-Free Minerals." *Geochim. Cosmochim. Acta* 45:2123-2135.

Strachan, D. M., R. P. Turcotte, and B. O. Barnes. 1982. "MCC-1: Standard Leach Test for Nuclear Waste Forms." *Nucl. Tech.* 56:306-318.

Van Iseghem, P., and B. Grambow. 1988. "The Long-Term Corrosion and Modelling of Two Simulated Belgian Reference High-Level Waste Glasses." In Scientific Basis for Nuclear Waste Management XI, eds. M. J. Apted and R. E. Westerman, Materials Research Society, Pittsburgh, Pennsylvania.

Wolery, T. J. 1983. EQ3NR - A Computer Program for Geochemical Aqueous Speciation-Solubility Calculations: User's Guide and Documentation. UCRL-53414, Lawrence Livermore Laboratory, Livermore, California.

APPENDIX A

CALCULATION OF SPECIFIC SURFACE AREA

APPENDIX A

CALCULATION OF SPECIFIC SURFACE AREA

Glossary

ρ	= glass density (g/m^3)
m	= glass mass (g)
S	= surface area (m^2)
S_s	= specific surface area (m^2/g)
N	= number of particles
r	= mean radius of particles (m)

Assuming the ground glass can be approximated by a spherical particle, the mass and surface area are given by

$$m = \frac{4}{3}N\pi\rho r^3 \quad (\text{A.1})$$

$$S = 4N\pi r^2. \quad (\text{A.2})$$

Solving Equation A.1 for N gives

$$N = \frac{3m}{4\pi\rho r^3}. \quad (\text{A.3})$$

Substituting Equation A.3 into Equation A.2 and simplifying gives

$$S = \frac{3m}{\rho r}. \quad (\text{A.4})$$

Setting $m = 1$ in Equation A.4, the specific surface area is given by

$$S_s = \frac{3}{\rho r}. \quad (\text{A.5})$$

Table A.1 lists the nominal sieve openings for 100 to 400 mesh screen sizes. The average particle *diameter* for a -100 +200 mesh powder is therefore 0.1118 mm and for a -140 +200 mesh powder 0.0889 mm. The specific surface area for any glass powder can be calculated by substituting the appropriate r value and appropriate density for the glass in Equation A.5.

Table A.1. Nominal Openings for Standard Test Sieves

Sieve Mesh Size	Nominal Sieve Opening (mm)
100	0.1499
120	0.1245
140	0.1041
170	0.0889
200	0.0737
230	0.0635
270	0.0533
325	0.0432
400	0.0381

APPENDIX B

ANALYTICAL DATA FROM STATIC TESTS

APPENDIX B

ANALYTICAL DATA FROM STATIC TESTS

Table B1. Analytical Data From Static Tests at 90°C with ES-4 ISV Glass (mg/L)^a

Test Type	Duration (d)	S/V (m ⁻¹)	τ (d/m)	pH (25°C)	Al	Ba	Ca	K	Fe	Mg	Mn	Na	Si	Sr	Ti
MCC-1	7	10	70	6.23	0.01	0.001	0.057	0.011	-0.048	0.018		0.01	0.67	0.001	
MCC-1	14	10	140	8.49	0.10	0.004	0.120	0.720	-0.005	0.059	0.007	0.14	1.41	0.001	-0.001
MCC-1	14	10	140	7.72	0.07	0.002	0.140	0.530		0.048	0.006	0.18	1.42	0.001	-0.001
MCC-1	14	10	140	7.12	0.18	0.006	0.180	0.320	-0.005	0.059	0.006	0.12	1.69	0.002	-0.002
MCC-1	28	10	280	8.71	0.43	0.011	0.250	1.310	-0.004	0.120	0.011	0.15	3.12	0.002	-0.004
MCC-1	56	10	560	8.69	1.15	0.019	0.570	-0.332		0.222	0.017	0.16	6.79	0.005	-0.901
PCT	7	88	616	7.12	0.12	0.007	0.376	0.350	-0.048	0.181	0.015	0.20	3.73	0.004	0.001
PCT	7	88	616	7.01	0.07	0.005	0.356	0.369	-0.048	0.163	0.011	0.20	3.42	0.003	0.001
PCT	7	88	616	7.67	0.43	0.005	0.452	0.511	-0.047	0.201	0.009	0.20	4.43	0.004	0.001
PCT	7	88	616	7.82	0.62	0.007	0.476	0.347	-0.048	0.221	0.012	0.24	4.81	0.004	0.001
PCT	7	88	616	7.69	0.32	0.006	0.358	0.364	-0.049	0.170	0.012	0.17	3.78	0.003	0.001
PCT	7	88	616	7.60	0.36	0.007	0.442	0.394	-0.044	0.189	0.011	0.19	4.33	0.004	0.001
MCC-1	91	10	910	8.32	1.63	0.022	0.661	2.511	0.010	0.363	0.021	0.43	8.56	0.007	0.005
MCC-1	91	10	910	5.10	0.35	0.033	0.715	2.297	0.013	0.375	0.041	0.38	8.05	0.008	0.003
MCC-1	91	10	910	7.34	0.62	0.024	0.694	2.614	0.007	0.391	0.028	0.42	8.23	0.007	0.003
PCT	14	88	1232	7.69	0.75	0.012	0.631	0.593	-0.047	0.304	0.021	0.30	6.56	0.005	0.001
PCT	14	88	1232	8.00	1.25	0.013	0.807	0.675	-0.039	0.386	0.014	0.40	8.58	0.006	0.003

