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ANL-94/34

The Effects of the Glass Surface Area/Solution Volume Ratio on Glass Corrosion: A Critical Review

by W. L. Ebert

Argonne National Laboratory, Argonne, Illinois 60439
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ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois 60439

**THE EFFECTS OF THE GLASS SURFACE AREA/SOLUTION
VOLUME RATIO ON GLASS CORROSION:
A CRITICAL REVIEW**

by

William L. Ebert

Chemical Technology Division

March 1995

MASTER

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VOLUME RATIO ON GLASS CORROSION:
A CRITICAL REVIEW**

William L. Ebert

ABSTRACT

This report reviews and summarizes the present state of knowledge regarding the effects of the glass surface area/solution volume (SA/V) ratio on the corrosion behavior of borosilicate waste glasses. The SA/V ratio affects the rate of glass corrosion through the extent of dilution of corrosion products released from the glass into the leachate solution: glass corrosion products are diluted more in tests conducted at low SA/V ratios than they are in tests conducted at high SA/V ratios. Differences in the solution chemistries generated in tests conducted at different SA/V ratios then affect the observed glass corrosion behavior. Corrosion of borosilicate waste glasses results in the dissolution of soluble components and the accumulation of sparingly soluble components in a layer on the surface of the corroding glass. Laboratory tests performed to distinguish the effects of the solution chemistry and the surface layer on the glass corrosion rate have shown the rate to be controlled primarily by the solution chemistry. Therefore, any testing parameter that affects the solution chemistry will also affect the glass corrosion rate. The results of static leach tests conducted to assess the effects of the SA/V are discussed with regard to the effects of SA/V on the solution chemistry. The effects of the SA/V ratio observed in static tests can be understood based on the effects of the solution chemistry on glass corrosion behavior. Test results show several remaining issues with regard to the long-term glass corrosion behavior: Can the SA/V ratio be used as an accelerating parameter to characterize the advanced stages of glass corrosion relevant to long disposal times? Is the alteration of the glass surface the same in tests conducted at different SA/V, and in tests conducted with monolithic and crushed glass samples? What are the effects of the SA/V and the extent of glass corrosion on the disposition of released radionuclides? These issues will bear on the prediction of the long-term performance of waste glasses during storage. The results of an experimental program conducted at ANL to address these and other remaining issues regarding the effects of SA/V on glass corrosion are described.

SUMMARY

This report reviews and summarizes the present state of knowledge regarding the effects of the glass surface area/solution volume ratio (SA/V) on the corrosion behavior of borosilicate waste glasses. The effects of the SA/V occur through dilution of corrosion products and through the effects of the corrosion products on the glass corrosion mechanism and rate. Therefore, the effects of the SA/V on glass corrosion are discussed in terms of the various effects of the solution chemistry. This is accomplished by first presenting a brief summary of glass structure, the processes leading to the alteration and dissolution of the glass, and laboratory tests that have been applied to characterize waste glass corrosion. This background information is required to characterize and quantify the corrosion rate and mechanism, and the influence of the SA/V on the corrosion behavior. Laboratory tests performed to elucidate the corrosion process are then reviewed to assess how various test results are related to the corrosion process and the importance of the solution chemistry on glass corrosion. The results of static leach tests conducted to assess the effects of the SA/V are then discussed based on what is known about the effects of the solution chemistry on glass corrosion. Of course, the understanding of glass corrosion behavior, the effects of the solution chemistry, the effects of the SA/V, and the interpretation of test results has not been achieved independently. The understanding of the effects of test conditions and various aspects of the solution chemistry has advanced simultaneously through the accumulation of a wide and varied data base. Remaining issues identified through the literature review include uncertainties regarding the use of tests at high SA/V to attain advanced stages of glass corrosion relevant to long disposal times, the relationship between tests conducted at different SA/V and between tests with monolithic and crushed glass samples, and the effects of the SA/V and the extent of glass corrosion on the disposition of released radionuclides. Finally, the results of an experimental program conducted at ANL to address remaining issues regarding the effects of SA/V on glass corrosion are summarized. Many of the published results are utilized in the literature review sections where they provide insight into the corrosion behavior and the effects of the SA/V.

Work done to distinguish the effects of the fluid chemistry and glass alteration layers on glass dissolution has shown that the effects of the fluid chemistry dominate the glass corrosion rate. The silicic acid concentration and the pH affect the rate most strongly, with the reaction to release silicic acid usually being modeled as the key corrosion step. The rate of silicic acid release from the glass depends primarily on the temperature, the glass composition, the concentration of silicic acid in solution, and the solution pH. For a given glass composition at a given temperature, any test conditions that affect the silicic acid concentration or the solution pH will affect the glass corrosion rate. The silicic acid concentration and the solution pH are affected by several factors: the release of other glass components as the glass corrodes; the volume of solution available to dilute released glass components; the refreshment of the leachate solution by solution flow, if any; and the formation of secondary phases. The relative importance of these will be determined by the glass and initial fluid compositions, and by the test method and test conditions used in a particular experiment. In general, the glass and solution compositions are determined by the waste form and the disposal site, while the test conditions may be selected to simulate particular environmental conditions, highlight a particular reaction step, or characterize the general corrosion behavior.

Glass corrosion is well-described by a rate equation that combines a rate coefficient that depends on the glass composition, a pH dependence, and a reaction affinity term expressed as the difference between the actual silicic acid concentration and a "saturation" value. Use of this equation requires the experimental determination of (1) the rate coefficient and its temperature dependence, (2) the pH-dependence of the rate coefficient, and (3) the saturation value to characterize the corrosion behavior of each particular glass. In addition, secondary phases that precipitate from highly concentrated solutions and control the silicic acid concentration after

long corrosion progress must be identified. The corrosion of borosilicate waste glasses can be described as occurring in three stages, which are related to the solution chemistry. The first stage occurs when the solution concentration of silicic acid is very low and the glass corrodes at a relatively high rate that depends on the glass composition and the solution pH. This is sometimes referred to as the "forward" or "intrinsic" rate, since it depends on the glass composition and is not affected by the affinity term. The second stage occurs as the silicic acid concentration increases to a maximum level and the glass corrosion rate decreases to a minimum value. The decrease in the corrosion rate is due to a decrease in the reaction affinity. The maximum concentration is usually referred to as the "saturation concentration" for the particular glass, as if the solution was saturated with respect to the glass phase. Of course, the solution cannot equilibrate with the thermodynamically metastable glass phase. However, the corrosion behavior of high-level waste glasses behaves as if the solution approaches saturation. The third stage occurs when secondary phases precipitate from solution to establish a silicic acid solubility limit that may be lower than the "saturation" concentration established by the glass during the second stage of corrosion. This affects the rate through the affinity term. We emphasize that secondary phases do establish thermodynamic solubility limits for glass components in contrast to the glass itself, which establishes "apparent" solubility limits.

Both the glass and leachant compositions and the testing method will determine which stage or stages of glass corrosion are observed in laboratory tests, and laboratory tests can be used to highlight particular aspects of the corrosion process. Tests with large solution volumes relative to the exposed glass surface area, such as MCC-1 and flow-through tests, will maintain dilute conditions and will be unaffected or weakly affected by silicic acid in solution. These tests are used to determine the intrinsic corrosion rate and the pH-dependence of the corrosion rate. Tests that allow silicic acid to accumulate in the solution, such as the MCC-3 test and the Product Consistency Test (PCT), show the effects of the affinity term on the corrosion rate and the approach to saturation. Under some test conditions, secondary phases will precipitate from the saturated solutions and the corrosion process may enter the third corrosion stage, although reaction times of several months or years may be required before precipitation occurs.

The corrosion behavior of high-level waste glasses in these three stages must be known to project their long-term performance. The SA/V ratio used in laboratory tests provides a means of driving the corrosion progress to these different stages within reasonable testing times to characterize the corrosion behavior of a waste glass. Characterization of the disposal site and anticipated storage conditions can be used to determine which corrosion stages will be important during the service life of the storage site. Test results describing the corrosion behavior can then be coupled with anticipated storage conditions in existing performance models to project glass durability over long-time periods.

1.0 INTRODUCTION

This report reviews and summarizes the present state of knowledge regarding the effects of the glass surface area/solution volume ratio (SA/V) on the corrosion behavior of borosilicate waste glasses. The effects of the SA/V occur through dilution of corrosion products and through the effects of the corrosion products on the glass corrosion mechanism and rate. Therefore, the effects of the SA/V on glass corrosion are discussed in terms of the various effects of the solution chemistry. This is accomplished by first presenting a brief summary of glass structure, the processes leading to the alteration and dissolution of the glass structure, and laboratory tests that have been applied to characterize waste glass corrosion. This background information is required to characterize and quantify the corrosion rate and mechanism, and the influence of the SA/V on the corrosion behavior. Laboratory tests performed to elucidate the corrosion process are then reviewed to assess how various test results are related to the corrosion process and the importance of the solution chemistry on glass corrosion.

The results of static leach tests conducted to assess the effects of the SA/V are then discussed based on what is known about the effects of the solution chemistry on glass corrosion. Of course, the understanding of glass corrosion behavior, the effects of the solution chemistry, the effects of the SA/V, and the interpretation of test results has not been achieved independently. The understanding of the effects of test conditions and various aspects of the solution chemistry has advanced simultaneously through the accumulation of a wide and varied data base. Remaining issues identified through the literature review include uncertainties regarding the use of tests at high SA/V to attain advanced stages of glass corrosion relevant to long disposal times, the relationship between tests conducted at different SA/V and between tests with monolithic and crushed glass samples, and the effects of the SA/V and the extent of glass corrosion on the disposition of released radionuclides. Finally, the results of an experimental program conducted at ANL to address remaining issues regarding the effects of SA/V on glass corrosion are summarized. Many of the published results are utilized in the literature review sections where they provide insight into the corrosion behavior and the effects of the SA/V.

Stabilization of high-level waste destined for disposal in a geologic repository will be accomplished by fixing the waste in a borosilicate glass matrix, pouring the glass into a stainless steel canister, placing the canister in a metal container, and placing the assembled waste package in a mined repository. The repository will be engineered so that radioactive waste components will be isolated from the biosphere for the tens of thousands of years that some will remain toxic. The radioactivity of high-level waste will be dominated initially by fission products that disintegrate primarily through beta decay (mainly ^{90}Sr [$t_{1/2} = 28$ yr] and ^{137}Cs [$t_{1/2} = 30$ yr], which will account for about 95% of the radioactivity for the initial 100 years [1]). While the presence of these radionuclides is an issue during waste vitrification, they are relatively short-lived and will be of minor consequence after about 500 years. Radionuclides having longer half-lives disintegrate primarily through alpha decay (mainly ^{239}Pu [$t_{1/2} = 24,400$ yr], ^{240}Pu [$t_{1/2} = 6580$ yr], and ^{241}Am [$t_{1/2} = 458$ yr]). These radionuclides will become the dominant source of radioactivity after about 500 years. Thus, while fission products present significant handling and processing difficulties, high-level radioactive waste forms must be designed to stabilize the long-lived transuranic elements for a period of on the order of 10,000 years or longer.

Radionuclides will be isolated from the accessible environment by multiple barriers. For radionuclides to be mobilized from emplaced waste forms, liquid water must contact and corrode first the container, then the canister, and then the waste glass. Radionuclides could then be released from the glass matrix as it corrodes and could enter the fluid either as dissolved or as

suspended material. The radionuclides could then be transported away from the repository by groundwater flow. Subsequent interactions with the surrounding geology could result in the radionuclides being removed from the groundwater by filtration or sorption onto mineral surfaces. Assessment of the suitability of a disposal system for the storage of radioactive waste requires that the performance of each barrier be evaluated. This report summarizes the current knowledge of the chemical durability of high-level waste glasses.

Corrosion and the subsequent dissolution of a glass waste form occur when interaction with a corrosive fluid causes chemical bonds to break and reform, generating an energetically more stable system. While glasses are thermodynamically unstable with respect to an assemblage of mineral phases compatible with their compositions, transformation in the solid state is effectively prohibited by very slow reaction kinetics. Contact between glass and a corrosive fluid provides an energetically favorable pathway for the glass to convert to other, more stable phases. Glass components may become solvated and enter solution, or may form new bonds and restructure to generate new solid phases at the glass surface. Secondary phases may also precipitate from the fluid as it becomes saturated. One goal of waste stabilization through vitrification is to produce glasses which corrode at sufficiently low rates that waste components remain immobile for long periods.

To evaluate the durability of a candidate waste glass, its long-term durability must be projected under anticipated environmental conditions in a repository. Such projections are made by using computer simulations that are based on models derived from laboratory experiments. Because of this experimental basis, the effects of various reaction parameters on glass corrosion are accounted for separately. For accurate modeling, it is important to understand the effects on the corrosion mechanism of the SA/V ratios and flow rates used in laboratory tests.

The effects of the SA/V used in static leach tests and the flow rate used in dynamic tests on the observed glass corrosion behavior can be understood (and related to glass corrosion behavior in the repository) in terms of the effects of the solution chemistry and, to a lesser extent, the alteration layers formed on the corroded glass. Laboratory tests conducted under controlled conditions may generate solutions which are very similar to fluids that may contact the glass during some period in the repository. It is important that the corrosion of waste glasses in the laboratory tests be understood in a way that can be used to simulate the corrosion of the same glasses in the repository. Therefore, both the corrosion mechanism of the glass and the relation between test results and the corrosion process must be understood.

The first part of this report provides background information, including descriptions of the glass structure and important corrosion processes, a simple model of the glass reaction to support experimental measures of the extent of glass corrosion in laboratory tests, a brief description of commonly used laboratory tests, and a discussion of how various SA/V ratios are attained in the laboratory. The observed effects of sample preparation techniques and various aspects of test protocols are also discussed.

The second part of this report consists of a review of key studies regarding the glass corrosion mechanism and the effects of testing conditions, including the solution chemistry, SA/V, and flow. It is important to recognize that the SA/V is a testing parameter that, by itself, does not directly influence the corrosion behavior. That is, in contrast to the solution chemistry and the test temperature, the glass is "unaware" of the SA/V being used, except for its effects on the solution chemistry as the glass corrodes. Because the SA/V used in a static leach test affects the dilution of glass components, the need to understand how the SA/V affects glass corrosion can be represented in terms of the need to understand the effects of the solution chemistry on glass corrosion. Therefore, a large part of this review is concerned with assessing important effects of the solution chemistry, including the effects of the solution pH and of various dissolved glass components, on the glass corrosion behavior to better understand the effects of

the SA/V. Experimental studies related to the effects of the SA/V can then be interpreted in terms of specific effects of the solution chemistry on the glass corrosion behavior. When reviewing the literature, the following are considered: the soundness of approach, quality of results, validity of conclusions, and consistency of results with other work. This evaluation is accomplished by presenting the data in the context of a description of the reaction path for the dissolution of high-level waste glass, and by assessing how specific test results contribute to this description. While the effects of the SA/V are of primary interest, only a limited number of tests have been performed so as to isolate the effects of the SA/V. Therefore, other effects arising from differences in the glass and leachant compositions, the glass geometry and/or preparation technique, the reaction temperature, and other test variables are considered when appropriate. In general, the papers reviewed will be discussed chronologically according to publication date, except when more recent work provides a basis for interpreting of earlier work. Although experiments performed with simple glasses having only a few components are utilized to demonstrate basic phenomena, the emphasis of the review is on multicomponent glasses relevant to high-level borosilicate glasses.

Four appendices are included: a list of definitions and acronyms is given in Appendix A, the compositions of glasses discussed in the text are compiled in Appendix B, and a derivation of the rate equation used in most computer simulations is given in Appendix C. Appendix D contains data spreadsheets for the ANL Task "Relationship between tests at high SA/V and MCC-1" performed during fiscal years 1991 through 1994.

2.0 DESCRIPTION OF GLASS STRUCTURE

A basic description of glass structure provides a convenient starting point for discussing the breakdown of a glass as it corrodes. Glasses used in high-level waste management are generated when a viscous melt cools sufficiently fast that elemental constituents become fixed in a noncrystalline arrangement. Although the bonding and coordination requirements of glass components are met in the glass phase, strain and distortion of chemical bonds cause the free energy of the glass phase to be higher than that of a corresponding crystalline state. This excess free energy will likely affect the energetics of the corrosion process.

Glasses have only short-range order, which corresponds to the hybridization of their component elements. Long-range periodic order is absent because bond lengths and bond angles may vary. High-level waste glasses are almost exclusively based on silicate glasses, and this review is focused on this family of glasses. The silicate network can be described as a three-dimensional polymeric matrix comprised of $[\text{Si-O}_{4/2}]$ units in which each silicon atom is bonded to four oxygen atoms in a tetrahedron (the tetrahedral coordination around the silicon atom is a result of its sp^3 hybridization) and each oxygen is part of an adjacent silica unit. Oxygen atoms bonded to two silicon atoms are often referred to as "bridging oxygen," since they bridge two silica units. The silicon-oxygen bonds have about 60% covalent and 40% ionic character. While the bond lengths and bond angles are constrained in individual tetrahedra, rotation about the bonds linking tetrahedral units contributes to the lack of long-range order.

Silicate glasses have been selected as host matrices for stabilization of radioactive wastes in part because most components of waste streams can be incorporated into the silica structure. The extent to which other elements can be incorporated depends on their size, coordination, and valence, as discussed in the following paragraphs.

Cations with a +4 charge and radii similar to silicon and can fit into four-coordinate sites are highly soluble in silicate glasses. Trivalent cations such as aluminum and iron(III) typically have coordination numbers of 6 and have low solubilities in silicate glasses (typically on the order of a few weight percent). However, the solubilities of these trivalent cations can increase significantly when they are associated with alkali metals and form paired cations such as $[\text{Na-Al-O}_{4/2}]$ units, which can substitute into four-coordinated sites (typically to levels in excess of 10 weight percent). The formation of such units perpetuates the silicate polymeric network. Boron can participate in such units (to form reedmergnerite groups $[\text{Na-B-O}_{4/2}]$, or it can hybridize as sp^2 and form a glass phase composed of trigonal planar $[\text{BO}_{3/2}]$ units. The coordination of these trigonal units is incompatible with the tetrahedral silicate network and results in phase separation. Boron may exist in both four-fold sites in the silicate phase and in three-fold sites in the borate phase. It is likely that an equilibrium exists between three- and four-coordinated boron in most alkali borosilicate glasses, and that the amounts of three- and four-coordinated boron depend on the alkali concentration. If sufficient alkali is present, boron will exist primarily in four-coordinated sites and yield a more durable glass. It is important to note that, because of the greater difference in charge potential, alkali ions will form stronger bonds with aluminate groups than with borate groups, so that reedmergnerite groups will form only after all aluminate groups (and most other trivalent cation groups) have been "charge balanced." In addition, cations with low charge density are more effective in "charge balancing" trivalent groups than those with high charge density. Thus, sodium is more effective than calcium.

Phosphorous exists in glass mainly in the +5 oxidation state and is sp^3 hybridized. Although phosphorous coordinates with four oxygen atoms, its +5 oxidation state requires one double bond, so that only three oxygen atoms are available for polymerization. The nonequivalence of the oxygen results in severe distortion of the phosphorous tetrahedra, which are not incorporated into the silicate network. Instead, a separate phosphorous glass phase may form. Phosphorous also readily forms apatites with alkaline earths, titanium, and zirconium. These apatites may occur as primary inclusions as the glass solidifies.

The behavior of monovalent and divalent cations in silicate glasses is strongly influenced by their ionic size, their coordination, and their ionic charge. Their bonds with oxygen are highly ionic. Oxygen bonded to monovalent cations cannot propagate the polymeric network, and those oxygen atoms are often referred to as "nonbridging oxygen." While the formation of nonbridging oxygen bonds decreases the rigidity of the network and the durability of the glass, the addition of alkali metals and alkaline earth elements to waste glasses is important because the presence of nonbridging oxygen lowers the melting point and allows waste forms to be processed under conditions that can be attained in commercial glass melters.