Table B1. Analytical Data From Static Tests at 90°C with ES-4 ISV Glass (mg/L)^a

Test Type	Duration (d)	S/V (m ⁻¹)	τ (d/m)	pH (25°C)	Al	Ba	Ca	K	Fe	Mg	Mn	Na	Si	Sr	Ti
PCT	28	88	2464	8.56	1.90	0.022	1.200	1.830	0.022	0.550	0.035	0.61	13.30	0.010	0.001
PCT	28	88	2464	8.00	2.82	0.015	1.510	1.110	0.201	0.720	0.023	0.77	17.00	0.011	0.022
PCT	28	88	2464	8.03	2.08	0.021	1.130	0.630	0.034	0.510	0.031	0.53	12.20	0.009	0.003
PCT	28	88	2464	8.64	2.79	0.022	1.300	1.250	0.003	0.590	0.028	0.64	14.90	0.010	0.022
PCT	28	88	2464	8.32	2.19	0.022	1.150	1.760	0.065	0.540	0.028	0.56	12.50	0.011	0.014
PCT	28	88	2464	8.06	1.65	0.020	1.050	0.960	0.024	0.480	0.030	0.51	11.40	0.009	0.002
PCT	56	88	4928	8.14	2.68	0.026	1.700	0.619	0.076	0.684	0.036	0.62	15.82	0.012	0.017
PCT	56	88	4928	8.01	3.18	0.020	1.566	0.342	0.118	0.704	0.021	0.69	17.62	0.012	0.031
PCT	7	880	6160	7.33	3.15	0.004	1.704	1.621	-0.024	0.883	0.008	1.22	19.16	0.009	0.003
PCT	7	880	6160	8.54	2.99	0.004	1.746	1.602	-0.004	0.899	0.007	1.23	19.25	0.010	0.003
PCT	7	880	6160	8.64	3.25	0.004	1.667	1.517	-0.033	0.862	0.010	1.19	18.88	0.009	0.002
PCT	91	88	8008	8.07	2.10	0.018	1.442	2.107	0.045	0.737	0.023	0.81	16.98	0.013	0.008
PCT	91	88	8008	8.11	3.13	0.022	1.542	1.695	0.170	0.770	0.028	0.75	17.19	0.013	0.039
PCT	91	88	8008	8.01	2.50	0.029	1.570	1.949	0.055	0.792	0.041	0.76	16.72	0.014	0.012
PCT	91	88	8008	5.44	2.55	0.090	2.508	2.852	0.112	1.183	0.127	1.29	24.35	0.023	0.003
PCT	91	88	8008	8.32	3.52	0.025	1.557	1.751	0.157	0.786	0.024	0.76	17.64	0.014	0.044
PCT	91	88	8008	8.31	3.40	0.024	1.549	2.345	0.263	0.820	0.030	0.78	17.23	0.014	0.061
PCT	14	880	12320	8.62	3.44	0.007	1.954	1.744	-0.027	0.969	0.007	1.37	21.00	0.011	0.003
PCT	28	880	24640	7.74	4.28	0.006	1.990	2.490	0.086	0.940	0.007	1.52	24.60	0.012	0.010
PCT	28	880	24640	8.05	4.28	0.007	2.160	2.160	0.027	1.050	0.005	1.49	24.90	0.012	0.002

Table B1. Analytical Data From Static Tests at 90°C with ES-4 ISV Glass (mg/L)^a

Test Type	Duration (d)	S/V (m ⁻¹)	τ (d/m)	pH (25°C)	Al	Ba	Ca	K	Fe	Mg	Mn	Na	Si	Sr	Ti
PCT	28	880	24640	8.06	4.14	0.006	2.210	2.360	0.023	1.040	0.011	1.50	23.90	0.012	0.002
PCT	56	880	49280	7.87	3.92	0.005	1.803	1.043	0.028	0.911	0.005	1.41	22.82	0.011	0.002
PCT	91	880	80080	8.23	3.96	0.006	1.946	2.939	0.081	1.105	0.009	1.71	23.17	0.012	0.008
PCT	91	880	80080	8.25	4.08	0.008	1.756	3.113	0.132	0.998	0.012	1.57	22.22	0.011	0.032
PCT	91	880	80080	7.92	3.28	0.007	1.898	2.749	0.033	1.094	0.014	1.79	22.73	0.012	0.006

^aNegative numbers indicate concentrations at or below analytical detection limit.