Cations of higher valence typically have large coordination shells and form strong bonds with oxygen. They will preferentially coordinate with oxygen atoms to fill their coordination shells at the expense of the silicate network. These groups are difficult to link into the silicate network and have very low solubilities in silica glasses. For example, most actinide elements are six-fold coordinated and are incorporated into alkali silicate glasses at levels that are typically less than 1 mol %. An important exception is uranium(VI), which forms uranyl units that are better accommodated in the network. Uranium has a solubility on the order of 10-15% in alkali silicate glasses, while other actinides have solubilities of 2-4 wt% [2]. The solubilities of highly coordinated components are increased by the presence of alkali metals, which generate nonbridging oxygen and provide charge balance.

Anions are generally large and highly polarizable, and do not fit into the silica structure. Most anions are volatilized at typical melting temperatures of glass waste forms (1050-1150°C) and their incorporation into silica glasses is usually not considered when discussing waste glass compositions. Only fluoride is incorporated into silica glasses to an appreciable extent (5-10 wt%) because of its ability to replace hydroxide (i.e., as $\equiv Si-F$). The other halides are volatilized, although these and other anions may occur in waste glasses as trapped gases or insoluble phases. Sulfur forms sulfates on the top of glass melt in joule-heated melters under oxidizing conditions, but it is only soluble in silica to about 1 wt%. Carbon and nitrogen are oxidized and volatilized, though some may remain trapped in the melt because of incomplete oxidation. Technetium [3,4] and other waste components, such as mercury, will volatilize during vitrification.

While anions generally do not contribute to the formation of the glass network, their presence in included phases may affect the corrosion behavior and performance of the waste form as these phases dissolve. For example, sulfate, phosphate, chloride, fluoride, nitrate, and nitrite all complex released radionuclides, and so the presence of these anions in the glass will affect the disposition of waste components as the glass corrodes.

Various components in the glass structure are commonly categorized in the literature as filling "network forming" and "network modifying" roles, depending on whether the component continues or discontinues the polymeric network structure, or, in other words, whether the component supports bridging or nonbridging oxygen. For several components, their role in the glass structure depends on the presence or absence of other components; i.e., boron requires charge compensation by an alkali metal. Such elements are sometimes called "intermediates," since they may fill network forming or network modifying roles depending on the glass

composition. While this terminology is not rigorous, it is used throughout the glass literature and will be used in this report when relating literature results and where it improves clarity. In general, silicon and trivalent elements that are coupled with alkali are referred to as network-forming elements, while monovalent and divalent elements are referred to as network-modifying elements.

2.1 Chemical Description of Glass Corrosion

Glass corrosion can be described as occurring through a series of chemical reactions to break one or more oxygen bonds such that component species become solvated and released into solution or the bonding scheme is altered in the corroded glass. The release of different species to solution will depend on the number and strength of cation-oxygen bonds that must be broken to generate a detached unit that can be solvated and enter solution. While the strain energy of a bond in a glass will affect some reactions, it can be assumed that the strain is removed as neighboring bonds are broken. When contacted by an aqueous solution, bonds are broken through hydrolysis reactions, which can be written as

where R and R' are cations that may or may not be linked to the glass network. Depending on the size and chemistry of the R and R' groups, the products of the reaction in Eq. 1 may remain part of the glass network or may be released into the leachate as either dissolved or colloidal material. Glass corrosion is conveniently discussed in terms of the identities of the R and R' groups based on their role in the glass structure. In the following discussion, the R group is considered to include cations capable of forming the polymeric network (i.e., network formers), primarily silicon, aluminum, and boron. Because of the high concentration of silica in most waste glasses, R is represented as $\equiv\text{Si-}$ in the following chemical reactions unless other elements are of specific interest. This depiction identifies the bond involved in the chemical reaction. The R' group may be either network formers, in which case Eq. 1 describes network corrosion, or elements that do not contribute to polymerization, such as monovalent and divalent cations.

2.1.1 Alkali Release

The reaction to release alkali metal ions from a nonbridging oxygen can be written as

where $\equiv\text{SiO-H}$ represents a silanol group and M^+ is an alkali metal ion. The net effect of the hydrolysis reaction is the replacement of an alkali atom in the glass by a proton from solution. Alkali release is often written as an ion exchange, since the reaction can also be written as

or

While Eq. 2b gives the basic reaction, an unhydrolyzed proton is not likely to be stable either in solution or in the glass [5]. Equations 2a, 2b, and 2c are related through the ionization reaction of water. Both the activity of the alkali ions in solution and the solution pH will affect the dealkalization of the glass through the equilibria described in Eqs. 2a-c. An expression for the equilibrium constant for Eq. 2b can be written as

$$\log K_{eq} = \log \left(\frac{=SiO-H}{=SiO-M} \right) + \log(M^+) + pH \quad (3)$$

Similar equilibrium constants can be written for Eqs. 2a and 2c, and the equilibrium constants for the three equations differ only by including K_w . Equilibrium constants have been measured for exchange of alkali metals and alkaline earths with silica gels by Dugger et al. [6]. They found the values of $\log K_{eq}$ (for the reaction as written in Eq. 2b) increased as $K < Na < Li \ll Mg, Ca$, and Sr . This ordering is consistent with the general order for ion-exchange of alkali metals and alkali earths, i.e., that the larger, more polarizable ion will be replaced in an ion-exchange solid by the smaller, less polarizable ion. Dugger et al. also concluded that the acidity of the silanol group in silica gel is similar to that of water. The equilibrium in Eq. 2b will be affected by the solution pH: high pH values will shift the equilibrium to the right and favor the formation of $(=SiO-M)$. Conversely, $(=SiO-Na)$ groups will be protonated more easily than $(=SiO-Li)$ groups as the proton concentration increases. This relationship is shown schematically in Fig. 1, where the results of Dugger et al. are utilized to describe the pH effect on the replacement of a proton on silanol with sodium, lithium, or calcium in a $1 \times 10^{-3} M$ solution of the exchanging ion [7]. For sodium, the silanol will exist entirely in the protonated form (as $=SiO-H$) at pH values below about 8, and entirely in the sodium form (as $=SiO-Na$) at pH values above about 12.5. Both forms will occur at pH values between about 8 and 12.5. Of course, the pH values bounding the regions of coexistence of $=SiO-H$ and $=SiO-Na$ will decrease as the solution concentration of sodium increases.

Fig. 1. Schematic Plot of the Fraction of $=SiO^-$ Sites Associated with Protons or Other Cations as a Function of pH in $1 \times 10^{-3} M$ Cation Solutions (after [7]).

Similar ion exchange reactions can be written for alkali bonded to oxygen atoms that are coordinated with aluminum, boron, or other elements. The alkali metal-oxygen bond energies will differ slightly from the energies for oxygen bonded to other R groups, such as $\equiv\text{AlO}-\text{Na}$, and so the values of K_{eq} will differ, although the relative order is expected to remain the same. By analogy to the behavior of aluminum described by Barrer and Klinowski [8], Bunker et al. reasoned that ion exchange at tetrahedral boron sites differs from exchange at silica sites, and that alkali will be released from nonbridging oxygen in the order $(\equiv\text{AlO}^-) < (\equiv\text{BO}^-) < (\equiv\text{SiO}^-)$ [9]. That is, an alkali metal ion will be released from a nonbridging oxygen bonded to silicon more easily than from a nonbridging oxygen bonded to boron or aluminum. This prediction is shown schematically in Fig. 2, where the pH-dependence of the ion exchange is shown for a $1 \times 10^{-3} \text{ M}$ solution of sodium. At pH values higher than the curves, protons on silanol groups will be replaced by sodium. At pH values lower than the curves, protons will remain with silanol groups. This is consistent with the experimental observation that sodium atoms bonded to silicate groups are released more easily than sodium atoms bonded to aluminate groups in aluminosilicate glasses [10].

Ion-exchange reactions may occur between the glass and the solution as a result of competition between alkali metals and alkaline earths. Ions bonded to the glass will be replaced by smaller ions having higher charge densities. Thus, alkaline earths and other divalent cations will replace alkali metals in the glass. The greater stability of these bonds may protect the glass against further corrosion, perhaps by inhibiting water penetration. For example, Buckwalter et al. found that the presence of lead, and to a lesser extent the presence of aluminum, in the leachate solution quenched the corrosion of PNL 76-68 glass [11]. Lehman and Kuchinski determined that both metallic lead and lead(II) oxide greatly reduced the extent of glass corrosion, relative to that occurring in the absence of any lead, while lead(I) oxide enhanced the extent of glass corrosion [12]. The incorporation of magnesium from solution has

Fig. 2. Schematic Plot of the Fraction of $\equiv\text{AlO}^-$, $\equiv\text{BO}^-$, and $\equiv\text{SiO}^-$ Sites Associated with Protons or Sodium as a Function of pH in $1 \times 10^{-3} \text{ M}$ Cation Solutions (after [7]).

also been found to reduce glass corrosion [13,14]. The mechanism by which these cations affect the corrosion cannot be determined conclusively based on the available experimental evidence. The effects may occur in the solution by the formation of secondary phases or at the glass surface.

2.1.2 Water Diffusion

While alkali metals at the glass surface may be immediately released through hydrolysis reactions upon contact with an aqueous fluid, continued alkali release requires a proton source, such as water, to diffuse into the glass to reach other alkali/nonbridging oxygen bonds. That dealcalization and water diffusion are interrelated is evidenced by the complimentary and S-shaped appearance of hydrogen and alkali profiles commonly obtained at the surfaces of corroded glasses. Experimental results have been interpreted based on the diffusion of molecular water and of hydronium into glasses. However, the small opening size in the silica polymeric structure is known to prevent diffusion of molecular water in many zeolites and fused silica [9]. In these materials, diffusion is instead explained as occurring through hydration/dehydration reactions between water and $\equiv\text{Si-O-Si}\equiv$ bonds [15-19]. Nevertheless, defects in the silicate structure may provide diffusion channels large enough to permit molecular water diffusion. For example, water diffusion is known to occur without affecting the silicate network of obsidian [20-23]. In most instances, tests of alkali silicate glasses in silicate solutions also show dealcalization to occur without affecting the silicate network [24]. The difference between water diffusion in silica glass or zeolites and water diffusion in alkali silicate is likely the presence of percolation channels in the latter formed by neighboring nonbridging oxygen atoms [25,26]. These channels may be formed by neighboring silanol groups in obsidian and by alkali and alkaline earths in waste glasses. It is likely that the presence of nonbridging oxygen and "the nature of cations bonded to non-bridging oxygen atoms control the hydration of silica-poor glasses" [27] and the rate of water diffusion into high-level waste glasses.

2.1.3 Dealcalization

Early work with simple alkali silicate glasses showed the release of alkali metal ions to be proportional to the square root of the reaction time initially, then to be proportional to time after longer reaction times. The root time dependence was interpreted to evidence a diffusion controlled rate and the linear time dependence a surface dissolution controlled rate. The diffusion coefficient for alkali release can be obtained from short-term leaching data as

$$D_{\text{exp}} = \frac{\left(\frac{\pi}{4C_i^2}\right)(M_i)^2}{(SA \cdot t^{1/2})^2} \quad (4a)$$

where D_{exp} is the effective diffusion coefficient in the experiment (in cm^2/s), C_i^2 is the concentration of alkali metal ion i in the glass (in moles/cm^3), M_i is the moles of alkali released into solution, SA is the surface area of the glass (in cm^2), and t is the reaction time (in seconds).

Alkali release from simple silicate glasses has been described in terms of the "interdiffusion" of hydronium and alkali metal ions, where the rate of dealkalization is related to the diffusivities of the exchanged species [28-33]. The interionic diffusion coefficient is given by

$$D = \frac{D_M D_H}{N_M D_M + N_H D_H} \quad (4b)$$

where D is the interionic diffusion coefficient, D_M and D_H are the diffusion coefficients of the alkali metal and hydronium ions in the glass, and N_M and N_H are the fractions of alkali metal and hydronium ions bonded to silanol groups in the glass [33]. This model is based on an alkali-for-hydronium exchange mechanism, as given in Eq. 2c, and is supported by the similarity between the hydrogen penetration depth and the alkali depletion depth and by the observation that the measured proton to alkali exchange ratio is often near three, as predicted by this model [34]. However, exchange ratios are affected by the glass composition, corrosion, and analytical conditions, and ratios as low as 1.5 have been measured [35,36]. This discrepancy between the expected and measured exchange ratios may be due in part to dehydration of the sample in the vacuum chamber, which would lead to lower measured amounts of hydrogen in the altered glass layers and thus lower exchange ratios [37]. The effects of dehydration should be greatest at the outer surface of the layer and become less significant deeper into the layer. Cooling the sample during analysis has been shown to reduce the loss of water during analysis in vacuum [33].

The measured proton/alkali ratio can be affected by other reactions that may obscure the stoichiometry of the exchange reaction. For example, hydrolysis of a silanol/alkali metal bond by molecular water according to Eq. 2a followed by sorption of a second water molecule would be expected to give a ratio of 3:1. Silanol is known to strongly sorb water which may remain fixed in silicate glasses even under vacuum conditions [38-41].

Leach tests of several sodium-potassium-silicate glasses in deionized water at room temperature showed the interdiffusion rate to be much more weakly dependent on the glass composition than would be calculated using the interdiffusion model [18]. Monolithic samples were leached in deionized water under conditions that were used to monitor the initial stages of dealkalization (i.e., where the kinetics were linear with the square root of time). While the SA/V used in the tests was not reported, it is assumed that the SA/V was sufficiently low to maintain dilute conditions. Results were reported by Bunker et al. for tests conducted at temperatures of 22, 30, and 60°C. The leachates were titrated with HCl or KOH solutions to maintain a constant solution pH as the glasses dissolved using the pH-stat technique. The net amount of acid added to the leachate was used to measure the extent of corrosion (dealkalization) and determine the effective diffusion coefficient. The interdiffusion model, which utilizes the bulk diffusivities of sodium and potassium, predicted the observed diffusion coefficient would have a minimum near the $0.5\text{Na}_2\text{O} \cdot 0.5\text{K}_2\text{O} \cdot 3\text{SiO}_2$ composition, whereas the data showed the coefficient to increase smoothly between $\text{Na}_2\text{O} \cdot 3\text{SiO}_2$ and $\text{K}_2\text{O} \cdot 3\text{SiO}_2$. In addition, the magnitude of the predicted coefficient was between 2 and 7 orders of magnitude smaller than the measured values. These results demonstrate that the diffusion of sodium and potassium in the glass does not control the kinetics of dealkalization. Instead, diffusion through the hydrated glass surface controls the dealkalization rate.

A different model assumes that it is molecular water, rather than hydronium, that diffuses into glass [5,42,43]. In this model, ion exchange reactions occur rapidly compared to the diffusivity of molecular water, which controls the rate of dealkalization [10]. This model is supported by the observation of molecular water in leached glasses by infrared spectroscopy [44-46], near infrared spectroscopy and differential thermogravimetry [39]. Of course, molecular water may also be produced by condensation of silanol within the glass (see below).

The mechanism of molecular water diffusion was proposed by Smets and Lommen to be operative in the pH range of 4 to 7. This mechanism is likely to be favored at the higher pH values commonly attained during corrosion of high-level waste glasses, where the concentration of hydronium is very low.

The dealkalization mechanism has been studied by corroding simple glasses and simulated waste glasses in isotopically labeled solutions. Pederson et al. reported that ^{18}O penetrated a soda lime silicate glass to the same depth as sodium was depleted, which they interpreted to indicate interaction between molecular water and the silica [47,48]. Their tests also showed that more oxygen was fixed to the glass than deuterium. This result indicates that the diffusing species, whether molecular or ionic water, was not incorporated into the glass through a simple insertion mechanism. The isotope effect observed by Pederson et al. when a sodium silicate glass was corroded in D_2O and in H_2O showed that a hydrogen (or deuterium)-oxygen bond was broken during the dealkalization process [48]. The authors ascribe the isotope effect to hydrolysis of the silicate network, rather than mass transport, for this glass.

In similar tests of more durable soda aluminosilicate glasses using isotopically labeled water, Pederson et al. did not observe a kinetic effect [49]. Their results implied that very little structural rearrangement occurred following ion exchange, and the authors suggest that the observed corrosion rate is controlled by mass transport instead of chemical reaction. These results are probably more representative of high-level waste glasses than are the results for soda silicate glasses discussed earlier.

The dealkalization of a soda-lime silicate glass was investigated in tests conducted at 100 and 200°C in deionized water, in a silicate solution (152 ppm SiO_2 and 37 ppm NaCl), and in a 5 M sodium chloride solution to discriminate between models based on molecular water and on hydronium diffusion [50]. Both the hydrogen and sodium profiles in the alteration layers were measured by using resonance nuclear reaction spectroscopy. Results for a sample reacted in silicate solution at 100°C is shown in Fig. 3. Shown in the plot are the measured concentrations of hydrogen and sodium and the "excess" hydrogen not required in the ion exchange reaction. Note that the decrease in the measured hydrogen concentration at the outer surface is probably due to dehydration in the vacuum. The hydrogen penetration and sodium depletion depths were the same after corrosion at 100°C in deionized water or in the silicate solution. This suggests the mechanism is controlled by an exchange reaction between sodium and a proton in both leachants. The "excess" hydrogen is due to the presence of molecular water, which accompanies the proton as it penetrates the glass. The depth of hydrogen penetration and sodium depletion was slightly greater in tests conducted in the silicate solution than those in the deionized water, which is consistent with less dissolution of the hydrated glass in the silicate solution. The hydrogen penetration and sodium depletion depths were much less after corrosion in the sodium chloride solution. Only the outer 20-60 nm of the glass was chemically altered. This dealkalization could have occurred by exposure to air prior to initiating the test, as has been shown by Hench [51]. These results suggest that in the absence of dealkalization, penetration of hydrogen (either as molecular water or as an ionic species) did not occur. In other words, stopping the ion exchange stopped the penetration of water into the glass.

Tests in deionized water and in the silicate solution conducted at 200°C showed similar behavior to that observed at 100°C except the alteration depths were greater. Tests in the NaCl solution at 200°C showed sodium to be depleted to a depth similar to that measured in tests conducted in deionized water. However, hydrogen had penetrated deeper into the glass than sodium was depleted. Thus, penetration of water occurred in the absence of dealkalization at 200°C.

Fig. 3. Measured Concentrations of (▲) Sodium, (■) Hydrogen, and (□) Excess Hydrogen at the Surface after Corrosion in Silicate Solution at 100°C (after [50]).

Dran et al. also found that the H/Na ratios increased from about 1.8 to about 2.5 in at 100°C, and from about 1.8 to about 2.5 in tests 200°C, in both tests in deionized water and in silicate solution, as the layer/glass interface was approached. The similarities in the ratios with depth suggest that the layers are not diffusion barriers. The slight increase in the ratio of H/Na as the pristine glass was approached was interpreted to indicate a decrease in the extent of alteration of the alkali-depleted layer, such as hydrolysis of the silicate network, and to represent the ion exchange reaction in the absence of competing reactions. The authors interpreted the results to indicate that ion exchange and water permeation occur simultaneously, and that the test conditions determine which is the controlling process [50]. At 100°C, ion exchange controls the dealkalization rate and controls the water penetration rate. High concentrations of alkali in the leachate slow the dealkalization and thereby hinder water penetration. At 200°C, some water penetration can occur independent of dealkalization.

The corrosion of a nuclear waste glass showed similar behavior [52]. Reaction in deionized water at 120°C resulted in anticorrelated hydrogen and sodium profiles, while reaction in a 5 M NaCl solution showed very little dealkalization. At 200°C, hydrogen penetrated into the glass to a greater depth than sodium was depleted in tests conducted in either deionized water or the NaCl solution. Dran et al. determined that the H/Na ratio never reached a value of three, and concluded that hydronium was not involved in the hydration reaction [52]. Other analyses showed the altered layer to restructure and insoluble components to accumulate in the layer [24]. The restructuring of the layer was found to generate molecular water due to the condensation of silanol groups (see Eq. 5a below) [47,53].

Studies with simple glasses have shown the models developed using test results for some glasses cannot be applied to some other glasses. The extension of models developed based on simple glasses may be even less applicable to complex high-level waste glasses. While the specific models may not be directly applicable, knowledge gained regarding the processes that occur in simple glasses provide valuable insight into the study of high-level waste glasses.

These different results also underscore the importance of accounting for how the test conditions influence the observed extent of corrosion and the effects of possible analytical artifacts. For example, the work of Bunker et al. shows the dealkalization reactions to be highlighted by maintaining dilute conditions and preventing the pH from increasing. The work of Dran et al. shows the dealkalization reactions to become quenched in the presence of high sodium solution concentrations. Different experimental approaches consistently show a net exchange of H^+ for M^+ during dealkalization, although uncertainties remain regarding the identity of the speciation of the inwardly diffusing proton source (H^+ or H_3O^+). Distinction of the proton source is of mostly academic interest, and is mostly irrelevant to characterizing the behavior of high-level waste glasses, where the release of waste components is of prime importance. The identity of the hydrogen species involved in a particular reaction may not be experimentally accessible due to the extremely high mobility of a proton in water. If, rather than an isolated water molecule, a linear array of molecules penetrates the glass through a percolation channel, then it may be impossible to distinguish whether the end of the array is H_2O or H_3O^+ .

2.1.4 Release of Network-Forming Elements

The degradation and eventual dissolution of the glass occurs by hydrolysis of the bonds forming its polymeric network. Hydrolysis of $\equiv Si-O-Si \equiv$ linkages may occur through nucleophilic attack on a silicon atom by either molecular water or hydroxide. Silicate glasses are typically quite resistant to acid attack. Hydrolysis is modeled to occur through formation of a five-coordinate activated complex, which may degrade by breaking the network bond [54,55]. The net reactions can be written as

or

Hydrolysis of terminal $-Si(OH)_3$ groups leads to dissolution of the glass as

or

The forward reaction in Eqs. 5a and 5b is referred to as a network hydrolysis reaction and the forward reaction in Eqs. 6a and 6b is referred to as a network dissolution reaction. The reverse reactions are referred to as condensation reactions. The $\equiv SiO^-$ groups generated in Eqs. 5b and 6b may become protonated by interacting with another water molecule. Note also that silicic acid is a weaker acid than silanol (at 90°C, $pK_a(\text{silanol}) = 6.8$ [56], and $pK_{a1}(\text{silicic acid}) = 9.7$ [57]). Because hydroxide is a stronger nucleophile than molecular water and because experimental observations indicate that the pH affects the rate of silicon release (see Sec. 3.3), it is likely that hydroxide is the dominant nucleophile in neutral and alkaline solutions, and that Eqs. 5b and 6b more accurately represent the hydrolysis reactions.

Similar reactions can be written for the hydrolysis of other terminal metal-oxygen bonds, such as for boron at terminal sites

The release of boron may consume a second hydroxide as

The pK_a for this reaction is 9.23 at 20°C [58] and 8.81 at 90°C (calculated from the results of [59]).

2.1.5 Glass Corrosion

It is clear from considering the dissolution of individual glass components that all processes are interrelated through their occurrence in a common fluid; that is, changes in the solution chemistry due to the release of one component will affect the release of other components. Hydroxide produced during alkali release through Eq. 2a, for example, will catalyze the hydrolysis of nearby $\equiv\text{SiO}-\text{B}=\text{}$ bonds. The replacement of alkali in the glass with protons and the hydrolysis of network bonds will increase water diffusion into the glass, and larger amounts of water in the glass will promote the ion exchange reactions. Water subsequently diffuses into the glass and exchanges with alkali metal ions that are encountered. The release of alkali metal ions via ion exchange generates hydroxide, which then catalyzes hydrolysis reactions.

The relative rates of individual reaction steps will also affect the overall glass corrosion rate because these rates control the number of reaction sites exposed. Initially, the number of reaction sites is proportional to the surface area. As water penetrates into the glass and a dealkalinized surface layer forms, the number of structural $\equiv\text{Si}-\text{O}-\text{Si}\equiv$ bonds available for reaction increases significantly and becomes proportional to the volume of the layer. Glass components having limited solubilities may participate in reactions to form alteration phases at the glass surface. The corroded surface layers of many waste glasses have been observed to be transformed into clays having compositions similar to the glasses on which they form [60-65].

2.2 Experimental Measures of the Extent of Glass Corrosion

As described in Section 2.1, contact of glass by an aqueous fluid results in the diffusion of water into the glass, partial dissolution of the glass, and physical and chemical alteration of the glass surface in contact with the fluid. Because glass corrosion involves several processes which lead to the alteration of the network, it is difficult to precisely define or operationally measure the extent of glass corrosion. Different measures of corrosion can be defined in terms of each reaction process, and different measures may be appropriate during different stages of corrosion and in different tests.

Although the extent of reaction may be determined by the depth of water penetration into the glass, the penetration depth of water is difficult to measure and is not a useful monitor of the glass corrosion. Indeed, diffusion of water into the glass does not constitute glass alteration or corrosion. The extent to which leachable components are released into solution provides an upper limit on the glass corrosion rate and an appropriate measure for modeling the release of leachable radioelements such as ^{137}Cs and ^{99}Tc . The release of other radioelements requires the dissolution of the glass matrix, and may be better modeled using the dissolution rates of matrix-forming elements such as silicon and aluminum.

The most common measure of the extent of corrosion utilizes the measured amounts of glass components that are released into solution. The mass of a component i in solution is usually normalized to the surface area and weight fraction of that element in the glass to permit the results of tests with different glass performed at different SA/V ratios to be compared directly. The normalized elemental mass loss of i , $NL(i)$, is given by the function

$$NL(i) = \frac{m_i - m_i^{\circ}}{SA \cdot f_i} \quad (9)$$

where m_i is the mass of element i measured in the leachate, m_i° is the mass of element i in the initial leachant solution, SA is the surface area of the glass, and f_i is the weight fraction of element i in the glass, i.e., $mass_i/mass_{glass}$. Alternatively, the masses in Eq. 9 can be replaced by the concentrations (c_i and c_i°) if the SA term is replaced by (SA/V) to give

$$NL(i) = \frac{c_i - c_i^{\circ}}{(SA/V) \cdot f_i} \quad (10)$$

If c_i and c_i° are expressed in g/m^3 and SA/V in m^{-1} , then $NL(i)$ will have units of g/m^2 . It is important to note that, because of the f_i term, the mass reflected in the units of $NL(i)$ is the mass of *glass*, not mass of species i . The value of $NL(i)$ represents the amount of glass that has reacted, as calculated using the solution concentration of species i , not the amount of i that is released.

If all glass components are released stoichiometrically, then the values of $NL(i)$ will be the same when calculated using the solution concentrations of different species. Otherwise, the value of $NL(i)$ will be different for each species used in the calculation. High-level radioactive waste glasses generally contain many chemical components that have different solubilities and may be released from the glass at different rates. They are usually found to corrode nonstoichiometrically under most test conditions. For example, Fig. 4 shows the normalized mass losses calculated from the solution concentrations of several major glass components for a glass relevant to the Defense Waste Processing Facility (DWPF) [66,67]. The glass was reacted in a tuff groundwater solution at 90°C following the Product Consistency Test (PCT) protocol (see Sec. 2.5.1.2). Alkali metals and boron are released to a greater extent, and uranium to a lesser extent, than silicon. Clearly, the measured extent of glass corrosion is highly dependent on the element selected to calculate $NL(i)$; for example, the integrated rate (see Sec. 2.3) based on lithium release is about 3.5 times that calculated based on the release of silicon. For this reason, the element used to calculate the normalized mass loss should be identified, and the corrosion of different glasses should be compared based on the release of the same element. Notice in Fig. 4 that the differences in the $NL(i)$ calculated using different elements become nearly constant for tests conducted beyond 181 days; the rates become much more similar after that time. This change may reflect a change in the corrosion mechanism occurs after about 181 days that results in nearly stoichiometric release. That is, these results imply that the reaction conditions, not the glass composition, determines whether corrosion occurs stoichiometrically or nonstoichiometrically. An increase in the leachate pH as the glass corrodes is responsible, in part, for the change from nonstoichiometric release to nearly stoichiometric release seen in the tests.

Early work with borosilicate glasses used silicon as a measure of the extent of glass reaction because it is the most abundant cation in the glass, is the predominant network-forming species, and is present in most glasses tested. However, the solution concentration of silicon may be a poor indicator of the extent of glass reaction because several glass components may be

Fig. 4. Product Consistency Tests Showing Nonstoichiometric Release of Glass Components. (○) Si, (■) K, (●) B, (▲) Na, and (◆) Li (after [67]).

released faster than silicon. Also, many secondary phases that have been seen to form during glass dissolution contain silicon. The solution concentration of silicon will not be indicative of the amount of silicon lost from the glass if some of the silicon released from the glass is contained in secondary phases. It should be noted that early work was conducted using tests at lower SA/V ratios in which the nonstoichiometry of glass corrosion was not as obvious as that seen in Fig. 4.

Water diffusion into the glass and the release of alkali metal ions and boron from the glass generally occur to a greater extent than hydrolysis reactions and the release of silicon and most other glass components. The solution concentrations of alkali metals and boron are often used as a measure of the extent of glass reaction in part because they represent the upper limit of elemental release rates (as seen in Fig. 4). A benefit of using boron as a measure of the glass reaction progress instead of an alkali metal is that boron is contained in very few secondary phases [68]. Alkali metals are contained in zeolite and clay phases that commonly form during glass corrosion. Boron release has been shown to be sensitive to reaction conditions such as temperature [69], pH [70,71], and silicic acid concentration [72,73], and it usually provides the best single measure of the extent of glass corrosion. The boron release has also been shown to be more sensitive to changes in the corrosion rate than other glass components [65]. In addition, boron is rarely present in appreciable concentrations in groundwaters or in other components that may be present in the repository, such as minerals in the host rock or engineering components, and so the glass corrosion can be measured in laboratory tests in the presence of other components without the need for background corrections.

One shortcoming of using boron to measure the extent of corrosion is that boron is not present in naturally occurring glasses or most historic and artifact glasses. These glasses have been used to derive correlations between glass corrosion rates and glass compositions that can be used during production of high-level waste glasses [74]. These glasses cannot be included in correlations based on boron release.

The analysis of the corrosion rate using solution concentrations can be complicated by the formation of colloids and suspended solids, including suspended glass particles in tests performed using crushed samples. While leachates are readily filtered, filtration may introduce artifacts as a result of sorption of material to the filtering material, dissolution of the filtering material, etc. Whether a leachate needs to be filtered or not should be determined by the test being conducted (i.e., if the sample is a monolith or crushed) and the information desired. If the purpose of the test is to determine how much material has been lost from the glass, then both dissolved and colloidal material should be included in the analysis. However, if crushed glass is used, coarse filtration may be required to remove suspended glass particles. If the purpose of the test is to provide a measure of the dissolved concentrations for use as simulation parameters for computer modeling, then filtration may be required. The need to filter solutions is sometimes incorporated into standard test protocols and sometimes left to the discretion of the experimenter. In either case, it is important to consider possible effects of filtration on the measured solution concentrations.

The normalized mass loss and normalized release rate (which is simply the normalized mass loss divided by the test time) provide upper bounds on the extent of corrosion (assuming that the element that is released to the greatest extent is used to calculate it) in terms of the mass of glass that dissolves per square meter of exposed glass surface and per unit time. Direct application of the normalized release to assessing the performance of a waste form requires knowledge of the surface area, from which can be calculated the fractional release rate.

The mass loss of a sample after corrosion can be measured directly and used as a measure of the extent of corrosion. The measured mass loss has been used extensively in tests utilizing monolithic samples. The measured mass loss is related to the normalized mass loss calculated based on solution analyses: the value of NL is an estimate of the total mass dissolved assuming the glass dissolves congruently, and will vary depending on which element is used in the calculation. Nonstoichiometric dissolution may result in NL being greater than the measured mass loss if the solution concentration of a leached component (alkali metal or boron) is used. Direct measurement of the mass loss is complicated by the formation of alteration layers or precipitation of secondary phases on the glass surface. Alteration layers and secondary phases are usually physically removed before measuring the mass loss.

Another measure of the extent of glass reaction is the thickness or mass of the outer glass layer that becomes depleted of alkali metals and boron as the reaction progresses. The thickness of the layer can be measured in cross section by using microscopic techniques, or the layer can be removed from monolithic samples and weighed directly. Neither the layer thickness nor the mass give an absolute measure of the extent of glass reaction; rather, they give the difference between the dissolution rates of the alkali metals and boron from the interior of the glass and the dissolution rate of silicon from the glass surface. The layer thickness will also be affected by drying the sample for analysis, sample preparation, and the environment in which the sample is analyzed. It is best to couple the layer thickness and solution data to attain a material balance for the reaction process [75,76]. Analysis of the reaction layer provides a valuable -and sometimes the only- measure of the extent of glass reaction in systems where solution analyses cannot be performed, such as in field tests [77-80] or in tests with water vapor [81]. In this case, the enrichment of insoluble components in the layer may provide a measure of the extent of etching of the reacted surface. That is, the amount of insoluble material left as residue on the reacting surface as the rest of the glass dissolves can be compared to the bulk concentrations to calculate the volume of glass that must have dissolved to generate the measured amount of residue. The total volume corroded (including the amount dissolved, if known, and the amount remaining in

the alteration layer) is related to the normalized release through the density of the glass. For example, the extent of corrosion leading to a layer with a uniform thickness can be estimated by multiplying the thickness of the layer (in μm) by the density of the glass (in g/cm^3) to obtain the normalized mass loss (in g/m^2).

The use of solids analyses to monitor the extent of glass corrosion should be related to a viable corrosion model and should assure material balance. For example, the precipitation of secondary phases on the surface should be excluded from the layer thickness measurements, since most of the material in these phases was made available after chemical alteration of the glass surface, and its inclusion would, in effect, double count the release of those components. In some cases, an abundant secondary phase may settle evenly across the glass surface so that the thickness of the layer formed by the secondary phase can be used as a measure of the reaction progress in lieu of the alteration layer formed on the glass. Otherwise, the general abundance of secondary phases can provide a qualitative measure of the extent of glass reaction.

2.3 Measurement of the Corrosion Rate

The corrosion rate is determined based on the measured extent of corrosion as a function of the corrosion time. Corrosion rates can be calculated based on any of the measures of the extent of corrosion discussed above. The corrosion rate can be expressed in terms of the overall extent of corrosion measured over the total time tested, which is referred to as the integrated rate, or as the rate at a specific time, which is referred to as the instantaneous rate. The difference between the two rates is illustrated using the measured lithium normalized mass losses from data in Fig. 4, where the normalized lithium mass loss is used to measure the extent of corrosion. These data are shown in Fig. 5. The integrated rate is determined by simply dividing the extent of reaction at the point of interest by the reaction time, and gives the average rate from the time the test was initiated through the time of interest. The integrated rate after a reaction time of

Fig. 5. Schematic Plot Showing Instantaneous and Integrated Rates for a Specific Reaction Time "A" on a Typical Release Curve.

560 days is calculated to be $(1.37 \text{ g/m}^2)/(560 \text{ days}) = 2.5 \times 10^{-3} \text{ g/m}^2/\text{d}$. The instantaneous rate is given by the tangent to the corrosion curve at the point of interest, and is usually approximated using data points collected after two different reaction times. From the lithium data in Fig. 5, the normalized lithium mass loss after 280 days is 1.10 g/m^2 and after 980 days is 1.47 g/m^2 so that the instantaneous rate from the data between 280 and 560 days is $(1.37-1.10)/(560-280) = 9.6 \times 10^{-4} \text{ g/m}^2/\text{d}$, and between 560 and 980 days is $(1.47-1.37)/(980-560) = 2.4 \times 10^{-4} \text{ g/m}^2/\text{d}$. The average instantaneous rate calculated from these test data is $6 \times 10^{-4} \text{ g/m}^2/\text{d}$, which is less than one-fourth the integrated rate. The values of the integrated and instantaneous rates are similar in the early stage of the corrosion when the glass corrodes rapidly, but become quite different beyond the steep portion of the curve at short reaction times. This is because all integrated rates account for the initial rapid corrosion while the instantaneous rate does not. Which rate is being discussed should be specified when presenting rate data.

Only the integrated rate can be determined for a test in which only one time period is used, as in product consistency tests. While the integrated rate can be used to compare the relative durabilities of different glass compositions under specific conditions, it is not particularly useful for characterizing the glass corrosion behavior or projecting long-term behavior. The integrated rate is strongly influenced by the initial corrosion rate and is only weakly sensitive to changes in the corrosion rate at long time periods. Also, rates that become constant at long time periods do not give constant integrated rates. Instantaneous rates are sensitive to changes in the corrosion rate and are better suited to characterizing the long-term corrosion behavior.

As described earlier, several different reactions and processes are involved during glass corrosion, and which controls the observed corrosion rate may change as the corrosion progresses and may differ under different test conditions. Utilizing a single measure of the extent of corrosion throughout the corrosion process may be misleading, since that measure may or may not track the rate-determining process. For example, dealkalization dominates the initial stage of glass corrosion while the release of silicic acid does not occur until the pH increases above about 9. Tracking the silicon concentration does not characterize the initial stages of corrosion. The intermediate stages of corrosion can be followed using the silicon concentration in solution, since the glass dissolves nearly stoichiometrically. Secondary phases formed after significant corrosion contain silicon, so that the silicon concentration no longer provides a good measure of the extent of corrosion. Likewise, measurement of the thickness of alteration layers depends on the relation between dealkalization and water penetration, which establish the layer/glass interface, and hydrolysis and dissolution, which establish the layer/solution interface. Early in the corrosion, dealkalization occurs rapidly and dissolution occurs slowly, while in the intermediate stages they occur at similar rates and during later stages precipitated material may settle on the glass surface. Therefore, it is important to relate what is being used to measure the extent of corrosion to the corrosion mechanism, and it is beneficial to utilize more than one measure to assure that changes in the rate controlling step will be accounted for.

2.4 A Phenomenological Model of Corrosion Behavior

As will be described in this report, an extensive and diverse range of laboratory tests have led to a generally accepted description of high-level waste glass corrosion. The following description of corrosion in a static system is referred to as phenomenological because it does not rely on the identification of specific chemical reactions or processes, rather it summarizes various experimental observations. It is presented to support the discussion of particular aspects of corrosion in later sections of the report. Figure 6 schematically illustrates the corrosion path followed as a waste glass corrodes under static conditions, where the extent of corrosion is indicated by the solution concentration of a highly soluble glass component. (Test conditions where the leachate solution is diluted due to flow or periodic replenishment will prevent the

Fig. 6. Schematic Plot Showing Corrosion Behavior as the Corrosion Progresses. Stage I occurs when the solution concentrations of glass components are too low to significantly affect the corrosion rate. Stage II occurs when glass components approach an apparent saturation concentration and slow the corrosion to a negligible rate. Stage III occurs when secondary phases maintain glass corrosion products at concentrations below the apparent saturation values.

environmental conditions from evolving. The change in the solution chemistry allowed by the test conditions will depend on the specific flow or replenishment rate. The effects of specific test conditions on the corrosion behavior are addressed below where pertinent.) The reaction path described by the curve is consistent with existing quantitative models discussed in Section 3. The reaction progress variable shown as the x-axis in the figure includes the effects of the reaction time and various test conditions, including the SA/V. The reaction progress variable is sometimes represented as the product of time and the glass surface area/solution volume ratio when plotting experimental data. However, it will be shown that the different pH values attained in tests at different SA/V complicate direct comparison of tests at different SA/V (see Section 4.2) [81].

The curve drawn in the figure describes the evolution of a particular corrosion system where the solution chemistry freely evolves as the glass corrodes over time. Hence, several important aspects of the system change as the reaction progresses, including the solution pH, the concentrations of glass components in the leachate, and the development of an alteration layer on the glass surface due to nonstoichiometric glass corrosion and secondary phase formation. The y-axis represents the measured amount of glass corroded, and different measures of the extent of corrosion will yield slightly different curves. The curve in Fig. 6 is meant to represent the solution concentration of a highly soluble glass component (such as boron). The corrosion rate is given by the derivative of the curve. Different curves will exist for different glass compositions, leachant compositions, and temperatures. The following discussion is with regard to a particular system of glass, leachant, and temperature.

The corrosion is described as occurring in three stages which are represented by changes in the solution concentration of boron in Fig. 6. (Boron is commonly used as a measure of the extent of glass corrosion and, for convenience of discussion, will be used for that purpose here.)