Table B2. Analytical Data From Static Tests at 90°C with Sample IC007C90IE (mg/L)

Test Type	Duration (d)	S/V (m ⁻¹)	τ (d/m)	pH (25°C)	Al	Ba	Ca	K	Fe	Mg	Mn	Na	Si	Sr	Ti
MCC-1	7	10	70	8.70	2.582	0.016	3.462	0.659	0.121	0.663	0.006	0.45	12.44	0.012	0.063
MCC-1	7	10	70	8.57	0.296	0.004	0.399	-0.705	0.003	0.019	0.002	0.09	1.761	0.002	-0.007
MCC-1	14	10	140	8.60	3.124	0.019	4.811	1.493	0.288	0.735	0.007	0.56	15.52	0.014	0.010
MCC-1	14	10	140	8.72	2.873	0.018	3.960	1.323	0.210	0.705	0.008	0.48	14.06	0.013	0.012
MCC-1	14	10	140	8.75	3.029	0.018	4.590	0.959	0.203	0.749	0.008	0.52	14.83	0.014	0.013
MCC-1	28	10	280		2.926	0.010	4.713	1.074	0.097	0.610	0.004	0.56	15.83	0.014	0.006
MCC-1	28	10	280	8.45	2.783	0.017	4.243	0.995	0.133	0.694	0.006	0.48	13.74	0.013	0.007
MCC-1	28	10	280		2.850	0.009	4.847	2.260	0.069	0.520	0.003	0.61	16.15	0.014	0.006
MCC-1	56	10	560	7.72	3.131	0.015	5.389	2.333	0.082	0.350	0.002	0.04	17.99	0.016	0.003
MCC-1	91	10	910	8.17	3.178	0.015	5.644	2.913	0.190	0.319	0.001	0.75	18.39	0.019	0.007
MCC-1	91	10	910	8.38	3.294	0.014	6.318	2.602	0.114	0.176	-0.001	0.80	19.80	0.019	0.004
MCC-1	91	10	910	8.31	3.070	0.013	5.460	2.832	0.121	0.240	-0.001	0.68	18.14	0.017	0.004
PCT	7	800	5600	8.80	3.160	0.010	6.002	2.193	0.065	0.183	0.001	1.37	20.71	0.017	0.044
PCT	7	800	5600	8.75	3.247	0.011	6.224	2.714	0.106	0.171	0.003	1.23	21.17	0.017	0.103
PCT	7	800	5600	8.79	3.087	0.009	5.765	2.354	0.035	0.129	0.001	1.20	20.55	0.017	0.016
PCT	28	209	5850		2.606	0.006	5.873	2.969	0.014	0.086	0.001	1.46	20.50	0.015	0.002
PCT	28	213	5976		2.697	0.005	6.280	2.076	0.014	0.044	0.001	1.51	21.42	0.015	0.001
PCT	14	800	11200	8.62	3.133	0.008	6.541	2.039	0.015	0.038		1.39	22.35	0.017	0.001
PCT	28	800	22400	8.60	2.955	0.006	6.581	2.782	0.078	0.029		1.52	22.09	0.017	
PCT	28	800	22400	8.53	2.930	0.006	6.520	1.840	0.080			1.51	21.70	0.017	

Table B2. Analytical Data From Static Tests at 90°C with Sample IC007C90IE (mg/L)

Test Type	Duration (d)	S/N (m ⁻¹)	τ (d/m)	pH (25°C)	Al	Ba	Ca	K	Fe	Mg	Mn	Na	Si	Sr	Ti
PCT	28	800	22400	8.72	2.890	0.006	6.001	18.5	0.100			1.49	21.5	0.016	
PCT	56	800	44800	8.67	2.847	0.009	6.640	3.429	0.158	0.165	0.004	9.99	22.00	0.018	0.008
PCT	91	800	72800	8.42	2.467	0.003	6.224	2.168	0.094	-0.028	-0.001	1.80	21.49	0.016	
PCT	91	800	72800	8.39	2.479	0.003	6.255	1.875	0.100	-0.024	-0.001	1.74	21.60	0.016	-0.001
PCT	91	800	72800	8.31	2.308	0.003	6.158	1.836	0.094	-0.021		1.73	21.11	0.015	-0.001

Table B3. Analytical Data From Static Tests at 90°C with IC027B90IE ISV Glass (mg/L)

Test Type	Duration (d)	SN/ (m ⁻¹)	τ (d/m)	pH (25°C)	Al	Ba	Ca	K	Fe	Mg	Mn	Na	Si	Ti
MCC-1	14	10	140	8.15	1.009	0.012	0.753	0.085	0.027	0.052	0.005	0.19	4.774	0
MCC-1	14	10	140	8.05	0.902	0.014	0.995	0.388	0.018	0.064	0.013	0.19	4.185	0
MCC-1	14	10	140	7.64	1.021	0.013	1.07	0.619	0.024	0.103	0.011	0.19	4.654	0.001
MCC-1	28	10	279.96		0.793	0.015	1.035	1.695	0.007	0.093	0.017	0.28	5.562	0.001
MCC-1	28	10	280	7.85	1.482	0.016	0.285	0.485	0.07	0.059	0.007	0.31	7.873	0
MCC-1	28	10	280.11		1.703	0.014	1.444	1.591	0.018	0.115	0.017	0.38	8.355	0.002
MCC-1	56	10	560	7.29	2.597	0.026	1.744	2.244	0.087	0.138	0.01	0.7	11.41	0.007
MCC-1	91	10	910	7.94	2.838	0.026	1.982	1.911	0.1	0.087	0.009	0.49	12.63	0.006
MCC-1	91	10	910	7.85	2.107	0.024	1.815	2.114	0.068	0.129	0.009	0.38	10.84	0.003
MCC-1	91	10	910	7.57	2.744	0.026	1.877	2.236	0.083	0.051	0.009	0.46	12.3	0.006
PCT	7	800	5600	7.88	3.911	0.009	2.566	1.25	0.019	0.133	0.005	1.18	17.8	0.007
PCT	7	800	5600	7.88	4.202	0.01	3.01	0.751	0.016	0.114	0.007	1.21	18.93	0.001
PCT	7	800	5600	7.88	3.852	0.012	2.758	1.549	0.023	0.105	0.015	1.17	17.4	0.012
PCT	28	218.49	6117.8		3.669	0.006	3.045	2.28	0.022	0.161	0.013	1.67	19.28	0.002
PCT	28	234.3	6560.5		3.999	0.005	2.996	1.982	0.021	0.164	0.009	1.64	19.85	0.003
PCT	14	800	11200	8.1	4.222	0.008	2.79	1.487	0.032	0.115	0.004	1.39	19.54	0.008
PCT	28	800	22400	7.85	4.37	0.006	3.03	1.96	0.08	0.1	0.009	1.6	20.5	
PCT	28	800	22400	7.83	4.142	0.009	2.915	1.801	0.091	0.1	0.005	1.57	19.78	0
PCT	28	800	22400	7.93	4.201	0.007	2.683	1.985	0.953	0.104	0.003	1.64	19.81	0.001
PCT	56	800	44800	7.98	4.144	0.008	2.743	2.843	0.106	0.155	0.005	1.73	20.14	0.004

Table B3. Analytical Data From Static Tests at 90°C with IC027B90IE ISV Glass (mg/L)

Test Type	Duration (d)	S/V (m ⁻¹)	τ (d/m)	pH (25°C)	Al	Ba	Ca	K	Fe	Mg	Mn	Na	Si	Ti
PCT	91	800	72800	7.89	4.148	0.008	2.96	1.847	0.123	0.127	0.007	1.95	20.2	0.003
PCT	91	800	72800	7.73	4.188	0.007	2.974	2.135	0.104	0.123	0.007	1.98	20.38	0.002
PCT	91	800	72800	7.87	4.093	0.006	2.951	2.357	0.087	0.136	0.006	2.02	20.19	0.002

Table B4. Analytical Data From Static Tests at 90°C with IC038B90IE ISV Glass (mg/L)

Test Type	Duration (d)	S/V (m ⁻¹)	τ (d/m)	pH (25°C)	Al	Ba	Ca	K	Fe	Mg	Mn	Na	Si	Sr	Ti
MCC-1	28	10	279.92	8.48	3.054	0.014	3.626	2.101	0.094	0.661	0.016	0.49	14.65	0.014	0.009
MCC-1	28	10	280.08	8.51	3.058	0.014	3.733	2.355	0.102	0.764	0.017	0.52	14.67	0.014	0.01
PCT	28	213.44	5976.3	9.02	3.054	0.005	4.295	1.991	0.015	0.184	0.001	1.47	19.87	0.013	0.002
PCT	28	235.93	6606.1	8.63	3.067	0.005	4.457	1.812	0.016	0.386	0.002	1.49	19.34	0.014	0.001

Table B5. Analytical Data From Static Tests at 90°C with IC038C90IE ISV Glass (mg/L)

Test Type	Duration (d)	SV (m ³)	τ (d/m)	pH (25°C)	Al	Ba	Ca	K	Fe	Mg	Mn	Na	Si	Sr	Ti
MCC-1	7	10	70	7.72	0.613	0.009	0.675	0.023	0.008	0.085	0.009	0.10	2.913	0.003	0.003
MCC-1	14	10	140	7.80	1.091	0.013	1.155	0.862	0.034	0.122	0.006	0.21	5.133	0.006	0.001
MCC-1	14	10	140	7.99	0.992	0.017	1.091	0.728	0.021	0.130	0.018	0.20	4.73	0.006	0.001
MCC-1	14	10	140	7.72	1.287	0.016	1.433	0.461	0.033	0.142	0.015	0.25	5.948	0.007	0.001
MCC-1	28	10	280	7.94	2.137	0.023	1.942	0.850	0.057	0.161	0.019	0.36	9.649	0.011	0.002
MCC-1	28	10	280		0.094	0.012	1.078	1.512	0.001	0.212	0.023	0.20	4.204	0.005	0.001
MCC-1	28	10	280		1.423	0.015	1.552	1.655	0.016	0.182	0.020	0.29	7.644	0.009	0.001
MCC-1	56	10	560	7.77	2.361	0.025	1.897	2.078	0.081	0.086	0.014	0.50	10.78	0.012	0.006
MCC-1	91	10	910	7.82	2.817	0.025	2.312	2.652	0.103	0.181	0.013	0.69	12.77	0.014	0.006
MCC-1	91	10	910	7.88	2.892	0.023	2.559	2.291	0.097	0.215	0.012	0.65	13.39	0.015	0.005
MCC-1	91	10	910	7.26	0.591	0.031	2.710	2.305	0.04	0.369	0.047	0.49	9.999	0.012	0.003
PCT	7	800	5600	8.4	3.925	0.009	3.942	1.035	0.024	0.466	0.008	1.39	19.38	0.016	0.009
PCT	7	800	5600	8.4	3.937	0.010	3.721	1.211	0.052	0.424	0.007	1.34	19.09	0.016	0.043
PCT	7	800	5600	8.48	3.820	0.011	3.319	2.200	0.090	0.391	0.006	1.35	18.59	0.016	0.087
PCT	28	232.26	6503.3		3.546	0.005	3.623	1.404	0.041	0.367	0.003	1.84	19.34	0.014	0.002
PCT	28	238.56	6679.8		3.695	0.005	3.750	1.268	0.028	0.351	0.003	1.82	19.86	0.014	0.002
PCT	14	800	11200	8.52	3.987	0.007	3.629	1.035	0.057	0.338	0.001	1.59	20.27	0.015	0.011
PCT	28	800	22400	8.01	4.053	0.007	4.082	1.740	0.086	0.362	0.002	1.90	20.89	0.016	0.001
PCT	28	800	22400	8.14	3.963	0.006	3.757	1.532	0.125	0.354	0.002	1.85	20.57	0.015	0.001
PCT	28	800	22400	8.21	3.980	0.006	4.036	1.422	0.088	0.373	0.003	1.82	20.53	0.016	0.001