During the initial stage of corrosion, referred to here as Stage I, corrosion occurs at a constant rate that depends on the glass composition and the solution pH that is often referred to as the forward rate. The boron concentration increases at a constant rate during this stage of the corrosion, and is not measurably affected by the presence of glass corrosion products in solution. The rate measured in the absence of effects of dissolved glass components (the forward rate) provides a valuable characteristic of the glass for modeling its long-term corrosion behavior. The forward rate is affected by the glass composition, the test temperature, and the solution pH. Different glass components may also be released at different forward rates. That is, the glass may dissolve nonstoichiometrically during Stage I. Dynamic tests conducted under high flow rates or frequent leachate replenishment can be used to maintain the corrosion in Stage I.

As the glass continues to corrode, the corrosion rate will eventually decrease as corrosion products accumulate in the solution. This is referred to as Stage II. The transition from Stage I to Stage II occurs gradually as the corrosion rate decreases as the solution becomes more concentrated in corrosion products. The distinction between Stages I and II is somewhat arbitrary, and depends, in part, on the ability to measure small changes in the corrosion rate. The rate will also decrease as alteration layers form on the glass surface and slow the release of glass components through mass transfer effects. The amounts of glass components released to solution will also decrease as the surface area of the dissolving glass decreases, although the dissolution rate itself will not necessarily decrease. Corrosion may continue in Stage II for very long time periods. Eventually, the rate will decrease to near zero as the chemical potential difference between components in the glass and in the solution approach minimum values. The rate measured experimentally is referred to as the "saturation rate," since the solution appears to become saturated with respect to the glass. Corrosion in Stage II is observed in most static leach tests where dissolved glass components are allowed to accumulate in the leachate solution.

Secondary phases will eventually precipitate from the concentrated solution and will alter the solution chemistry. The change in the solution chemistry will then affect the corrosion rate. The formation of secondary phases that affect the corrosion rate marks the beginning of Stage III. The extent to which secondary phases affect the corrosion rate depends on the assemblage of phases formed and how they alter the solution chemistry. In fact, some secondary phases form without measurably affecting the corrosion rate. Clays commonly form early during the corrosion of borosilicate waste glasses, but their formation does not measurably affect the corrosion rate. This is probably because their effect on the solution chemistry is negligible. The increased corrosion rate shown in Fig. 6 is similar to that observed for high-level waste glasses under some test conditions. Corrosion in Stage III has been observed in static leach tests conducted at very high SA/V and elevated temperatures where highly concentrated leachate solutions are generated.

The corrosion rate observed in Stage III depends on the relative rates of secondary phase precipitation and glass dissolution, that is, whether or not the secondary phases equilibrate with the solution. If secondary phases precipitate fast enough to maintain equilibrium with the solution, then the observed glass dissolution rate will be higher than that prior to secondary phase formation and will be constant (as shown in the figure). If secondary phases precipitate slowly, such that the solution remains supersaturated, then the observed rate will slow over time. The ability of the secondary phases to remove glass components from the solution will also depend on the relative surface areas of the glass and the secondary phases. Continued corrosion in Stage III involves the maturation of the assemblage of secondary phases as the initially formed phases are replaced by more stable phases in a paragenetic sequence. As more stable phases form, the solution chemistry and glass corrosion rate will be affected. Corrosion of waste glasses for extremely long time periods will be controlled by changes in the solution chemistry that occur during the evolution of the assemblage of secondary phases.

Glass corrosion in Stages I and II is commonly observed in most laboratory tests where the corrosion products are allowed to accumulate in the leachate solution. Progression into Stage III is not commonly seen in laboratory tests simply because the glass corrosion does not reach Stage III under the test conditions used within the reaction times tested. Several examples exist where the last data point collected is observed to be higher than expected based on the trend shown by tests conducted for shorter time periods; these tests have likely progressed to Stage III. By performing tests under conditions that lead to highly concentrated solutions within a relatively short time, the corrosion progress of most glasses can be driven to Stage III.

The corrosion behavior of a glass in each Stage cannot be predicted from its measured behavior in any other Stage because what controls the corrosion rate is different in each Stage. In Stage I, the corrosion is controlled by the glass composition and leachant pH. In Stage II, it is controlled by the solution chemistry (primarily silicic acid concentration and pH) which is determined by the glass composition. In Stage III, the corrosion rate is controlled by the solution chemistry that equilibrates with the assemblage of secondary phases that is formed. While the compositions of the secondary phases is determined in part by the glass composition (the solution from which they precipitate is composed of dissolved glass), the identity of secondary phases that form initially and the effects of their formation on the glass corrosion rate must be determined experimentally.

The prediction of glass corrosion behavior over long time periods requires the corrosion progress curve of the waste glass be known and related to storage conditions. This requires characterization of the glass corrosion behavior using several test methods to determine (1) the forward reaction rate, (2) the solution saturation conditions and the saturation rate, and (3) the identity of the assemblage of secondary phases formed and determination of how they affect the corrosion rate. Once the corrosion curve of the glass is determined, how far along the curve corrosion will progress under particular disposal scenarios can be estimated based on projected groundwater flow rates, amounts of groundwater expected to contact the waste packages, chemical effects due to interaction of the groundwater with the surrounding geology, radiation effects, etc. This report now focuses on the test methods used to describe the glass corrosion behavior and experiments conducted to establish the curve and analytical model of the corrosion rate.

2.5 Standard Test Methods

Laboratory tests are performed on glass waste forms to provide information on their chemical durabilities. Test results can be used to guide formulation of waste form compositions and to monitor their production, or to determine waste form corrosion mechanisms and predict their long-term durabilities under potential disposal environments. The information needed will determine the test methodology that should be used and the required detail of analysis of the reacted sample. Tests designed to monitor the processing of waste forms are discussed in detail because of the extensive database that is available for these tests and because of the characterization of the glass corrosion process that these tests provide. Other tests have been designed to highlight a particular corrosion step or to provide values for use in reaction models.

The American Society for Testing and Materials (ASTM) has issued a Standard Practice for relating laboratory tests to the prediction of glass corrosion (ASTM C1174-91: "Prediction of the Long-Term Behavior of Waste Package Materials Including Waste Forms Used in the Geologic Disposal of High-Level Nuclear Waste"). In this Practice, several categories of tests are identified, including attribute tests, characterization tests, accelerated tests, service condition tests, analog tests, and confirmation tests. The purpose of these tests is to provide parametric values to be used in support of a mechanistic model of the corrosion process and to verify the relevance of the model. The Practice does not identify specific test procedures to be used as attribute tests, characterization tests, accelerated tests, etc., rather it "covers the general approach

for proceeding from the statement of a problem in prediction of long-term behavior of materials, through the development and validation of appropriate models, to formulation and confirmation of actual predictions" [82].

The corrosion of a glass observed in a laboratory test can be utilized simply as a measure of the response of the glass to those particular test conditions, or the results can be further interpreted as a measure of a characteristic property of the glass, usually with respect to a specific corrosion model. Different test methods have been developed to compare the responses of different glasses under standard test conditions without regard to the corrosion mechanism. These tests have been created to assist in the development of waste forms and the monitoring of product consistency. Other tests have been developed to characterize the corrosion behavior of a glass under conditions believed to highlight a certain aspect of the corrosion process. In some cases, the same test method can be used to compare the response of a glass to that of a reference glass and to ascertain a corrosion characteristic of the glass. Of course, the distinction between a response test and a characterization test is primarily in how the results are utilized. However, as will be discussed in detail in this report, the corrosion mechanism should be considered when interpreting the results of response tests because corrosion of different glasses under the same test conditions may result in different characteristic corrosion processes dominating the observed responses. That is, the test may measure different corrosion characteristics for different glasses and, therefore, not measure the same aspect of corrosion for different waste forms. This may or may not affect the utility of the test for determining product consistency.

Service condition tests are designed to simulate environmental conditions anticipated to exist during waste disposal, such as temperature, amount and chemistry of groundwater, surrounding geology, etc. In general, the test conditions used to characterize the glass corrosion behavior are very different from those anticipated to exist during actual storage of the glass. This is because the characterization tests are typically designed to isolate one aspect of the corrosion behavior, while the corrosion of the glass in service condition tests is affected by several interactions simultaneously. Additionally, in an actual disposal site, most conditions are expected to vary over time, while constant environmental conditions are usually maintained in characterization tests. For example, the temperature in a repository is expected to decrease over time while most tests are conducted at a constant temperature. Water can enter the repository only very slowly, first as water vapor, then as small volumes of liquid water. Laboratory tests are generally conducted with much larger volumes of water than are likely to contact the waste in a repository, and generally do not replicate the slow increase in the amount of water present. Thus, the results of characterization tests can only be related to repository performance after careful consideration of the anticipated environmental conditions and an understanding of the glass corrosion mechanism and how it is affected by parameters such as the temperature, water volume and fluid chemistry, and how these conditions change over time. Although specific service conditions and the evolution of the environment in a waste repository are not discussed in this review, the relevance of specific test methods designed to characterize the corrosion behavior of waste glasses to glass performance under likely storage conditions is discussed.

Laboratory tests of chemical durability are performed by contacting the glass sample with a solution and measuring changes in either the glass or the solution. Standard testing procedures have been developed to directly compare results obtained at different laboratories for different glass compositions. These tests can be categorized as dynamic tests, in which the solution is continuously or periodically refreshed, or as static tests, in which the leachate is not refreshed during the test. (Note that in the present context, the specification of static and dynamic refers only to the replacement of leachate solution as the test is conducted.) Tests may be performed on either monolithic samples or crushed samples to provide various glass surface areas that may be contacted by different solution volumes. A summary of test methods used to measure the

chemical durability of high-level waste glasses has been compiled by the International Atomic Energy Agency (IAEA) and is included with other test methods in Table 1 [83]. Some commonly used tests relevant to further discussions in this report are discussed in more detail below.

2.5.1 Static Tests

2.5.1.1 MCC-1 Test

The MCC-1 static leach test (sometimes referred to as MCC-1P) was developed by the Materials Characterization Center to provide a comparison of the durabilities of candidate waste forms developed for the stabilization of high-level nuclear wastes [84]. The method calls for placing a monolithic specimen of known geometric surface area into a volume of leachant such that the SA/V is 10 m^{-1} . The test may be performed in deionized water, a reference silicate solution, or a reference brine. The reference silicate and brine compositions as analyzed in Round Robin tests conducted for the MCC-1 are given in Table 2. The silicate solution is dominated by sodium and silicon, while the brine is dominated by sodium and potassium. Tests are to be conducted in perfluoralkoxy (PFA) Teflon containers, unless the absorbed dose to the container due to the radiation field will exceed 10^4 rad (100 Gy) over the course of a test, in which case a vessel constructed from an inert material (such as stainless steel or fused silica) is to be used. Teflon vessels cannot be used in high radiation fields because of the possibility of fluoride being leached from the vessel [85]. The reference temperature is 90°C and the reference test time is 28 days, although temperatures of 40 or 70°C and other testing times can be used. (The MCC-2 test procedure is a variation of the MCC-1 procedure for reaction temperatures greater than 90°C [86].) The measured solution concentrations of component species are used to monitor the extent of glass corrosion.

Monolithic specimens can be prepared for testing by fracturing, cutting, or grinding the sample material. The surface finish resulting from the surface preparation is known to have a measurable effect on the measured corrosion rate, which increases with surface roughness (see Section 2.7). The increase is due to both the increase in the actual surface area with the surface roughness (the geometric surface area generally does not account for the surface finish) and the increased surface strain. Test results for short-term tests of a few days are strongly influenced by the initial surface condition and the presence of fines. The MCC-1 test provides a solution-dominated system in that the leachate remains dilute as the glass dissolves. Because there is little interaction between released glass components and the glass, the corrosion is strongly influenced by the glass composition. However, tests conducted for long periods may be affected by the leachate solution chemistry. Samples corroded in MCC-1 tests show details of the chemical and physical alteration of the glass surface that cannot be seen in other tests. Detailed analyses of monolithic samples corroded during this test have provided valuable insight into the mechanism controlling the initial stage of corrosion [87-89].

The precision of the MCC-1 test method was evaluated by Kingston et al. by testing glasses with tightly controlled key procedural steps and experimental conditions [90]. Aware of the effects of surface finish on the test result, they ground all test specimens to a 600-grit final finish to improve the uniformity of the samples. They determined that "all the imprecision contributed by the test parameters and the glass samples combined was found to be insignificant compared to the analytical technique used for leachate analysis under these conditions" [90]. Analytical error is typically near 10-15% of the amount present for multicomponent analyses required during the performance of these tests. The precision determined from the MCC Round Robin test program was 6-11% within a laboratory and 25-40% between laboratories [91]. Much of the difference between laboratories was probably due to sample preparation.

Table 1. List of Test Methods (adapted from [83])

Test Name	Temp. (°C)	Leachant	Flow Rate	Sample Description	Ref.
Soxhlet	50-100	DIW	1.5 mL/min	Plate, S = 3 cm ²	1
Modified Soxhlet	35-100	DIW	Variable	Plate or crushed	2
Hot-Cell Soxhlet	100	DIW	80 mL/h	Beads, plates, chips	3
MCC-5 Soxhlet	100	DIW	1.5 mL/h	Plate, S = 4 cm ²	4
Soxhlet (PNC)	70, 100	DIW	60-225 mL/h	Bar, S = cm ²	5
HIPSOL (HT Soxhlet)	100-300	DIW	100-900 mL/h	Powder or block	6
IAEA	25	DIW	Periodic replacement	Cylinder	7
ISO Buffer	23-100	DIW, buffers, or sea water	Periodic replacement	Monoliths	7
ANS 16.1	25	DIW or synthetic seawater	Periodic replacement	Cylinders, S/V = 10 m ⁻¹	8
DLT	25	DIW or synthetic seawater	Periodic replacement	Cylinders, variable	9
Powder (P1)	95-200	DIW	Periodic replacement	100-200 µm	10
Powder (P2)	40-100	DIW	Daily replacement	100-150 mesh	11
MCC-4	40, 70, 90	DIW or reference groundwater	0.1-0.001 mL/min	Plate	4
Low Flow	25-90	DIW	1 mL/wk	Plate	12
Dynamic	35-90	DIW	3-1200 mL/h	Crushed or monolith	13
Grain Titration	100	DIW	Static	Crushed	14
Time-Dependent Method	20-60	Buffer	Static	Disc	12
MCC-1	40, 70, 90	DIW or reference groundwater	Static	Monolith, S/V = 10 m ⁻¹	4
MCC-2	110, 150, 190	DIW or reference groundwater	Static	Monolith, S/V = 10 m ⁻¹	4
MCC-3	40, 90, 110, 150, 190	DIW or reference groundwater	Static (agitated)	Crushed, 149-175 µm, or <45 µm	4
HTLC (CEC)	90, 110, 150, 190	DIW	Static	S = 4 cm ²	15
Autoclave (HMI)	150-200	DIW or brine	Static	Beads, chips	16
Autoclave (KfK)	100, 150, 200, 250	DIW or brine	Static	Cylinders, S= 20 or 5 cm ²	17
Repository Simulation	25-90	Granitic water		Plate, S= 3 cm ²	12
Waste/Water/ Rock Leach	98	DIW or granitic water		Cube, S = 6 cm ² 20 g granite powder	5
MCC-14	25-290	Repository groundwater	Static or periodic sampling	Monoliths or powders	4
PCT A	90	DIW	Static	Crushed, 74-149 µm	18
PCT B	Variable	Variable	Static	Crushed	18
ALT	Variable	DIW	Periodic replacement	Cylinder	19
TCLP	25	Acetate solution	Static (agitated)	Crushed (<0.375 in.)	20

(contd.)

Table 1 - contd.

Test Name	Temp. (°C)	Leachant	Flow Rate	Sample Description	Ref.
Cal. WET	25	DIW or citrate solution	Static (agitated)	Crushed	21
MEP	25	Acetate solution followed by synthetic acid rain			20
MWEP	25	DIW or synthetic site water	Static	Crushed or monolith	22
ELT	25	DIW	Static	Crushed	9
ANC	25	Nitric acid solution	Static	Crushed	9
SCE	25	Acid solution	Static	Crushed	9
Unsaturated Test	25-90	Repository groundwater	75 µL/3.5 days (variable)	Monolith	23
Vapor Hydration	70-250	DIW	Static	Monolith	24
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Table 2. Analysis of Leachants by Participants in the MCC Round Robin of the MCC-1 Test^a

Element	Silicate Solution Average, ppm	Brine Solution Average, ppm
B	0.161	1.412
Ca	0.072	1.014
Cs	0.110	0.520
Fe	0.006	0.867
K	0.782	20650
Na	49	37214
Sr	0.004	0.094
Si	22.76	3.596
pH	7.582	6.48

^aNumber of significant figures reported by Johnston and Daniel [91].

While the MCC-1 method was originally designed to compare the relative chemical durabilities of candidate waste forms, the test can be used to characterize several aspects of the corrosion process in conjunction with a corrosion mechanism. Tests conducted for short periods (on the order of 28 days or less, depending on the reactivity of the waste form) provide a simple and useful measure of the glass corrosion rate under dilute conditions. Variations of the MCC-1 method conducted in buffer solutions or in solutions spiked with various glass components can be used to determine the effects of the leachate chemistry on the glass corrosion rate [92]. Longer term tests can be run to monitor the alteration of the glass surface during corrosion to investigate the corrosion mechanism [87,93-97].

Application of the MCC-1 test to various materials has several advantages. First, heterogeneous waste forms can be analyzed before and after testing to monitor the corrosion behavior of the different phases that are present. Second, sample characterization prior to testing can be performed to assure that representative samples of heterogeneous materials are tested. Third, samples can be prepared with geometric shapes amenable to post-test physical and chemical analytical methods, such as fracture tests and chemical analysis of the reacted surface. One disadvantage of the method is that it may be difficult to prepare monolithic samples of glasses that have not been annealed for testing. The removal of strains in the glasses during annealing may affect the performance of the glass in the test. Preparation of samples of radioactive glasses may also be difficult if remote handling is required.

The MCC-1 test conditions are not representative of disposal conditions that are currently under consideration. The very low SA/V used in the MCC-1 test represents the highly unlikely event of the head space of a waste canister being instantaneously filled with groundwater. More likely, water vapor will first fill the canister and condense on the glass. The amount of water contacting the glass would slowly increase, possibly filling the head space if no drainage from the canister occurred. The glass would continually corrode as more water entered the canister, such that the highly dilute conditions measured in the MCC-1 test may never occur under disposal conditions.

The MCC-1 test has recently been standardized by the ASTM through committee C-26 as Standard C1220-92, "Test Method for Static Leaching of Monolithic Waste Forms for Disposal of Radioactive Waste" [98]. Despite possible difficulties in application with

radioactive samples and its being irrelevant to service conditions, the MCC-1 test provides a simple method of comparing the relative durabilities of different homogeneous waste glass compositions under conditions that are sensitive to the glass composition. The results of MCC-1 tests conducted for short time periods (typically 7 days or less) provide a useful estimate of the forward corrosion rates of candidate waste glasses. The MCC-1 tests are easier to perform than other tests designed to measure the forward corrosion rate, such as Soxhlet and single-pass flow-through tests, which are described below.

2.5.1.2 Product Consistency Test

The Product Consistency Test (PCT) [74] was developed for use at the DWPF to monitor the chemical durabilities of the glasses to be produced and assure that all waste forms exceed the benchmark measure of durability [99,100]. Variations of the PCT are used to monitor the product consistency (PCT method A) and to characterize the corrosion behavior (PCT method B). In the PCT method A, a mass of crushed glass in the size fraction between 100 mesh and 200 mesh sieves is combined with a mass of deionized water ten times that of the glass. The test time is seven days and the test temperature is 90°C. At the end of the test, the leachate solution is filtered using a relative coarse filter (a 0.45- μm filter is called for in the procedure) to remove suspended glass. This solution is analyzed to measure the boron, sodium, and silicon concentrations. The PCT method A was developed to remotely monitor the consistency of glass produced by the DWPF. The PCT method A specifies that stainless steel reaction vessels be used, since the glasses to be tested will be highly radioactive and other materials, such as Teflon, may be affected by the radiation fields [85]. The PCT method A is a "response test," in which the response of a sample glass is compared to the benchmark response of a reference glass, which for the DWPF has been identified as the "SRL EA" glass [100].