Table B5. Analytical Data From Static Tests at 90°C with IC038C90IE ISV Glass (mg/L)

Test Type	Duration (d)	S/V (m ⁻¹)	τ (d/m)	pH (25°C)	Al	Ba	Ca	K	Fe	Mg	Mn	Na	Si	Sr	Ti
PCT	56	800	44800	8.07	3.713	0.008	3.701	3.251	0.109	0.430	0.003	1.82	19.92	0.015	0.005
PCT	91	800	72800	7.86	3.563	0.006	3.938	2.344	0.080	0.395	0.002	2.39	20.08	0.015	0.002
PCT	91	800	72800	7.91	3.556	0.005	3.833	2.161	0.077	0.356	0.002	2.41	19.99	0.015	0.001
PCT	91	800	72800	8.07	3.622	0.005	3.862	1.506	0.087	0.339	0.002	2.30	19.98	0.015	0.002

Table B6. Analytical Data from Static Tests at 90°C with IC038D90IE ISV Glass (mg/L)

Test Type	Duration (d)	S/V (m ⁻¹)	τ (d/m)	pH (25°C)	Al	Ba	Ca	K	Fe	Mg	Mn	Na	Si	Sr	Ti
MCC-1	28	10	280.02	8.02	0.938	0.015	1.149	0.828	0.007	0.091	0.016	0.30	6.365	0.007	0.001
MCC-1	28	10	280.09	8.03	0.887	0.012	0.877	1.926	0.022	0.117	0.011	0.35	6.768	0.006	0.004
PCT	28	224	6271.2	8.61	3.925	0.007	2.789	1.736	0.017	0.135	0.006	1.52	19.16	0.014	0.002
PCT	28	241	6744.9	8.71	3.911	0.006	2.957	1.929	0.015	0.147	0.011	1.63	19.49	0.014	0.002

Table B7. Analytical Data From Static Tests at 90°C with IC048B90IW ISV Glass (mg/L)

Test Type	Duration (d)	S/V (m ⁻¹)	τ (d/m)	pH (25°C)	Al	Ba	Ca	K	Fe	Mg	Mn	Na	Si	Sr	Ti
MCC-1	7	10	70	7.77	0.864	0.008	0.840	0.690	0.026	0.207	0.003	0.20	4.288	0.004	0.012
MCC-1	14	10	140	7.92	2.211	0.016	2.270	0.971	0.118	0.545	0.012	0.35	10.04	0.009	0.010
MCC-1	14	10	140	8.31	2.244	0.014	2.108	0.668	0.119	0.475	0.008	0.37	10.23	0.009	0.007
MCC-1	14	10	140	7.29	0.218	0.008	0.382	-0.182	0.020	0.075	0.006	0.08	1.44	0.002	
MCC-1	28	10	280		2.962	0.014	2.738	2.904	0.124	0.720	0.014	0.52	13.70	0.012	0.012
MCC-1	28	10	280		2.914	0.014	2.710	2.522	0.129	0.706	0.014	0.51	13.45	0.012	0.013
MCC-1	28	10	280	7.84	2.505	0.019	2.269	1.129	0.118	0.520	0.014	0.42	11.64	0.011	0.007
MCC-1	56	10	560	8.01	3.305	0.020	3.163	2.193	0.129	0.801	0.013	0.47	15.47	0.013	0.008
MCC-1	91	10	910	7.93	3.298	0.018	3.037	2.166	0.152	0.688	0.01	0.64	15.52	0.013	0.007
MCC-1	91	10	910	8.15	3.735	0.018	3.698	2.583	0.185	0.885	0.011	0.68	17.37	0.015	0.013
MCC-1	91	10	910	8.08	3.417	0.018	3.181	2.263	0.180	0.763	0.01	0.60	15.88	0.014	0.010
PCT	28	174	4877		3.365	0.005	3.870	2.048	0.020	0.592	0.002	1.50	19.95	0.017	0.003
PCT	28	176	4916		3.171	0.006	3.569	2.325	0.019	0.398	0.001	1.44	19.37	0.012	0.003
PCT	7	800	5600	8.50	3.909	0.008	3.808	1.763	0.018	0.838	0.002	1.14	19.67	0.014	0.012
PCT	7	800	5600	8.64	3.819	0.007	3.617	0.008	0.045	0.851	0.001	1.08	19.20	0.013	-0.007
PCT	7	800	5600	8.50	3.895	0.008	3.667	2.009	0.045	0.827	0.001	1.15	19.50	0.014	0.011
PCT	14	800	11200	8.40	3.760	0.007	3.944	1.296	0.030	0.575	0.001	1.32	20.43	0.014	0.020
PCT	28	800	22400	8.13	3.680	0.005	4.040	1.777	0.078	0.637	0.001	1.48	20.47	0.013	0.001
PCT	28	800	22400	8.36	3.543	0.005	3.575	1.875	0.153	0.460		1.42	20.26	0.013	0.001
PCT	28	800	22400	8.31	3.764	0.005	4.077	1.765	0.077	0.583	0.001	1.46	20.66	0.013	0.001

Table B7. Analytical Data From Static Tests at 90°C with IC048B90IW ISV Glass (mg/L)

Test Type	Duration (d)	S/V (m ⁻¹)	τ (d/m)	pH (25°C)	Al	Ba	Ca	K	Fe	Mg	Mn	Na	Si	Sr	Ti
PCT	56	800	44800	8.10	3.437	0.007	4.187	3.009	0.080	0.430	0.002	2.28	21.27	0.013	0.004
PCT	91	800	72800	8.13	3.173	0.005	4.038	1.964	0.074	0.260	-0.001	1.83	21.24	0.013	
PCT	91	800	72800	8.04	2.971	0.004	4.112	3.038	0.074	0.359	-0.001	1.92	21.69	0.013	-0.001
PCT	91	800	72800	7.86	2.967	0.013	3.688	11.59	0.127	0.904	0.008	2.12	19.48	0.015	0.024

Table B8. Analytical Data From Static Tests at 90°C with IC048C90IW ISv Glass (mg/L)

Test Type	Duration (d)	S/N (m ⁻¹)	τ (d/m)	pH (25°C)	Al	Ba	Ca	K	Fe	Mg	Mn	Na	Si	Sr	Ti
MCC-1	28	10	280	8.62	2.840	0.014	2.535	2.315	0.091	0.639	0.014	0.50	13.24	0.012	0.010
MCC-1	28	10	280	8.20	1.282	0.015	2.279	1.989	0.114	0.600	0.018	0.44	10.89	0.010	0.005
PCT	28	226	6336	8.44	3.369	0.005	3.611	2.981	0.029	0.495	0.002	1.53	19.80	0.012	0.004
PCT	28	247	6911	8.18	3.392	0.005	3.661	2.412	0.021	0.512	0.001	1.73	19.83	0.012	0.003

Distribution

**No. of
Copies**

**No. of
Copies**

OFFSITE

12 DOE/Office of Scientific and
Technical Information

M. W. Shupe
DOE/Office of Technology
Development
EM-541, Trevion II Building
12800 Middlebrook Road
Germantown, MD 20874

T. D. Anderson
DOE/Office of Technology
Development
EM-551, Trevion II Building
12800 Middlebrook Road
Germantown, MD 20874

D. L. Biancosino
DOE/Office of Technology
Development
EM-551, Trevion II Building
12800 Middlebrook Road
Germantown, MD 20874

G. E. Voelker
DOE/Office of Technology
Development
EM-50, Trevion II Building
12800 Middlebrook Road
Germantown, MD 20874

K. E. Hain
DOE/Office of Technology
Development
EM-55, Trevion II Building
12800 Middlebrook Road
Germantown, MD 20874

S. T. Lien
DOE/Office of Technology
Development
EM-54, Trevion II Building
12800 Middlebrook Road
Germantown, MD 20874

S. M. Prestwich
DOE/Office of Technology
Development
EM-52, Trevion II Building
12800 Middlebrook Road
Germantown, MD 20874

J. E. Baublitz
DOE/Office of Environmental
Restoration
EM-40, Trevion II Building
12800 Middlebrook Road
Germantown, MD 20874

L. H. Taylor
DOE/Office of Environmental
Restoration
EM-43, Trevion II Building
12800 Middlebrook Road
Germantown, MD 20874

**No. of
Copies**

J. J. Fiore
DOE/Office of Environmental
Restoration
EM-42, Trevion II Building
12800 Middlebrook Road
Germantown, MD 20874

J. C. Lehr
DOE/Office of Environmental
Restoration
EM-44, Trevion II Building
12800 Middlebrook Road
Germantown, MD 20874

S. A. Mann
DOE/Office of Environmental
Restoration
EM-44, Trevion II Building
12800 Middlebrook Road
Germantown, MD 20874

W. Bixby
DOE/Office of Restoration &
Waste Management
EM-60, Forrestal Building
U.S. Department of Energy
1000 Independence Avenue SW
Washington, DC 20585