The PCT method B allows variation in the size fraction of the glass used, the glass mass-to-leachant mass ratio, the leachant composition, the test temperature, and the test duration. The PCT method B was designed to investigate the effects of various parameters on the response of a specimen so that the results could be used in the further development of waste forms. In effect, the PCT method B encompasses all variations of crushed-sample, static leach tests. The method does not specify the use of a particular reaction vessel; studies have shown the effect of the vessel used in the test to generally be small [67,101,102].

There is some concern regarding the effect of using crushed glass in tests, since waste forms placed in storage will not be crushed and since uncertainties exist regarding the effects of the particle size on the test results. Particle size effects are very difficult to separate from other effects in laboratory tests, in part because the surface area of crushed glasses is hard to measure (see Section 2.6) and the surface area may decrease significantly over the course of the test (see Section 2.9). The surface area is an important test parameter that is approximately proportional to the number of reaction sites available for each corrosion process (water diffusion through channels, ion exchange, and hydrolysis). The surface area is usually combined with the solution volume in a single test parameter, namely SA/V. The SA/V of a test determines the rate of change of solution concentrations as a waste form dissolves, which affects the corrosion rate. The PCT method A avoids addressing surface area effects by specifying that glass of a particular size fraction be used in the test. An acceptably reproducible surface area can then be attained by accurately weighing the glass. However, different glass compositions may fracture differently, and so the total surface areas in tests with different glasses may not be the same. This is probably an unavoidable limitation of using crushed glass.

Also, glasses of different density will result in different numbers of grains being used in the test. That is, fewer grains are present in 1 g of a heavy glass than in 1 g of a lighter glass. If the grains of heavy and light glass are the same size, as assumed for the

same sieve fraction, then less surface area will be present in tests with the heavy glass than with the light glass. The solution concentrations of components released from the heavy glass will be lowered due to the smaller surface area. The solution concentrations could be normalized by multiplying by the densities of the glasses, although this is not done in practice. The correction factors would be of the order of the differences in densities. That is, a glass that has a density 10% higher than another glass is expected to have about 10% less surface area per gram and so the solution concentration is expected to be about 10% lower due to the lower surface area, irrespective of the durability of the glass.

While the surface area is not specified by the PCT procedure, it must be known to compute the normalized mass loss, which is needed to compare PCT results for various size fractions to other test results. (Use of the PCT method A as a response test does not require computation of the normalized mass loss, since all results are compared to other PCT method A results.) The surface area is usually calculated by assuming that the crushed particles have a convenient geometric shape, such as a sphere or a cube, that can be related to the sieve size. Such an approximation suggests the SA/V used in the PCT method A test, for glasses having a density of about 2.7 g/cm^3 , is about 2000 m^2 (see also Section 2.6).

Despite the uncertainty in the absolute surface area, the SA/V used in the PCT method provides a convenient means of accelerating the approach of the solution chemistry to saturation. The reaction system is dominated by the surface area of glass that is available for reaction (in contrast to the MCC-1 method in which the system is dominated by solution). Because the corrosion of a very small amount of glass produces large changes in the solution chemistry under high SA/V conditions, the effects of the solution chemistry and the glass corrosion products on the glass corrosion process are highlighted. Tests that are run for only a few days may attain solution compositions that MCC-1 tests would require several years to attain. Alteration of the glass surface progresses rapidly in response to the solution chemistry, and alterations observed after a year or more in MCC-1 testing can be observed within a few days in PCT testing [81]. Similarities in the alteration of the glass surface demonstrate that the corrosion mechanism is the same under MCC-1 and PCT conditions. Thus, PCT tests can easily be used to accelerate the corrosion process, and PCT tests run for periods between a few days and several months can be used to map the evolution of the solution chemistry as it approaches saturated conditions. These tests can be used to provide parameters to mechanistic models used to predict the long-term behavior of waste glasses under a wide range of corrosion conditions. The composition of the saturated solution is needed for computer modeling of the corrosion process (see Section 6). In addition, secondary phases can be generated in PCT tests conducted for periods on the order of several months or more, depending on the glass durability. These tests can be used to determine the corrosion rate attained upon the formation of secondary phases, which represents the long-term corrosion rate. Analysis of the corroded solids in tests which have progressed into the long-term corrosion stage provides insight into the corrosion mechanism, allows corrosion products to be identified, and confirms that the corrosion mechanism has not changed [65].

Saturated solution conditions, secondary phase formation, and the long-term corrosion rate are attained faster in tests at higher SA/V. Higher SA/V conditions can be attained by simply performing the PCT test using -100+200 mesh glass at a greater glass mass-to-solution volume ratio or by using a smaller particle size. Again, the absolute surface area provided by a particular grain size can only be approximated, although the relative surface area is expected to scale with the mass of glass used in the test. Therefore, tests performed with different masses of the same grain size are expected to scale according to the masses used. Since the rate decrease of surface area as the glass corrodes will be higher for smaller particles, tests with different grain sizes may not scale in a predictable fashion. The loss of surface area will affect the actual SA/V of the test and complicate comparison of the results to tests at other SA/V

(see also Section 2.9). The effect will depend on the relative durability of the glass being tested: durable glasses will show less effect than non-durable glasses. If different grain sizes are used to attain a particular SA/V, some tests should be conducted using glass in the 100 to 200 mesh size fraction to relate the performance of the glass being tested to other glasses and to assess the effects of the grain size.

Complications of using crushed glass include the possibilities that non-representative sampling may occur due to preferential sieving, that the physical shapes of the glass particles affect the measured corrosion rate (i.e., glasses which fracture to produce particles with high energy surfaces and sharp edges will react more than glasses which fracture to form low energy surfaces without sharp edges, regardless of the differences in the intrinsic reactivities of the glasses).

Other complications may arise when applying the test method to some materials. Non-representative samples may be taken when crushing heterogeneous waste forms due to the generation of different particle size distributions during fracturing and sieving. Interpretation to the test results may also be difficult if the relative surface areas of various phases exposed to the leachate are not known. Highly soluble phases may be partially or totally dissolved during the washing step to remove fines and so not be detected during the test.

The PCT has been standardized by the ASTM as Standard C1285-94, "Standard Test Methods for Determining Chemical Durability of Nuclear Waste Glasses: The Product Consistency Test (PCT)" [103].

2.5.1.3 MCC-3 Solubility Test

The PCT test described above is similar to the MCC-3 test, which was designed to measure the maximum solubility of a waste form in the solution of interest [86]. The MCC-3 test is a crushed glass test that originally specified that glass passing through a 325 mesh (about 40 μm) screen is to be reacted with a mass of solution ten times the mass of the glass. Tests were to be conducted until the solution composition did not vary with the reaction time (i.e., until the glass "saturated" the solution). However, application of the test method showed that (1) constant solution compositions were not achieved within a few weeks, and (2) the solution composition was dependent on the size fraction of the glass used. The first observation is a direct result of the glass reactivity, while the second is due to the different SA/V that result from the different total surface areas of different sieve fractions. As discussed in this report, tests conducted at higher SA/V are expected to generate more concentrated solutions. This was observed in MCC-3 tests conducted with PNL 76-68 glass [86]. The MCC-3 procedure has been modified such that (1) a single size fraction is specified, and (2) the test vessel is continuously agitated during the test. The PCT test has effectively replaced the MCC-3 test in most laboratories, although all of the complications associated with the use of crushed glass found during development of the MCC-3 tests are common to the PCT procedure.

The development of the MCC-3 test was based on the premise that different glasses having different compositions would generate saturated solutions having different concentrations. Crushed glass was used to accelerate the saturation of the solutions. However, the observed effects of the particle size and the SA/V on the solution composition show that unique solution compositions are not obtained, and that the solution compositions depend on the test conditions. The difference in the solution concentrations are due in large part to the fact that the glasses corrode nonstoichiometrically and that the pH achieved in the test depends on the SA/V that is used.

2.5.1.4 Argonne National Laboratory Vapor Hydration Tests

The vapor hydration test developed at Argonne is a static test in which a monolithic sample is exposed to water vapor in a sealed vessel [104-108]. At relative humidities above about 80%, a thin film of water condenses on the sample in which the glass corrodes [41]. Tests are commonly performed in 100% relative humidity at temperatures above 100°C, although tests have been conducted at lower humidities and temperatures. These tests combine the effects of high SA/V and high temperature to accelerate the corrosion process. At the completion of a test, the sample is removed from the vessel, and the reacted surface is analyzed. Corrosion of the sample results in the formation of discrete secondary mineral precipitates and alteration of the glass surface. The solution evaporates from the sample when the test is terminated and is not available for analysis. Material is left as a residue on the corroded sample as the solution evaporates. While most secondary phases form during the test, some secondary phases may form as the fluid evaporates from the sample. These can usually be distinguished by consideration of the solubilities and morphologies of the phases. (Evaporite phases are expected to contain highly soluble components, such as boron, and are usually small, poorly crystalline or amorphous, and widely distributed.) This material can be redissolved by recondensing vapor on the sample at room temperature then removing the solution using a micropipette [81], or by submerging the entire sample in deionized water, as in an MCC-1 test [109]. Both methods have shown high concentrations of soluble glass components to exist on the sample surface, though the latter method is complicated by the disintegration of the layer itself.

The extent of corrosion is determined by measuring the volume of glass that has been altered or the volume of alteration phases generated. Alteration products generally include a clay layer that uniformly covers the sample and precipitated phases that nucleate at various sites on top of or within the clay layer. The extent of corrosion is conveniently measured using the thickness of the alteration layer, which can be assumed to be proportional to the volume of glass corroded. Secondary phases precipitated on the surface are generally excluded from the measurement of the layer, since they are typically localized. Most high-level waste glasses tested to date have formed relatively uniform clay layers at the glass surface that were used to quantify the extent of corrosion [81,104-108]. The relative amounts of secondary phases formed have also been used to compare the extents of reaction [109]. Secondary phases can be analyzed *in situ* or removed from the sample for analysis and identification.

These tests provide a convenient means of generating secondary phases for analysis within a short period. A knowledge of the assemblage of phases formed is needed to simulate the corrosion process using computer models (see Section 6). In addition, these tests provide a qualitative measure of the effect of secondary phase formation on the long-term corrosion rate; that is, if the formation of secondary phases increases the corrosion rate. This information is important to final waste form development because the long-term corrosion rate depends on the glass composition and the assemblage of phases formed. From these test results, it can be determined whether a candidate final waste form will show an unacceptable increase in the corrosion rate upon secondary phase formation. Such compositions can therefore be avoided. While similar information can be obtained from PCT B tests run for very long time periods, vapor hydration provides results within a few days or weeks.

Vapor hydration tests are performed in a batch mode, in which a separate test is run for each period. Tests can be run for several different durations to determine when the secondary phases form and how they affect the corrosion rate. Vapor hydration tests can be performed at any temperature, though higher temperatures significantly accelerate the individual reaction rates (i.e., diffusion, ion exchange, and hydrolysis reactions). The rate measured at high temperatures can be extrapolated to lower temperatures if the activation energy is known, although the rate-determining step may be different at lower temperatures. Tests at lower temperature may also be required to confirm that the same secondary phases form, but the

known stability ranges of specific phases can be utilized instead. This information is incorporated into geochemical equilibration computer codes such as EQ3NR, which is being utilized to simulate long-term glass corrosion behavior [110]. However, since the EQ3NR and other equilibration codes require complete specification of the solution chemistry, application of vapor hydration data is currently restricted to identifying the phases that form experimentally for comparison to those that are predicted to form by the model [111], due to the lack of solution data. Nevertheless, identification of the assemblage of secondary phases is crucial to long-term modeling, since these phases control the glass corrosion rate. Phases identified to form in vapor hydration tests are generally different than those predicted to form by computer simulation [111,112]. The difference is likely due to the fact that the phases found to form experimentally are generally those favored kinetically while the thermodynamically most stable phases are predicted to form in the computer models. The phases formed initially will likely be replaced by more stable phases through diagenetic reactions over long time periods. In fact, paragenetic sequences have been observed in vapor hydration tests conducted over extended time periods [113,114].

Corrosion in a vapor environment represents a high SA/V system dominated by the solution chemistry, since very little glass must dissolve to saturate the thin film of sorbed water. While the geometric surface area of the specimen is easily measured, the volume of water in contact with the glass specimen will vary as the corrosion progresses. Initially, only about 50 to 100 monolayers condense on the glass [41] to form a film estimated to be about 0.01 μm thick (assuming each monolayer is about 0.15 nm thick [115]). This is equivalent to an SA/V of $1 \times 10^8 \text{ m}^{-1}$. Additional water condenses as the glass corrodes during the test, until a maximum of about 0.05 mL condenses on a sample having a surface area of about 1.5 cm^2 . (Enough water is added to the test vessel to allow 0.05 mL to condense.) If the water is evenly distributed, then the minimum SA/V is about 3000 m^{-1} , similar to that of the PCT test. However, a significant amount of corrosion will have occurred before the SA/V approaches 3000 m^{-1} in the vapor hydration test, whereas the SA/V is about 2000 m^{-1} at all times in the PCT test, so the two tests cannot be directly compared. Glass corrosion will be more progressed in vapor hydration tests than in PCT tests because of the initially higher SA/V in the vapor environment. Nevertheless, the glass corrosion path is expected to be the same in long-term PCT tests and in short-term vapor hydration tests. Indeed, hydrothermal leach tests similar to the PCT that were conducted at high temperatures have resulted in the formation of secondary phases within a year [60,116].

2.5.1.5 Buffered Solution/pH-Stat Tests

In pH-stat tests, a constant solution pH is maintained by the periodic addition of strong acid or base solutions. Tests in buffer solutions and pH-stat tests are performed to suppress changes in the solution pH as the waste form corrodes. These tests are equivalent to those performed in buffer solutions, except buffers contain counter-ions that could affect the corrosion of the waste form. Strong acid and base solutions are used in pH-stat tests to assure that the counter-ions will not interfere with the waste form.

Corrosion of borosilicate waste glasses generally results in an increase of the solution pH as a result of dealkalization. The increased pH then affects the glass corrosion rate. External control of the solution pH suppresses the natural pH change during corrosion of borosilicate glasses, although glass corrosion products are free to build up in the solution. Control of the fluid pH by materials other than the glass may be important in disposal conditions. For example, large amount of backfill material may maintain lower fluid pH values than those attained during laboratory tests conducted in the absence of these materials.

Neither pH-stat nor pH buffering constitute test methods *per se*, rather they are modifications of other test methods. The need to periodically add acid or base in the pH-stat technique may restrict application to some test methods, and the technique is not widely used in testing high-level waste glasses. The use of buffers to control the solution pH during both static and dynamic laboratory tests is common (see Section 3.3).

2.5.2 Dynamic Tests

2.5.2.1 Soxhlet Tests

Soxhlet tests provide a simple and convenient method for assessing glass corrosion rates under highly dilute conditions. Several variations of the soxhlet test have been utilized. Soxhlet tests are performed by reacting a sample with condensed fluid in a refluxing apparatus. Water is boiled from a reservoir, recondensed in a reflux tube, then allowed to drip into a small cup that contains the sample. The solution in the sample cup is refreshed as condensate (leachant) drips into the sample cup and leachate solution is displaced from the sample cup and drips into the reservoir. Complete replacement typically occurs every few minutes; the precise replenishment time depends on the refluxing conditions. The refluxing action provides an effective flow rate that can be varied by controlling the boiling of the liquid and the condensation of the vapor. Tests are usually run to attain a flow rate sufficiently high that the solution in the sample cup remains highly dilute and does not affect the corrosion rate. The corrosion rate can be measured by periodically removing a sample of the solution in the reservoir for analysis. Different reaction temperatures can be tested by varying the pressure in the apparatus to maintain boiling over a wide temperature range [69]. Because the leachant solution is recondensed vapor, only deionized water or an azeotrope solution can be used as a leachant. Soxhlet tests can be used to provide a measure of the corrosion rate in highly dilute solutions and over a wide range of temperatures. Either crushed or monolithic specimens can be used: however, test conditions must be such that crushed glass is not flushed out of the sample cup. Most tests are conducted using monolithic samples. The MCC-5 Soxhlet test specifies the use of a monolithic sample and an all-Teflon apparatus [86]. Teflon is specified to prevent contamination of the solution by Pyrex glassware.

Soxhlet tests are commonly utilized to measure the initial corrosion rate of glasses as a function of the temperature [69,117]. The glass continues to react at the forward rate because the leached glass components accumulate in the reservoir as leachate solution drips from the reaction cell, while the freshly distilled leachant accumulates in the reaction cell. The fluid contacting the sample remains highly dilute, thus maintaining the forward corrosion rate, while the fluid in the reservoir becomes concentrated, which facilitates analysis. Soxhlet tests have also been performed for long corrosion times on a single sample by periodically replacing the solution in the reservoir with fresh deionized water [118]. Soxhlet tests have also been used to characterize the alteration of the surface after extensive corrosion [119]. Note that the alteration phases formed during the Soxhlet tests are due to differences in the solubilities of different glass components and are not necessarily identical to secondary phases formed from the saturated leachate in static tests. The leachate solution in contact with the sample in Soxhlet tests remains highly dilute.

2.5.2.2 Single-Pass Flow-Through Tests

In single-pass flow-through tests, leachant solution is passed over a monolith surface or through crushed glass at a constant rate. The sample is contained in a reaction cell that can be modeled as a well-stirred reactor with leachant solution entering the cell and leachate solution exiting the cell at the same rate. Solution is pumped through the cell by a peristaltic pump; the solution leaving the cell is periodically collected and analyzed. The single-pass flow-through apparatus designed at Lawrence Livermore National Laboratory has been

described by Weed et al. [120] and by Coles [121,122]. The corrosion rates of glass waste forms are known to increase with the leachant flow rate up to a maximum corrosion rate, which is the forward reaction rate (see Section 5). Low flow rates can be used to simulate conditions in particular disposal sites. Alternatively, solution can be pumped through the cell at a rate high enough that the glass reacts at its characteristic forward rate. The forward rate has been reached if further increases in the flow rate do not increase in the corrosion rate. The forward corrosion rate is an important characteristic of waste forms that is needed for computer simulations of corrosion behavior (see Section 6).

The forward rate represents the maximum corrosion rate of the waste form at a particular solution pH and temperature. The corrosion rate is known to decrease as silicic acid and other glass components become concentrated in the solution. The leachant solution used in a single-pass flow-through test can be varied to measure the effects of the solution composition on the forward reaction rate. Such tests have been applied to determine the effects of pH, and of silicon, calcium, aluminum, and other glass components on the glass corrosion rate [70,71]. In addition, the reaction cell can be placed in an oven to determine the effects of temperature on the corrosion rate. Such tests are valuable in assessing the corrosion mechanism of waste forms, and they provide values for parameters needed to simulate their long-term corrosion behavior (see Section 6).

Flow tests are usually conducted for several days at a constant flow rate, with samples being collected periodically until the analyses show that a constant (sometimes referred to as "steady-state") rate has been reached. Determination of the corrosion rate requires the glass surface area be known at all times. Tests are complicated by the need to maintain negligible concentrations of corrosion products at the glass surface but to generate leachates that can be analyzed to monitor the reaction rate. The test data must also be interpreted to account for the effects of the initial surface finish of monolithic samples, the presence of fines, the reduction in surface area as the glass corrodes, and the possible formation of surface layers and precipitates. Fines present on the freshly prepared sample provide a very high initial surface area that usually cannot be quantified, and so data from the initial reaction is usually ignored. Tests must also be conducted so that the loss of surface area during the test is negligible. Therefore, crushed glass samples should be sufficiently large that the surface area does not decrease significantly over the test duration. Monolithic samples provide a readily measured but small surface area. Crushed glass samples provide a higher surface area, but the surface area is difficult to quantify. Crushed glass samples are also more likely to contain fines. Also, the surface area of crushed glass samples decreases faster than that of monolithic samples. The loss of surface area will lead to a decrease in the measured corrosion rate over time. The loss of surface area can be determined by directly measuring the reacted glass or estimated based on the solution analyses and the test duration. Therefore, intermediate reaction times most faithfully represent the corrosion rate, while results from short times are complicated by fines and the results from long times are complicated by the decrease in surface area. In practice, rates are computed from data showing "steady state" behavior, i.e., for which the solution concentrations remained nearly constant with time [70,71].