S. P. Cowan
DOE/Office of Waste
Management
EM-30, Trevion II Building
12800 Middlebrook Road
Germantown, MD 20874

J. E. Lytle
DOE/Office of Waste Operations
EM-30, Forrestal Building
U.S. Department of Energy
1000 Independence Avenue SW
Washington, DC 20585

**No. of
Copies**

C. W. Frank
DOE/Office of Tech. Dev.
EM-50, Forrestal Building
U.S. Department of Energy
1000 Independence Avenue SW
Washington, DC 20585

R. P. Whitfield
DOE/Office of Env. Rest.
EM-40, Forrestal Building
U.S. Department of Energy
1000 Independence Avenue SW
Washington, DC 20585

T. J. Atterbury
Battelle Columbus
505 King Avenue
Columbus, Ohio 43201-2693

D. Berry
Sandia National Laboratories
Department 6620
Albuquerque, NM 87185-5800

D. H. Brandy
U.S. Department of Energy
Albuquerque Field Office
P.O. Box 5400
Albuquerque, NM 87115

B. E. Campbell
Geosafe Corporation
2950 George Washington
Way #A
Richland, WA 99352-1615

Peter Colombo
Brookhaven National Laboratory
Building 703-50 Rutherford
Upton, NY 11973

**No. of
Copies**

J. Corones
AMES Laboratory
329 Wilhelm Hall
Iowa State University
Ames, IA 50011

B. Crary
HAZWRAP
P.O. Box 2003, MS 7606
Oak Ridge, TN 37831-7606

N. W. Dunbar
NMBMMR
New Mexico Institute of Technology
Socorro, NM 87801

J. Eckwert
Sandia National Laboratories
P.O. Box 5800, Division 6621
Albuquerque, NM 87185-5800

W. Fitch
U.S. Department of Energy
Idaho Field Office
785 DOE Place
Idaho Falls, ID 83402

D. R. Gibby
Martin-Marietta Energy Systems
K-25 Environmental Management
Division
K-25 Plant, Building 0303-8
Oak Ridge, TN 37831-7314

B. Haas
Ames Laboratory
7 Spedding Hall
Iowa State University
Ames, IA 50011

**No. of
Copies**

J. E. Hanson
Geosafe Corporation
2950 George Washington
Way #A
Richland, WA 99352-1615

J. S. Haselow
Westinghouse Savannah River
Company
P.O. Box 616
Aiken, SC 29801

J. C. Haugen
U.S. Department of Energy
Chicago Field Office
9800 South Cass Avenue
Argonne, IL 60439-4837

M. D. Herd
EG&G Idaho
P.O. Box 1625
Idaho Falls, ID 83415

A. C. Heywood
P.O. Box 808
7000 East Avenue
MS L-207
Livermore, CA 94550

G. H. Hewig
Battelle Europe
Am Romerhof 35
D 6000 Frankfurt/Main 90

W. Holman
U.S. Department of Energy
San Francisco Field Office
1333 Broadway
Oakland, CA 94612

**No. of
Copies**

4 G. K. Jacobs
 Oak Ridge National Laboratory
 P.O. Box 2008
 Oak Ridge, TN 37831-6036

 C. M. Jantzen
 Westinghouse Savannah River
 Company
 P.O. Box 616
 Aiken, SC 29801

 R. Knowlton
 Sandia National Laboratories
 Department 6620
 Albuquerque, NM 87185-5800

 J. Koger
 Martin Marietta Energy Systems
 P.O. Box 2009
 Oak Ridge, TN 37831-8097

 P. E. MacDonald
 EG&G Idaho
 P.O. Box 1625
 Idaho Falls, ID 83415

 W. J. Madia
 Battelle Columbus
 505 King Avenue
 Columbus, Ohio 43201-2693

 B. G. Maiden
 Battelle Columbus
 505 King Avenue
 Columbus, Ohio 43201-2693

 H. Maier
 Battelle Europe
 Am Romerhof 35
 D 6000 Frankfurt/Main 90

**No. of
Copies**

12 R. K. Major
 Nuclear Regulatory Commission
 Advisory Committee on Nuclear
 Waste
 Mail Stop P-315
 Washington, DC 20555

 A. Malinauskas
 Oak Ridge National Laboratory
 P.O. Box 2008, MS 6135
 Oak Ridge, TN 37831-6135

 S. K. Merrill
 EG&G Idaho
 P.O. Box 1625
 Idaho Falls, ID 83415

2 J. Moore
 U.S. Department of Energy
 Oak Ridge Field Office
 P.O. Box E
 Oak Ridge, TN 37831

 H. D. Murphy
 Los Alamos National Laboratory
 P.O. Box 1663
 Los Alamos, NM 87545

2 M. Naney
 Oak Ridge National Laboratory
 P.O. Box 2008
 Oak Ridge, TN 37831-6036

5 D. F. Nickelson
 EG&G Idaho
 P.O. Box 1625
 Idaho Falls, ID 83415

**No. of
Copies**

R. L. Norlund
EG&G Idaho
P.O. Box 1625
Idaho Falls, ID 83415

K. Nuhfer
Environmental Management
Westinghouse Materials Co.
of Ohio
P.O. Box 398704
Cincinnati, OH 45239-8704

M. G. O'Rear
U.S. Department of Energy
Savannah River Field Office
RFD #1, Bldg. 703A,
Rm E208 North
P.O. Box A
Aiken, SC 29802

P. T. Owen
Martin Marietta Energy Systems
P.O. Box 2003
Oak Ridge, TN 37830-7256

J. L. Peterson
EG&G Rocky Flats, Inc.
P.O. Box 464
Golden, CO 80402-0464

J. Phelan
Sandia National Laboratories
Department 6620
Albuquerque, NM 87185-5800

J. M. Pope
West Valley Nuclear Services
Company
P.O. Box 191
West Valley, NY 14171

**No. of
Copies**

M.A.H. Reimus
Los Alamos National Laboratory
P.O. Box 1663
Los Alamos, NM 87545

L. Rogers
EG&G Energy Measurements,
Inc.
P.O. Box 1912, MS RSL-11
Las Vegas, NV 89125

P. A. Saxman
U.S. Department of Energy
Albuquerque Field Office
P.O. Box 5400
Albuquerque, NM 87115

R. F. Smiecinski
U.S. Department of Energy
Nevada Field Office
P.O. Box 98518
Las Vegas, NV 89109

2 B. P. Spalding
Oak Ridge National Laboratory
P.O. Box 2008
Oak Ridge, TN 37831-6036

J. F. Sproull
Westinghouse Savannah River
Company
P.O. Box 616
Aiken, SC 29801

J. L. Steele
Savannah River Site
SRL, 773 A, A208
Aiken, SC 29802

**No. of
Copies**

R. R. Stiger
EG&G Idaho
P.O. Box 1625
Idaho Falls, ID 83415

2 J. Sweeney
U.S. Department of Energy
Oak Ridge Field Office
P.O. Box E
Oak Ridge, TN 37831

Technical Library
Sandia National Laboratories
Department 6620
Albuquerque, NM 87185-5800

Technical Library
Battelle Columbus
505 King Avenue
Columbus, Ohio 43201-2693

L. Tyler
Sandia National Laboratories
Department 6620
Albuquerque, NM 87185-5800

R. Tyler
U.S. Department of Energy
Rocky Flats Field Office
DOE Building 1166
Golden, CO 80402-0464

2 C. L. Timmerman
Geosafe Corporation
2950 George Washington
Way #A
Richland, WA 99352-1615

**No. of
Copies**

T. L. Walton
Westinghouse Savannah River
Company
P.O. Box 616
Aiken, SC 29801

J. L. Yow, Jr.
Lawrence Livermore National
Laboratory
7000 East Avenue
Livermore, CA 94550

ONSITE

5 DOE Richland Field Office

E. A. Bracken	A6-95
J. K. Erickson	A5-19
R. D. Freeberg	A5-19
J. D. Goodenough	A5-19
D. E. Trader	A5-90

23 Westinghouse Hanford Company

M. R. Adams	H4-55
W. C. Alaconis	L4-92
H. Babad	R2-08
J. D. Berger	LO-18
J. W. Cammann	H4-14
K. R. Fecht	H4-56
R. L. Gilchrist	L5-63
W. E. Hamilton	L6-78
H. D. Harmon	R2-52
J. O. Honeyman	B1-31
K. N. Jordan	L4-88
C. H. Kindle	H4-55
M. R. Korenko	B3-08

**No. of
Copies**

R. E. Lerch	B2-35
R. M. Marusich	H5-32
M. M. McCarthy	N3-13
H. E. McGuire	B3-63
J. W. Shade	R4-03
T. M. Wintezak	L4-92
D. D. Wodrich	L6-27
R. D. Wojtasek	L4-92
B. A. Wolfe	G6-02
J. G. Woolard	H4-55

52 **Pacific Northwest Laboratory**

L.M.G. Ballou	P7-18
E. G. Baker	P8-38
W. F. Bonner	P7-41
K. A. Borgeson	P7-41
G. H. Bryan	P7-25
J. L. Buelt	P7-41
H. C. Burkholder	P7-41
C. C. Chapman	P7-41
R. E. Einziger	P7-14
R. W. Forester	K6-88
P. R. Hrma	P8-37

**No. of
Copies**

D. W. Koppenaal	P7-07
W. L. Kuhn	P8-38
N. J. Lombardo	K7-02
P. S. Lowery	K7-15
J. Luey	P7-34 (3)
S. C. Marschman	P7-18
J. L. McElroy	P7-46
B. P. McGrail	K2-38 (10)
K. M. Olson	P7-14 (3)
M. E. Peterson	P7-41
T. D. Powell	P7-34
R. L. Skaggs	K6-77
S. C. Slate	K1-19
S. L. Stein	BSRC
D. M. Strachan	K2-38
R. B. Stults	K2-20
M. T. Thomas	R1-03
L. E. Thompson	P7-34
J. S. Tixier	P7-34
K. D. Weimers	P7-14
C. L. Widrig	K1-21
S. L. Woosley	P7-34
Publishing Coordination	
Technical Report Files	(5)

**DATE
FILMED**

4 / 20 / 93