The large volumes of leachant required and the large volumes of leachate solution generated may preclude use of this test in cases where a limited volume of leachant is available, such as when groundwaters are used, or where disposal of the leachate solution is of concern, such as when radioactive glasses are used.

2.5.2.3 Periodic Replenishment Tests

Several test methods have been developed in which leachate solution is periodically removed from an ongoing static test and replaced with an equal volume of fresh leachant solution. Such replacement tests can be used to simulate very low flow rates that cannot

be attained by using mechanical pumps [123,124]. Either monolithic or crushed samples can be used, and different leachant compositions can be used. Specific test methods have different replacement schedules and call for different fractions of the total solution volume to be replaced. For example, the test designated by the International Organization for Standardization (ISO) calls for the entire solution volume to be replaced daily for the first week, every week for eight weeks, monthly for six months, and twice yearly thereafter [125]. The American Nuclear Society test ANS 16.1 and variations of the ANS 16.1, such as the Dynamic Leach Test (DLT) and the Accelerated Leach Test (ALT), also require the total solution volume of leachate to be exchanged periodically. The variations differ in the replacement schedules. For example, the ALT calls for replacement of the entire solution after 2, 5, and 17 hours and then daily for 10 days. The total accumulated contact time in the ALT is 11 days [126]. Other replacement tests replace only a small fraction of the leachate that is removed for analysis [127].

The effect of the solution replacements on the corrosion of the waste form depends on the amount of leachate replaced and on the frequency of the replacement. If the solution is refreshed often and completely, as in the initial stages of the ISO and ALT methods, reaction products will be prevented from accumulating in the solution and the solution will remain dilute. The solution chemistry will have a minor effect on the corrosion of the waste form under these conditions. These tests will be similar to static tests with very low SA/V. If only a small amount of leachate is replaced, reaction products will accumulate and the solution will approach saturation. Under these conditions, the corrosion rate will slow between refreshments but then attain a slightly higher rate after refreshment until the solution returns to near saturation.

Differences in the presentation of test results in terms of an incremental leach rate and the cumulative leach rate, as in the ISO method have been discussed by Ogard and Bryant [128], who caution against possible misinterpretation of test results. Plotting the rate against the reaction time increment does not account for when a particular time increment occurred. In effect, the sample is assumed to be unaffected by previous leaching increments. This assumption is not valid for most waste glasses, which dissolve nonstoichiometrically under most conditions. Formation of an altered surface layer is known to affect glass corrosion, though only slightly for most waste glasses (see Section 3.1). The leach rates measured between samplings will be affected by the duration of the leaching increment because of the feedback effects of corrosion products, which tend to decrease the corrosion rate. Plotting the extent of reaction against the reaction time increments will group rates obtained at common durations and more clearly show the effects of changes in the solution composition. In contrast, plotting against the cumulative time will more clearly show the effects of the generation of a surface layer.

The perturbing behavior of leachate replacement is shown in Fig. 7, where the boron concentrations measured over seven exchange intervals with $\Delta V/V = 0.25$ and $\Delta t = 28$ days ($T_r = 112$ days) are plotted against the cumulative exposure time [127]. The boron concentration is seen to rise over each interval and then decrease by about 25% upon replenishment. The observation that the maximum and minimum concentrations reach nearly constant values after three exchanges (four exchanges corresponds to complete volume replacement under these conditions) indicates that the corrosion layer, which is assumed to continually increase with time, has a negligible effect on the corrosion rate. The perturbations seen in the boron concentration due to leachate replacement will similarly affect the silicic acid concentration and the pH. Indeed, it is probably the periodic reduction in the silicic acid and the pH that affect the glass corrosion rate and lead to the cycling in the boron concentration (see Section 3).

Fig. 7. Measured Boron Concentrations in 28-Day Replacement Tests (after [127]).

Refreshment of the corroding fluid may significantly affect the glass corrosion mechanism by preventing the solution from becoming saturated for long periods, and long-term tests conducted in static solutions and those conducted with partial replenishment may show very different glass performance. Leachant replacement results in a cycling of the corrosion process as the control over the corrosion rate alternates between the glass and the solution chemistry under highly dilute and more concentrated solutions, respectively. These perturbations may have a significant effect on the observed corrosion behavior (see Sec. 5.0). Even small perturbations of an otherwise static test may prevent precipitation of secondary phases and progression of the corrosion into the long-term stage. Replacement tests cannot be conveniently used for characterizing the long-term corrosion process because of uncertainties about the effects of the solution perturbations. They are better used as response tests to index durabilities of candidate waste forms relative to a benchmark of performance. Even for comparative purposes, it is important that the same replacement schedules be followed.

An effect that has been observed in partial replenishment tests that has not been observed in continuous flow tests is transient leach rate excursions [129,130]. These excursions are characterized by a sudden temporary increase in the leach rate. Barkatt et al. have speculated that these excursions are due to cracking of the surface layer and sudden exposure of fresh glass, but have not provided convincing evidence. They also point out that the excursions are transient and do not appear to significantly affect the corrosion rate of typical high-level waste glasses (see Sec. 3.1).

2.6 Measurement of Surface Area

Most test procedures require a measurement of the glass surface area used in the test. The glass surface area that is available for reaction must be known to determine the reaction rate from the measured solution compositions in all tests methods. Monolithic samples may be prepared with regular shapes that can be measured with calipers. Finely crushed glasses have high surface areas, which can be measured by means of gas adsorption. In this technique, the amount of an inert gas that condenses on a sample of powdered glass is measured at a temperature near the boiling point of the gas. The amount of gas condensed on the sample is

measured by the pressure or volume change in the system upon exposure to the sample, or by direct weighing of the sample before and after sorption. When one monolayer of gas is sorbed onto the glass, the surface area of the glass covered by gas can be calculated from the known size of the gas molecule or atom and the amount sorbed. Inert gases such as nitrogen and krypton are usually used for surface area measurements. What is measured in these analyses is the surface area accessible to nitrogen or krypton, which may be different than that accessible to water molecules. The surface areas measured using different gases may also be different. For example, the surface areas measured using adsorption of N_2 was about twice that measured using adsorption of Kr [131]. The area measured by the adsorption of Kr was about 15 times the area estimated using the sieve size. These gases are larger than water and so may not contact glass in small pores or channels that are accessible to water. Of course, geometric measurement of monoliths will not include pores or cracks in the glass, either. The surface area that is available during the reaction may be greater than that measured geometrically or by gas adsorption.

The surface area accessible to water may be measured by the sorption of water vapor directly. The interaction of water vapor with the glass surface may play an important role in the reactivity of waste glasses in a hydrologically unsaturated repository, where the glass will likely be contacted by water vapor before liquid water reaches it. Water vapor is known to chemisorb strongly onto surface silanol groups even at very low water vapor pressures. Sorption onto other sites is generally weak. Water sorbs initially onto the silanol sites and then forms small drops of several molecules that are anchored to the glass surface by the silanol group. As more water condenses, the drops coalesce to minimize the water surface area and form a thin film covering the surface. The surface density of silanol groups depends on the glass compositions and has an upper value of about 4.6 silanol groups/nm² in silica gel [40]. As the silica content of the glass decreases, the surface density of silanol groups will also decrease. Sorption studies on a typical waste glass composition showed that increased amounts of water may condense as a result of reaction between the glass and the sorbed water [41]. Exposed alkali metal ions can ion exchange with condensed water if enough water is present to hydrate the released ion. The increased hygroscopicity of the condensate will result in more water condensing on the glass.

Alternatively, the grain sizes of crushed glass can be approximated on the basis of the specific sieve fraction used in the test by assuming the grains have a simple geometry, such as cubic [132], spherical [133], or tabular [134]. The specific shape does not significantly affect the computed surface area. In this approach, the average dimension of the particle is taken as the average mesh opening of the sieve fraction used, and the distribution of the particles is assumed to be Gaussian and centered on the average mesh size. The surface area per unit mass is then computed on the basis of the glass density.

For example, consider crushed glass in the sieve fraction of -100+200 mesh where the particles are approximated to be spheres. The opening of the 100 mesh sieve is 0.149 mm and the opening of the 200 mesh sieve is 0.74 mm. The arithmetic average opening is 0.112 mm. The volume and surface area of a sphere can be readily calculated as $\text{volume} = \pi d^3/6$ and $\text{surface area} = \pi d^2$, where d is the diameter. A sphere having a diameter of 0.112 mm has a volume of $7.4 \times 10^{-7} \text{ cm}^3$ and a surface area of $3.9 \times 10^{-4} \text{ cm}^2$. If the density of the glass is assumed to be 2.7 g/cm^3 , then there are 5×10^5 glass particles per gram and $2 \times 10^{-2} \text{ m}^2$ of surface area per gram of glass. Larger size fractions will have smaller surface areas per gram and smaller size fractions will have larger surface areas per gram. Glasses with densities greater than 2.7 will have less surface area per gram and glasses with densities less than 2.7 will have more surface area per gram.

A photomicrograph of glass in the -100+200 mesh sieve size fraction is shown in Figs. 8a and 8b for two different glasses. It is not obvious that the particles of either glass could be well represented by cubes, spheres, or tablets. The crushed glasses shown in Figs. 8a and 8b also have different particle habits. The particles of crushed WV 6 glass are relatively uniform in size and shape. The particles are generally free from sharp edges, having been rounded when crushed. Three distinct morphologies of SRL EA glass particles can be seen in the photomicrograph: chunk-like particles with a somewhat spherical shape, rod-like particles, and plate-like particles. The particles of SRL EA glass have sharper fracture points and edges than the particles of WV 6 glass. The WV 6 particles are all of a similar size, whereas the SRL EA particles have a wider size distribution, and are generally bigger than the WV 6 particles. Clearly the crushed WV 6 and SRL EA glasses have different surface areas per unit mass. In addition to differences in the total surface area, the particles with sharper edges and points are likely to be more reactive than the particles with more rounded surfaces simply due to the higher surface energy of the sharper particles. The fracture behavior depends largely on the cooling of the glass from the melt: rapidly quenched glasses are expected to fracture to form particles with sharper edges than annealed glasses.

The exposed surface areas of various size fractions have been compared experimentally in terms of the extent of corrosion under test conditions that are sensitive to the surface area. Fig. 9 shows the sodium concentrations in solutions attained in tests using different size fractions of crushed glass. In these tests, glass mass/solution mass ratios were selected to yield a common SA/V, namely 77 m^{-1} , based on a geometric approximation of the particle surface area [135]. The results show that the sodium release is the same for all size fractions, within experimental error, and that the glass corrosion rate is not affected by the particle size over this time period. Longer reaction times are expected to show particle size effects because smaller particles will lose surface area faster. These results imply that the geometric approximation is sufficient, compared to the testing errors.

Several studies have compared the surface area estimated by sieve size to that measured by adsorption based on the experimentally measured extent of reaction. For example, Oversby compared the extent of corrosion of various sieve fractions of crushed SYNROC in modified ISO tests at 90°C [131]. The surface areas of the different size fractions were measured by Kr adsorption and the geometric surface area was calculated by assuming the grains to be cubes having an edge length equal to the average sieve opening. Tests were conducted for one, two, and three days, where the entire leachate volume was replaced and analyzed daily. Tests were conducted using the sieve fractions listed in Table 3. The surface area measured by adsorption, the surface area calculated by assuming the grains to be cubes, and the approximate SA/V (based on the geometric surface area) are given for each size fraction.

Oversby reasoned that all size fractions should corrode at a similar rate under these conditions. The leach rates of aluminum, calcium, sodium and silicon calculated using the geometric surface areas are consistently higher for tests conducted at lower SA/V ratios. The sodium release rates are included in Table 3. The differences are consistent with the effects of the SA/V ratio observed in other tests. Using the surface areas measured using gas adsorption, SA (ads), which were higher than the geometric surface areas, SA (geom.), amplifies the differences in the different size fractions. Oversby concluded that the smaller differences calculated using the geometric surface areas were more consistent with her expectation that all size fractions would corrode at a similar rate. She estimated that the "real" surface area of the crushed glass was about 1.5 times that calculated geometrically due to the grain shapes.

(a)

(b)

Fig. 8. Photomicrographs of -100+200 Mesh Size Fraction of Crushed Glass

Fig. 9. Test Results for Different Size Fractions of Crushed Glass: (□) -4+10 Mesh, (■) -45+60 Mesh, and (○) -325+400 Mesh (after [134])

Table 3. ISO Test Results [131]

Size Fraction, mm	SA (ads.), ^a cm ²	SA (geom.), ^b cm ²	SA (ads.)/V, m ⁻¹	NL(Na) ^c
177-246	187.7	3.16	20	5.6
246-417	27.0	3.93	24	5.0
417-840	39.3	4.94	31	3.7
840-1650	69.2	6.95	44	2.1

^aSurface area measured by gas adsorption.

^bSurface area estimated based on sieve size.

^cIn g/m²d.

Oversby also compared the results of a 3 day MCC-1 test using a monolithic sample having a known geometric surface area to the results of a one day ISO test [131]. The leach rate measured in the MCC-1 test was in good agreement with the results of the ISO test where the "real" surface area was used (that is, 1.5 times the surface area calculate by assuming cubic geometry).

Tests were conducted by Barkatt et al. to compare the reactivities of monolithic samples to that of crushed samples [136]. Tests were conducted by placing a monolithic sample of measured geometric surface area and a known mass of crushed glass samples of the size fraction -42+60 mesh in a Teflon reaction vessel with deionized water and placing the sealed vessel in a 70°C oven. Enough water was added in an effort to maintain low SA/V conditions throughout the test periods. The water was replaced at frequent intervals and analyzed. Tests were conducted using three different glasses, PNL 76-68, and unidentified CUA and SRL glasses. By comparing the silica concentrations measured after several reaction intervals in tests with monolithic with that measured in tests with crushed glass, Barkatt et al. concluded that the effective surface area of the crushed glass in the size range -42+60 was $97 \pm 11 \text{ cm}^2/\text{g}$. This is in good agreement with the value calculated assuming spherical particles, which is about $117 \text{ cm}^2/\text{g}$ (average mesh opening is $190 \mu\text{m}$). The range of silica concentrations attained in the tests ranged between 1.65 and 11.1 ppm in tests with the PNL 76-68 glass, between 0.09 and 0.59 ppm in tests with the CUA glass, and between 1.92 and 42.3 ppm in tests with the SRL glass. The higher concentrations attained in tests with the SRL glass may have reduced the corrosion of both monolithic and crushed glasses slightly. However, similar concentrations were attained in tests with monolithic and crushed glasses, so the comparison of surface areas is expected to be unaffected.

Using the measured extent of reaction to estimate the surface area of crushed glass is based on the premise that tests at similar products of the SA/V and the reaction time generate equivalent solution compositions. This approach is best applied by using tests that are dominated by the initial stages of corrosion and maintain dilute solution conditions, under which the build-up of glass corrosion products has a negligible effect on the corrosion rate.

Because of the uncertainty in the measuring the precise surface area of crushed glass, some test protocols (for example, the PCT) specify only the size fraction and mass of crushed glass to be used, rather than the surface area or the SA/V. Crushed glass is sieved to isolate a specific size fraction, which is carefully washed to remove fines from the particles. (The PCT procedure includes the option of not washing the crushed sample to remove fines if the presence of highly soluble included phases is anticipated or determined. Of course, the presence of fines will then likely result in higher solution concentrations of all elements present in the fines. The trade-off of losing soluble material when washing fines from the sample or including the effects of fines in the test result is a consequence of using crushed glass. It may be possible to wash the crushed glass with a nonaqueous solvent in which the phases have low solubility.) It is assumed that the total surface area per mass of crushed glass is reproducible for a given size fraction. The fracture behavior probably depends on the glass composition and the method used to crush the glass. Tests can be performed at different SA/V either by using crushed glass of different size fractions or by using different masses of crushed glass of the same size fraction. While the SA/V attained is only approximate for either method, using the same size fraction allows direct comparison of tests at different relative SA/V, since the area per gram, while it may not be known very accurately, can be expected to be constant. That is, the error introduced by using sieve size to approximate the surface area per gram of glass will cancel in test results at different $\text{mass}_{\text{glass}}$ to $\text{mass}_{\text{solution}}$ ratios.

2.7 Effect of Surface Finish

The actual surface area of a monolithic glass sample that is available for reaction in a laboratory test will be affected by how the sample is prepared. Different surface finishes may be achieved by cutting with various wafering blades, grinding with abrasives, fracturing, flame polishing, etc. The microscopic surface areas generated during sample preparation may differ significantly among samples that have the same geometric surface areas but were prepared differently. For example, it has been reported that abrasion with 100-grit increased the surface area of PNL 76-68 glass available for reaction by about 33% and abrasion with 600-grit increased the surface area by about 17% over the geometrically measured surface area [137].

The release of glass components during corrosion will increase with the surface roughness because of both the greater area and the higher strain. Because the strain induced by the abrasion penetrates into the glass to a finite depth, corrosion becomes equivalent after the corrosion front penetrates to that depth. The effects of surface finish are expected to be most important during the initial stages of glass corrosion and to diminish as the corrosion process proceeds. Tests used to measure the effects of surface finish and surface strain on glass corrosion must be sensitive to the initial stages of glass corrosion and minimize the effects of the solution chemistry on the corrosion. Such tests must maintain dilute solution conditions and be dominated by the reaction of the topmost millimeter of glass, and typically they must be short-term tests (on the order of a few days).

Differences in reactivities have been measured for samples with different surface finishes. In Soxhlet tests with R7T7 glass, the normalized boron mass loss was higher by a factor of about two for a saw-cut sample compared to the loss for a sample polished with 1 micron diamond powder [138]. In MCC-1 leach tests with PNL 76-68 glass, the boron loss was higher by factor of about 3 for glass ground to 100-grit finish compared to loss for a sample polished with 1 micron diamond powder [137]. The differences between samples polished with 1 micron diamond powder and samples ground to a 600-grit finish were small (less than about 20%) for both glasses. The soxhlet and MCC-1 test methods highlight the initial stages of glass corrosion during which the effects of materials released to solution on the continued glass corrosion are small, and emphasize the effects of the glass surface area. These test results indicate that the higher surface areas of the more coarsely ground surfaces (i.e., the saw-cut sample and the sample with a 100-grit finish) generate higher solution concentrations than the samples with the finer polish (1 micron finish).

Tests were performed by Bates et al. to assess the importance of surface strain to glass reactivity in a water vapor environment at elevated temperatures [139]. Glass samples were either polished, fractured thermally, or fractured by percussion. Some glasses were thermally annealed after fracturing to partially relieve the stresses induced during fracturing. It was found that the annealed samples and those polished to 600-grit were the least reactive and that the fractured and 320-grit samples reacted similarly. Thus, both stresses and increased surface area promoted corrosion under these conditions.

Fractures may occur at the sample surface during mechanical preparation of monolithic samples (core drilling, wafering, etc.). These fractures may be removed by subsequent annealing or grinding of the surfaces. The effect of such surface fractures on test results depends on the crack width and whether water can enter the crack. An investigation of the effect of various crack widths was carried out by Perez and Westsik following a MCC-1 type test procedure [140]. Various crack sizes were simulated by placing different sized platinum wires between monolithic samples. They found that leaching from cracks was less than that from free surfaces, and that leaching was nil from tight cracks. However, analysis of corroded monoliths in cross-section have shown dealcalization to occur to similar depths around surface cracks and at free surfaces [73]. This suggests that solution analyses are not sufficiently sensitive to distinguish the effects

of leaching from cracks. Surface analysis also shows that little or no etching occurs in the crack, while significant etching may occur on free surfaces. Thus, material remains contained within the crack as the glass corrodes. Insight into the effects of cracks is provided by an understanding of the effects of the solution chemistry on glass corrosion. Of course, the effects of the cracks will disappear as the corrosion front penetrates into the glass deeper than the cracks.

2.8 Measurement of Leachant Volume

Measurement of the leachant volume is trivial in static leach tests because the amount of leachant is added to the reaction vessel either by volume or by weight. However, several test procedures will be discussed for which the leachant volume or effective leachant volume is not directly measurable. These will include partial-replenishment tests, in which the leachate is periodically refreshed; continuous flow tests, in which the leachant is continuously flowing past the glass; and static tests in water vapor, in which the leachant is condensed on the sample from the vapor phase. The dynamic tests require the definition of a contact or residence time in which a measurable volume of solution tends to react with the glass in a quasi-static system. Alternatively, the time required to completely replace the leachate solution in a confined volume can be determined.

In a vapor system, water may condense on the sample to form a film many monolayers thick. (A monolayer is a film one molecule thick covering the surface.) The amount of condensed water depends on the glass surface composition, the water vapor pressure, and the temperature. If the sorbed water is corrosive towards the surface, for example, if dealkalization occurs, a hydrophilic solution may be generated that will increase the amount of water that is condensed. While the amount of water condensed at ambient temperatures can readily be measured [41], condensation at higher temperatures is not easily quantified. Instead, the maximum volume of water that can be supported on the sample without dripping off can be approximated and used as the leachant volume.

Initially, only about 50 to 100 monolayers condense on the glass [41] to form a film estimated to be about $0.01\ \mu\text{m}$ thick (assuming each monolayer is about $0.15\ \text{nm}$ thick [115], i.e., equals the diameter of a water molecule). The formation of this layer occurs rapidly [41]. For a sample having a surface area of about $2\ \text{cm}^2$, about $2 \times 10^{-6}\ \text{mL}$ of water would be required to form a layer $0.01\ \mu\text{m}$ thick. The corresponding SA/V can be easily calculated by considering the $0.01\ \mu\text{m}$ thick layer to form on a plane surface, such that the volume of water per square meter is about $1 \times 10^{-8}\ \text{m}^3$ and the SA/V is about $1 \times 10^8\ \text{m}^{-1}$. Additional water condenses as the glass corrodes and releases alkali metals and other glass components into the film during the test. The amount of water on the glass increases steadily until a maximum of about $0.05\ \text{mL}$ condenses on the sample. Only enough water to allow $0.05\ \text{mL}$ to condense is added to the test vessel. Condensation of more than $0.05\ \text{mL}$ is likely to result in water dripping from the sample. For a sample having a surface area of about $1.5\ \text{cm}^2$, and assuming that the $0.05\ \text{mL}$ of water is evenly distributed, the minimum SA/V achieved during the test is about $3000\ \text{m}^{-1}$.

The actual SA/V of the system will decrease from the values calculated from the initial surface area of the glass and the amount of water added because some of the water will diffuse into the glass as corrosion occurs. This water is no longer available to dilute corrosion products. Alteration layers on the surface of corroded high-level waste glasses are generally found to include about 4 to 7 wt% water [46]. Assuming the layer to contain 7 wt% water, the density of water in the layer of a glass having an initial density of $2.7\ \text{g/cm}^3$ would be about $0.2\ \text{g/cm}^3$. Formation of a $10\ \mu\text{m}$ thick layer would consume about $2\ \text{mL}$ of water per square meter ($2 \times 10^{-5}\ \text{mL/cm}^2$). For a sample having a surface area of about $2\ \text{cm}^2$, about $4 \times 10^{-5}\ \text{mL}$ of water would be consumed by formation of a $10\ \mu\text{m}$ thick layer. Note that this is about 20 times

the water needed to form the initial 10 μm thick film on the glass surface, but is negligible compared to the formation of a 0.05 mL drop of water as corrosion proceeds. Presumably, water entering the glass is replaced by freshly condensed glass as alkali metal ions are released into the surface film.

Water is also consumed as crushed glass corrodes. If the alteration layer contains about 4 to 7 wt% water, then the maximum amount of water consumed can be calculated from the known volume of glass in the test. For a PCT A test, 1 gram of glass is added to 10 g of water. About 0.07 g of water is required to hydrate all the glass to 7 wt% water. This is negligible compared to the 10 g added to the test.

2.9 Changes in SA/V as the Glass Corrodes

Changes in either the surface area of the glass available for reaction or the volume of solution available to dilute the corrosion products as the reaction progresses will change the SA/V of the system. Changes in the solution volume can usually be assumed to be negligible, as discussed in the preceding section. The surface area may either decrease or increase as the glass corrodes. Consider the simple case of spherical particles. The initial radius is given by r_i and the radius after corrosion is given by r_f . The initial surface area is simply $4\pi r_i^2$ and the final surface area is simply $4\pi r_f^2$, so that the net loss in surface area is $\Delta A = 4\pi(r_i^2 - r_f^2)$. The volume of material dissolved is given by $\Delta v = (4/3)\pi(r_i^3 - r_f^3)$, and the mass dissolved is given by $\Delta v\rho$, where v is the volume of the particle and ρ is the density of the glass. Since the differential change in the volume decreases as the square of the change in radius ($dV/dr = 4\pi r^2$) and the differential change in the surface area decreases proportional to the change in radius ($dSA/dr = 8\pi r$), the volume (and mass) of the particle will decrease faster than the surface area (until the radius is less than 2). The SA/V of the system will change directly proportional to the change in the surface area as the glass corrodes, assuming that the solution volume remains constant. Therefore, the initial SA/V can be defined as $4\pi N r_i^2/V$, where N is the number of particles and V is the solution volume, while the final SA/V is $4\pi N r_f^2/V$. The initial SA/V is greater than the final SA/V.

The decrease in surface area is illustrated by the photomicrograph of a crosssectioned corroded particle of SRL EA glass in Fig. 10. The glass was reacted in tuff groundwater at 90°C at an SA/V ratio of 20,000 m^{-1} for 98 days. The inner core of material is pristine SRL EA glass and the outer crust has been depleted of soluble glass components. The crust was formed as soluble glass components dissolved and sparingly soluble components formed the outer layer. As the particle continues to corrode, soluble elements are released from the surface of the inner core of glass, which has a much smaller surface area than the original particle.

The model of the glass as a dissolving sphere and the analytical relation between the radius, surface area, and volume has the important consequence that if the glass dissolves at a constant rate (i.e., the mass loss is proportional to t^1), then the surface area (and the radius of the particle) will decrease proportional to the square of time (t^2). Conversely, if the radius decreases linearly over time, the mass loss will decrease proportional to the square root of time ($t^{1/2}$). Hence, the observed time dependence of the corrosion rate provides evidence of the rate-controlling phenomenon: if the corrosion rate is controlled by alteration of the glass, through processes such as diffusion through alteration layers, then the radius of the particles is expected to decrease linearly with the reaction time and the mass loss is expected to decrease with the

Fig. 10. Photomicrograph of Corroded SRL EA Glass Grain

square root of the reaction time. If the corrosion rate is controlled by the solution, then the mass loss is expected to increase linearly with the reaction time, and the radius of the particle (and the surface area) will decrease as the square of the reaction time.

For example, the results of tests conducted at ANL show the boron concentration to increase linearly with respect to time in tests conducted with SRL 202 glass at an initial SA/V of 20,000 m⁻¹ for 364 days or longer (see Section 4.2). This indicates that the volume of glass corroded was constant with respect to time and that the radii of the glass particles decreased at an ever increasing rate (proportional to t^2). Since any mass transport effects would result in a root time dependence, the corrosion layers are seen not to affect the long-term corrosion rate. Instead, the long-term corrosion rate is controlled by the solution chemistry.

Nonuniform dissolution can lead to the formation of pits and pores and a net increase in the surface area. Nonstoichiometric corrosion can also lead to an increase in the effective surface area for some glass components. For example, boron may be depleted from the glass to a depth corresponding to the surface for r_f , while much of the silicate structure remains in the boron-depleted region. Thus silicon-oxygen bonds are exposed to water throughout the altered volume, while boron-oxygen bonds are exposed only on the surface at r_f . While the number of reaction sites for boron release continually decreases as the glass corrodes, the number of reaction sites for silicon release continually increases.

Knowledge of the maximum possible values of the normalized mass loss function is important when conducting long-term tests to (1) recognize effects of shrinking particles, and (2) avoid having glasses corrode too much (or completely). Glasses that completely corrode during the test cannot be used to determine the corrosion rate, since it cannot be determined when the corrosion was complete. The maximum possible normalized mass loss values can be calculated by assuming the grains to be spheres of uniform diameter. Consider the complete

reaction of 1 gram of -100+200 mesh glass containing 3 elemental wt% boron to completely dissolves in 10 mL of deionized water. The surface area of the size fraction is $2 \times 10^{-2} \text{ m}^2/\text{g}$. The normalized mass loss based on the release of 0.03 g boron can be calculated as

$$\text{NL(B)} = \frac{\left(\frac{0.03 \text{ g B}}{10 \text{ mL}} \right)}{\left(\frac{1 \text{ g glass}}{10 \text{ mL}} \right) (2 \times 10^{-2} \text{ m}^2/\text{g}) \left(\frac{0.03 \text{ g B}}{\text{g glass}} \right)}$$

which equals $1/(2 \times 10^{-2} \text{ m}^2/\text{g}) = 50 \text{ g/m}^2$. The maximum NL(B) depends on the size fraction of the glass but is independent of the SA/V. Smaller size fractions have larger maximum NL values. A similar calculation can be performed for monolithic samples. For a right cylinder, the surface area per gram is $1 \times 10^{-3} \text{ m}^2/\text{g}$ for a glass density of 2.7 g/cm^3 and the maximum NL value is 1000 g/m^2 .

The loss of surface area during the test is usually not accounted for when computing corrosion rates from test data, though it is clearly important when evaluating the corrosion rates of poorly durable glasses or of durable glasses over very long time periods. The solution results are normalized to the surface area to obtain a measure of the rate that is independent of the glass surface area, such that rates measured in tests with different initial surface areas can be compared. Because the surface area of the starting material is used, the normalized mass function only describes the integrated extent of dissolution, i.e., the average rate between the start of the test and the point at which the solution concentration is analyzed (see Sec. 2.3). The normalized mass losses computed after two different reaction times may not provide a valid approximation of the instantaneous corrosion rate since the surface area of the glass will be less than that at the start of the test. The difference may or may not be significant, depending on the reaction times and the corrosion rate of the glass. For example, the perimeter of the remaining glass core in Fig. 10 is less than half that of the outer crust. The corresponding surface area of the core is certainly less than half that of the original surface. Expressing the continued corrosion rate in terms of the normalized mass loss, which is calculated using the original surface area, clearly underestimates the corrosion rate. An appropriate means of estimating the effects of decreasing surface area on the long-term corrosion rate of crushed glass samples needs to be developed.

3.0 OBSERVED EFFECTS OF THE SOLUTION CHEMISTRY ON GLASS CORROSION

The composition and chemistry of the fluid that contacts a high-level waste glass will affect the glass corrosion rate and the release of glass components into solution. The chemistry of the groundwater that may eventually enter the repository will be determined initially by its interactions with the atmosphere and various phases in the host rock, then with engineering components such as metal containers and backfill materials, and by radiolysis of the gas phase and the groundwater itself. The amounts of dissolved air, iron, and other redox-sensitive species are likely to establish the redox potential of the fluid which initially contacts the waste glass [141]. The chemistry of the solution in contact with the glass will change considerably as the glass corrodes and glass components are released into solution. These changes will affect both the corrosion rate of the glass and the disposition of various glass components as the glass reacts. The relative importance of interactions with the surrounding environment and the corroding waste glass to the fluid chemistry will depend strongly on the volume of groundwater that contacts the waste form; the chemistry of small volumes of groundwater will be more affected by interactions with the waste form than that of larger volumes. Since a key design goal of engineered high-level waste repositories will likely be to minimize the amount of groundwater that may contact waste, it may be assumed that the fluid chemistry will be dominated by glass corrosion products. This may not be the case if the entering fluid is a brine, however. In order to assess the long-term corrosion behavior of waste glasses, it is necessary to understand how their long-term corrosion is affected by the chemistries of both the fluid initially contacting the waste and that generated after interaction.

Static leach tests commonly show the amounts of glass components released into solution to decrease in a parabolic fashion with the reaction time (i.e., approximately in proportion to the square root of time). As described in Section 2.4, the interaction of a high-level waste glass with an aqueous solution results in the dissolution of the soluble glass components and the formation of alteration layers containing less soluble glass components at the glass surface. Both the solution composition and the alteration layer may affect the continued rate of glass corrosion. The decrease observed in the corrosion rate over time may be due either to mass transport through the layers that develop as the glass corrodes or to the concentrations of glass components in the solution, which slow the individual reactions releasing those components.

It should be emphasized that concentration gradients are likely to exist in the solution, especially when layers form. A nearly static fluid having a chemistry that may be very different from the bulk solution will exist within the layer and at the surface of the glass [142]. While the composition of the bulk solution is analyzed in laboratory tests, it is the chemistry of the static fluid at the glass surface that will affect the corrosion rate. While the chemistry of the static fluid may be difficult to measure, possible differences should be borne in mind when modeling glass corrosion behavior.

Static leach tests have also shown high-level waste glasses to dissolve nonstoichiometrically. This may be due to the incorporation of some elements into secondary phases [143], or simply to different release kinetics. Several models have been proposed based on these observations, including models based on mass transport dominance [18,75,119,144-148] and surface reaction dominance [149-154]. Both mass transport and surface reaction effects probably affect the corrosion of most glasses, although one or the other effect may be dominant for specific glass compositions or reaction conditions.

3.1 Tests Conducted to Distinguish between Mass Transport and Surface Reaction Effects

Consideration of solution results alone is not sufficient to distinguish whether mass transport or chemical reaction effects dominate measured corrosion behavior, since static leach test data can be fitted equally well by both diffusion and surface reaction models [143]. Instead, other tests have been performed to make this distinction. These tests typically involve comparison of the extent of reaction of glass samples in two sets of conditions: (1) with and without the physical removal of alteration layers and (2) with and without removal of corrosion products from the solution through leachant replacement. If the surface reactions control the glass corrosion rate, then changes in the solution composition as the glass corrodes are expected to dominate the corrosion rate, and the presence or absence of an alteration layer should not affect the measured extent of reaction. If mass transport controls the glass corrosion rate, then the presence or absence of an alteration layer is expected to dominate the corrosion rate, and changes in the solution composition will have little effect.

Chick and Pederson conducted tests based on the MCC-1 protocol and three variations of it test to assess the effects of the solution composition and mass transfer through the alteration layer on the corrosion rate [155]. The compositions of the three glasses used are given in Table 4. In the first series of tests, glass A was reacted in deionized water under MCC-1 conditions for several reaction times between 7 and 56 days. The measured boron release in these tests is shown as filled circles in Fig. 11. The normalized boron mass loss increased rapidly over the first few days and then leveled off at longer reaction times. In the first variation of the MCC-1 test, samples of glass A were reacted for 56 days and then removed from the leachate, carefully rinsed, and placed in fresh deionized water and reacted for additional times up to 56 days. This variation tests the corrosion of a sample having an alteration layer in a leachate solution without any reaction products, where only the alteration layer is expected to affect the reaction. The upper dashed line in Fig. 11 shows the trend observed through the initial 56 days of reaction translated to days 56 through 112. Data from tests continued after the layers were

Table 4. Compositions (wt%) of Three Glasses Used by Chick and Pederson [155]

Oxide	Simulated Waste Glass		
	A	B	C
Al ₂ O ₃	6.1	6.9	5.9
B ₂ O ₃	4.2	-	9.1
CaO	2.1	7.1	-
CeO ₂	-	-	1.2
Fe ₂ O ₃	11.5	12.8	0.5
Gd ₂ O ₃	-	-	9.1
Li ₂ O	6.7	-	4.6
MoO ₃	-	-	1.7
Na ₂ O	8.3	9.0	9.7
Nd ₂ O ₃	-	0.1	2.6
NiO	1.1	0.7	0.3
P ₂ O ₅	0.8	-	2.2
SiO ₂	57.5	59.0	48.1
UO ₂	-	1.8	-
Others	1.7	2.6	5.0

Fig. 11. Variations of MCC-1 Test with Glass A: (○) MCC-1 Tests, (●) MCC-1 Tests in which Leachate is Replaced by Fresh Leachant after 56 Days (after [155]).

removed were expected to follow this curve if the corrosion was affected only by the solution chemistry. The lower dashed curve shows the extrapolation of the trend observed during the initial 56 days through 112 days. Data from tests continued after the layers were removed were expected to follow this curve if the corrosion was affected only by the mass transport through the layer. The normalized boron mass loss from glass A in tests continued in fresh leachant after 56 days shows a rapid increase after the leachate was replaced with fresh leachant, followed by a leveling off at longer times. This behavior is similar to that shown during the initial 56 days of reaction and indicates that the layer formed during the first 56 days of reaction had a minor effect on the corrosion rate during the second 56 days of reaction.

A different glass, glass B, was also reacted under MCC-1 conditions for times up to 56 days. The normalized silicon mass loss values measured in the MCC-1 tests are shown in Fig. 12 (open circles). The silicon release from glass B follows the same trend as the boron release from glass A, namely an initial rapid rise followed by a leveling off. In a second variation of the MCC-1 test, samples of glass B were reacted for half the prescribed test duration and then removed from the leachate and replaced with fresh (unreacted) samples which were reacted for the remaining time. This test measures the reactivity of a glass sample having no alteration layer in a leachate solution that contains dissolved glass constituents from the corrosion of the first sample. In this variation, the corrosion products remaining in the leachate are expected to affect the corrosion rate. The normalized silicon mass losses for these tests (filled circles in Fig. 12) continue the trend established by the initial glass samples during the first 56 days of reaction. This result indicates that the solution components from the corrosion of the first glass specimen dominate the corrosion rate of the second glass specimen. The observation that the normalized silicon release from the variant tests (in which the samples were replaced after half the reaction time, filled circles in Fig. 12) is slightly higher than that from the standard tests (open circles in Fig. 12) indicates that the layer does have a minor effect on the glass corrosion rate.

Fig. 12. Variations of MCC-1 Test with Glass B: (○) MCC-1 Tests, (●) MCC-1 Tests in which Glass Samples are Replaced by New Samples after Half the Scheduled Reaction Time (after [155]).

In a third variation of the MCC-1 test, samples of a third glass, glass C, were removed from the leachate solution, rinsed, and placed in fresh leachant every 7, 14, or 28 days. For comparison, glass C was also reacted for several different time periods under standard MCC-1 conditions; these data are plotted as circles in Fig. 13. (Note that this test variation is a periodic replacement test. See Sections 2.5.2.3 and 5.) Under these conditions, the alteration layer is expected to continue to grow on the monolithic glass sample, while the glass reaction products are periodically removed from the solution. It can be seen in Fig. 13 that the normalized sodium mass loss increases nearly linearly, with the rate of increase highest in the tests with the greatest solution replacement rate. Tests in which the leachate was replaced every 7 days maintain the most dilute solutions, but also generate the thickest alteration layers. Thus, for this glass, the solution effects are more important than the mass transfer effects. The results from these variations of the MCC-1 test all show that the solution chemistry has a much greater effect on the glass corrosion rate than does the generation of an alteration layer. This implies that mass transport of water and reaction products (i.e., boron from glass A, silicon from glass B, and sodium from glass C) through the alteration layer is faster than the reactions leading to their release from the glass. We note also that boron, sodium, and silicon fill different roles in the glass structure and are released via different reaction mechanisms (see the Introduction to Sections 2 and 2.1) Nevertheless, the release of all three is controlled primarily by the solution chemistry.

Grambow and Strachan used a different modification of the MCC-1 method to study the effects of the alteration layer on the corrosion of two glass compositions in deionized water and in a MgCl_2 solution [152]. Monolithic glass samples were reacted following the MCC-1 procedure for 57 days. At that point, samples in some tests were removed from the leachate, and the layers that had formed were physically removed from the samples. The remaining glass was then replaced in the same leachate and allowed to react for up to an additional 91 days (for a total

Fig. 13. Variations of MCC-1 Test with Glass C: (■) MCC-1 Tests, MCC-1 Tests in which Leachate is Replaced by Fresh Leachant Every (Δ) 7 Days, (○) 14 Days, and (□) 28 Days (after [155]).

reaction time of 148 days). Other tests were conducted without disruption, for comparison. The normalized boron and silicon mass losses for the glass MCC 76-68 (also known as PNL 76-68) and C31-3 glass in deionized water are plotted in Fig. 14. The boron and silicon releases are essentially the same whether the layer was removed or not, which shows that the layer had little effect on the corrosion rate.

In other tests in which MCC 76-68 and C31-3 glasses were reacted in a 0.001 M MgCl_2 solution, removal of the layer resulted in an increase in the normalized boron and silicon mass losses, as shown in Fig. 15. However, while the layer formed on the MCC 76-68 glass in the MgCl_2 solution may have been protective, Grambow and Strachan emphasized that the normalized release of boron (and to a lesser extent of silicon) from C31-3 glass was greater in the MgCl_2 solution than in deionized water, both with and without the layer present. This observation indicates that the increase is due to the solution chemistry and not the layer. Grambow and Strachan suggested that the affinity for glass corrosion was increased in the presence of magnesium because of the formation of a magnesium silicate, such as sepiolite, which reduced the silicic acid concentration in solution and accelerated the corrosion. Sepiolite is formed by the following reaction:

While magnesium was abundant in the leachant solution, silicic acid had to be supplied by corrosion of the glass. The reaction to form sepiolite (or perhaps another magnesium-bearing phase, such as magnesium-montmorillonite) approached equilibrium as silicic acid was released by corrosion of the glass during the initial 57 days. Removal of the layer after 57 days caused an increase in the forward reaction to generate more sepiolite. Sufficient magnesium was present in the solution, but additional silicic acid was generated by an increase in the glass corrosion rate.

Fig. 14. Variations of MCC-1 Test with (a) MCC 76-68 and (b) C31-3 Glass in Deionized Water: (○) B in Tests in which Layer is Removed after 57 Days, (●) B in Tests in which Layer is Not Removed, (□) Si in Tests in which Layer is Removed after 57 Days, and (■) Si in Tests in which Layer is Not Removed (after [152]).

Fig. 15. Variations of MCC-1 Test with (a) MCC 76-68 and (b) C31-3 Glass in MgCl_2 Solution: (○) B in Tests in which Layer is Removed after 57 Days, (●) B in Tests in which Layer is Not Removed, (□) Si in Tests in which Layer is Removed after 57 Days, and (■) Si in Tests in which Layer is Not Removed (after [152]).

The observed effect depends on the relative rates of sepiolite precipitation, which consumes silicic acid, and glass corrosion, which supplies silicic acid. The observation that the silicic acid concentration also increased upon removal of the layer implies that both glasses corrode faster than sepiolite precipitated. The effect of the silicic acid concentration on the glass corrosion rate is summarized in the next section.

Similar tests were conducted on the glass SM58LW11 in a MgCl_2 -dominated brine by Conradt and Roggendorf [156,157]. They periodically interrupted MCC-2 type tests (conducted with monolithic samples at 120 and 200°C and an SA/V ratio of 3.4 m^{-1}) and removed the corroded sample. Next they manipulated the sample by one of the following methods: (A) remove the alteration layer from the glass and replace layer and glass into same leachate; (B) remove the alteration layer, dry it at 110°C, weigh it, and then replace layer and glass into same leachate; (C) remove sample and place it in fresh leachant, with layer intact; (D) remove layer and place the sample in fresh leachant; or (E) replace the sample without further manipulation. Other tests were conducted without interruption for comparison. The results of tests conducted at 120°C are shown in Fig. 16. The one test conducted following method E was consistent with uninterrupted tests and showed that cooling the test vessels and reheating them did not affect the results. Tests following methods A and B, where the layers were removed then the samples replaced in the same leachate, both showed slightly greater corrosion than untreated tests. This result implies the layers had a slight protective effect. The results of manipulations A and B were deemed indistinguishable by the authors. Tests following methods C and D, where the samples were periodically placed in fresh leachant) both showed significantly greater corrosion than the untreated tests. The presence of the layer in C and the absence of the layer in D had little effect on the extent of corrosion, however. This finding demonstrates that the physical removal of the layer does not measurably affect the continued corrosion of the glass.

Fig. 16. Measured Extent of Corrosion vs. Time for Tests with Various Treated Samples: (○) Uninterrupted Static Tests, (◇) Tests with Manipulations A or B, (●) Tests with Manipulation C, (Δ) Tests with Manipulation D, and (◆) Tests with Manipulation E (see text, after [156]).

The results of tests with similar manipulations at 200°C lead to the same conclusions, except the removal of the layers in treatments A and B had no measurable effect. These results demonstrate that the layer has little or no effect on the corrosion rate of this glass in a brine solution, while the corrosion products that build up in the leachate slow the corrosion.

The effects of the MgCl_2 brine solutions observed by Grambow and Strachan and by Conradt and Roggendorf at first appear contradictory. While the tests conducted by Grambow and Strachan are similar to methods A and B used by Conradt and Roggendorf, Conradt and Roggendorf replaced both the removed layer and the glass back into the solution, while Grambow and Strachan replaced only the glass. Therefore, while the mass transport effects are eliminated in both methods, chemical interactions continue between the layer and the solution in the Conradt and Roggendorf methods A and B tests. If a secondary phase such as sepiolite is present in the layer, then its influence on the solution chemistry and the silicic acid activity will continue after being separated from the glass. That is, the perturbation of the equilibrium proposed by Grambow and Strachan will not occur if the sepiolite phase remains in contact with the solution. The different glass compositions tested by Grambow and Strachan and by Conradt and Roggendorf may also affect the comparison of the test results. (C31-3 glass has a higher content of aluminum and barium and a lower content of boron and alkali.) The difference in reaction temperature probably also affected the tests. Conradt and Roggendorf found the effect of layer separation to decrease as the temperature increased.

The results of Chick and Pederson, Grambow and Strachan, and Conradt and Roggendorf all show the solution chemistry to dominate the glass corrosion rate during the initial stages of glass corrosion (Stages I and II as described in Section 2.4). Alteration layers formed during glass corrosion have only a minor mitigating effect in these tests. The importance of this with regard to the effects of the SA/V on glass corrosion is that the effects of the SA/V occur solely through the solution chemistry. That is, the SA/V affects the solution chemistry and the solution chemistry is the primary controlling influence on the glass corrosion rate. However, differences in the solution chemistries generated in tests at different SA/V will affect the alteration layers formed on the glass surface as the glass corrodes.

It is important to recognize that the test methods used to assess the effects of the layer all maintained dilute conditions such that solutions did not approach saturation during the test period. Very different effects of the layers on glass corrosion have been observed in various tests that generated saturated solutions. An understanding of the corrosion mechanism under these conditions is essential, since these conditions are expected to dominate the long-term corrosion behavior. Unfortunately, the very slow kinetics that occur under saturated conditions make it difficult to characterize the corrosion mechanism or measure the corrosion rate under these conditions. Some test results have been interpreted to suggest that the effect of the alteration layers on the glass corrosion rate become more significant as the leachate solution becomes saturated and the hydrolysis reaction to release silicic acid slows to a negligible rate. Some researchers have suggested that ion exchange reactions determine the long-term glass corrosion rate [158-160], while Grambow et al. have suggested that water diffusion through the alteration layers controls the corrosion rate under silica-saturated conditions [161]. This issue is discussed in Sec. 3.5.

Barkatt and co-workers have observed transient increases in tests with several glass compositions during periodic replenishment tests that they attributed to cracking or sloughing of the alteration layer to expose fresh glass beneath the layer [127,162,163]. However, such transient excursions have only been observed in periodic replenishment tests, and may be an artifact of that test method (see also Section 2.5.2.3). Clear evidence of cracking or spalling has not been demonstrated. Also, possible effects of decreasing particle size need to be accounted for in long-term tests. In fact, examination of the data presented by Sang et al. show no obvious

"spike" of boron release beyond the range of experimental error. What is observed is an initially higher rate at the beginning of the test, which may be due to the presence of fines and reaction of high energy sites such as points and sharp edges on the crushed glass.

3.2 Observed Effects of Silicic Acid Concentration on Glass Corrosion

Since most waste glasses contain roughly 50 wt% SiO_2 , it is not surprising that leach tests have shown the silicic acid solution concentration to strongly affect the glass corrosion rate. For example, Pederson et al. [72] conducted MCC-1 tests (28 days, 10 m^{-1} , 90°C) with PNL 76-68 glass in solutions having various silicic acid concentrations, including two groundwaters. Figure 17 shows the normalized boron release from PNL 76-68 glass plotted against the silicic acid concentration. The glass corrosion rate, as measured by the boron solution concentration, decreases as the silicic acid concentration increases up to about 100 ppm. At silicic acid concentration higher than that, the glass corrodes at a very low and approximately constant rate. The open symbols represent silicic acid solutions, while the two solid symbols represent groundwaters. The observation that all data fit the same trend indicates that other components in the groundwater have only a minor effect on the corrosion rate.

The same dependence of the rate on the silicon concentration is shown in Fig. 18, where the corrosion rate of JSS-A glass, as measured by the boron release, is plotted against the ion activity product of silica in 28-day leach tests at 90°C at SA/V ratios of 10 m^{-1} [154,164]. From these results, it is clear that the maximum corrosion rate occurs when the solution contains no silicic acid and the minimum rate occurs when the silicic acid concentration attains a so-called "saturation" concentration. Further increases in the ion activity product (or silicic acid concentration) do not affect the corrosion rate.

The presence of silicic acid in the leachant solution has also been shown to affect the corrosion rates of high level waste glasses using a flow-through apparatus [71]. Some results are discussed in Section 3.2.

The effect of the concentration of silicic acid in the leachant solution becomes insignificant in tests at high SA/V, since silicic acid released from the glass dominates the solution after very short time periods. This is shown in tests conducted to assess the effects of SA/V on the corrosion of glasses relevant to the DWPF (see Sec. and Appendix IV B) in the boron concentrations in tests conducted at 90°C and at SA/V of about 340 and $20,000 \text{ m}^{-1}$ in deionized water and in a tuff groundwater solution containing about 45 ppm silicon [66]. For example, the boron concentrations in tests in deionized water and in groundwater at 340 m^{-1} conducted for 14 days were about 10 and 3 ppm, respectively, while the corresponding silicon concentrations are about 65 ppm in tests in deionized water and about 75 ppm. This shows the higher initial silicon concentration results in less corrosion in tests in groundwater than in deionized water. In contrast, the boron concentrations are about 77 ppm and silicon concentrations are about 280 ppm both in tests in deionized water and in groundwater at $20,000 \text{ m}^{-1}$ after only 3 days. This shows the silica released from the glass during the test quickly dominates the solution concentration, and the extent of corrosion in these tests is independent of the initial leachant composition.

A model in which the glass corrosion rate is controlled by the amount of silicic acid in solution has been developed by Grambow [153,154]. This model is based on an equation derived to represent the dissolution rates of silicate minerals. The rate limiting step is modeled to be the hydrolysis of a silane bond to release silicic acid into solution and form a silanol group in the glass. The net reaction given earlier in Eq. 6b, can be written to include the activated complex as

Fig. 17. Boron Release from PNL 76-68 Glass vs. Silicic Acid Concentration (○) Synthetic Solutions and (●) Groundwaters (after [72])

Fig. 18. Glass Corrosion Rate (as Measured by Boron Release) vs. the Ion Activity Product of Silicic Acid (after [154])

where the species in brackets represents the activated complex. The formation of the activated complex involves the adsorption of a water molecule onto the terminal silicon atom, which then becomes coordinated to five oxygen atoms, and the formation of a hydrogen bond between one of the hydrogen atoms of the adsorbed water molecule and the bridging oxygen. The mechanics of this reaction have been described by Lasaga and Gibbs [55], who point out that five-fold coordinated silicon is a stable structure. In addition, they show that adsorption of water to silicon through electron donation by the oxygen is a stable configuration, though less stable than sorption to silanol groups through electron acceptance of the hydrogen [55]. The breaking of the bond between the terminal silicon and bridging oxygen atoms shown in the activated complex (and dissociation of the water molecule) results in the release of silicic acid.

An expression describing the rate of this reaction was derived by Aagaard and Helgeson [54]. (A discussion of the derivation is presented in Appendix B.) The rate of the hydrolysis reaction to release silicic acid given in Eq. 6a has been modeled on the basis of irreversible thermodynamics; the rate is expressed as the product of a forward rate term and a term representing the chemical affinity for the reaction to release silicic acid from the glass [54]. The chemical affinity can be expressed in terms of the difference between the actual silicic acid concentration and a "saturation" concentration of silicic acid. Expressed this way, the reaction affinity decreases as the solution concentration of silicic acid approaches the "saturation" concentration [153]. In its simplest form, the rate of silicic acid release can be expressed as

$$\frac{d(n(\text{H}_4\text{SiO}_4))}{dt S} = k_+ \left(1 - \frac{[\text{H}_4\text{SiO}_4]}{[\text{H}_4\text{SiO}_4]_{\text{sat}}} \right) \quad (13)$$

where $n(\text{H}_4\text{SiO}_4)$ is the number of moles of silicic acid released from the glass into solution, S is the surface area of the glass, k_+ is the rate coefficient for the hydrolysis reaction, $[\text{H}_4\text{SiO}_4]$ is the molar silicic acid concentration in solution, and $[\text{H}_4\text{SiO}_4]_{\text{sat}}$ is the molar "saturation" concentration of silicic acid. The term $(1 - [\text{H}_4\text{SiO}_4]/[\text{H}_4\text{SiO}_4]_{\text{sat}})$ is the reaction affinity. Note that the rate as written in Eq. 13 has the units of moles silicic acid per time per area. Variations of Eq. 13 are often written in which the surface area term, S , is moved to the right-hand side of the equation. This change facilitates the use of experimental data for which the silicic acid concentration is measured after various reaction times (see below). It is important to remember that such an expression (with S on the right) gives the rate at which the solution composition changes, but not the corrosion rate of the glass. The corrosion rate of the glass is given in Eq. 13.

In general, the solubility limits of dissolved species are established by solid phases with measurable thermodynamic equilibrium constants. Glasses are thermodynamically metastable and cannot equilibrate with solutions. Instead, they appear to approach an apparent "saturation" concentration at which the dissolution rates become very low. While no solid phase that establishes the "saturation" concentration has been identified, empirically determined "saturation" concentrations are usually found to be intermediate between the solubilities of quartz and amorphous silica. This model and the concept of silica "saturation" have been found to describe the observed effects of the silicic acid concentration on the glass corrosion rate and the "roll over" behavior seen in Fig. 4 well.

On the basis of observations in dynamic tests, another term is sometimes included to account for the diffusion of material through the alteration layer that forms on the glass surface during corrosion [154,165]. This term may be important for glass compositions with relatively large values of k_+ but will not be further considered here.

Two limiting cases to Eq. 13 exist: when the affinity term is near one (when $[H_4SiO_4] \ll [H_4SiO_4]_{sat}$), and when the affinity term is near zero (when $[H_4SiO_4] \approx [H_4SiO_4]_{sat}$). When the affinity term is one, the rate is given simply by k_+ . When the affinity term is zero, the corrosion rate predicted by Eq. 13 is also zero. Experiments conducted under these limiting conditions have demonstrated the need to modify Eq. 13. First, tests under conditions for which the affinity term remains near one have shown the corrosion rate to be sensitive to the solution pH. Second, tests under conditions for which the affinity term approached zero suggest that the rate does not reach zero even after very long reaction times; rather it approaches a minimum value that may be two or three orders of magnitude less than k_+ [67,116,166-168]. These observations and the modifications of the rate equation made to account for them are discussed below.

In a slightly different approach, Bourcier has represented the reaction affinity in terms of all elements found in the alteration layer which forms on the reacted glass, instead of only silicon. The silicic acid concentration in the affinity term in Eq. 13 is replaced by the activity product of components which remain in the layer, while the silicic acid saturation value is replaced by a term computed using the solubilities of several phases modeled to comprise the layer [143]. The layer composition is measured experimentally, but is well approximated by simply excluding soluble alkali metals and boron from the glass composition. Layers typically include silicon, aluminum, iron, calcium, and magnesium. The solubility products calculated using this approach are similar to those regressed from experiments, since the calculated values are dominated by the contribution of silica. This approach identifies the phase controlling the "saturation" of the solution to be the alteration layer. That is, the terms $[H_4SiO_4]$ and $[H_4SiO_4]_{sat}$ are replaced by terms including other components in addition to silicic acid. The approach is appealing since the alteration phases are more likely to be equilibrated with the solution than is the glass. In fact, in the computer model used by Bourcier, the alteration layer is comprised of phases that have equilibrated with the solution. However, the reaction that the affinity term is meant to represent occurs between the glass and the solution.

3.3 Observed Effects of the Leachate pH on Glass Corrosion

The effect of the leachate pH on the corrosion of waste glasses has been measured under conditions where the silicic acid concentration remained low throughout the test. From Eq. 13, the corrosion rate under these conditions can be expressed as $\text{rate} = k_+$. To express its pH-dependence, the rate coefficient can be written as $k_+ = k_f (H^+)^h$, where k_f is the pH-independent rate coefficient and h is an experimentally determined value. In the limit where the silicic acid concentration is zero, the rate can be written as $\text{rate} = k_+ = k_f (H^+)^h$ or as $\log(\text{rate}) = \log(k_f) - h (\text{pH})$. Workers at Lawrence Livermore National Laboratory have measured the effects of the solution chemistry on the corrosion rate of high-level waste glasses by using a single-pass flow-through device. In these experiments, pH buffers were added to the leachant solution to measure the effects of the leachant pH on the glass corrosion rate. The solution leaving the reaction cell was sampled periodically until constant solution concentrations of key glass components were measured. The constant concentrations indicated that apparent "steady state" conditions had been reached, where it could be assumed that the effects of fines and surface stresses were eliminated (see below).

Figures 19a and 19b show the release of several elements from the CSG glass in tests conducted at 70°C at pH values of about 6 and 10 [112]. The CSG glass is a simple, five-component glass that was designed to simulate a compositionally more complex DWPF glass. It contains, in oxide weight percent, 11.7 Al₂O₃, 8.4 B₂O₃, 6 CaO, 18.2 Na₂O, and 55.7 SiO₂. At pH 6, boron, calcium, and sodium are released stoichiometrically and at a higher rate than are silicon or aluminum. At pH 10, all five elements are released stoichiometrically. This is because the solubility of silicon is greater at pH 10 (see below). Notice that the release of boron, calcium, and sodium is similar at pH 6 and pH 10. The increased release rate observed for aluminum is not due to an increase in its solubility. Instead, aluminum probably is removed from the glass stoichiometrically with silicon as the surface disintegrates and it is flushed by the solution flow. Aluminum is commonly observed to be retained on the glass surface as insoluble residue in static tests, many of which attain high pH (e.g., see Appendix D).

Figure 20 shows the corrosion rate of CSG glass at 70°C (measured by the solution concentrations of glass components) as a function of the leachant pH, as measured in a single-pass flow-through apparatus [70]. The logarithm of the rate, as measured by the silica release, increases linearly with the pH over the range 6 < pH < 13. The same pH effects have been observed in tests with DWPF reference glasses [71]. The increase in the rate with decreasing pH below about pH 6 is probably due to reactions involving aluminum (see below).

A similar pH dependence was observed in modified MCC-1 tests (SA/V = 2 m⁻¹) with monolithic samples of a defense waste reference glass (DWRG) glass in buffer solutions at 90°C [169]. The measured silica concentration had a minimum between pH values of pH 4 and pH 7 and increased at both lower and higher pH values.

Figure 21 shows the corrosion rate of R7T7 glass in static tests similar to MCC-1 tests performed at 90°C [92]. The reaction times varied with the pH to measure the rate under dilute conditions. Short testing times (a few hours) were used at high pH values and longer testing times (up to three weeks) were used at low pH values. These results also show the corrosion rate to increase linearly with the pH in neutral and alkaline solutions. The slope of the pH dependence (h) has a value of about 0.4 for both the simulated DWPF glass at 70°C [70] and the French glass R7T7 at 90°C [92]. We note that the tests with R7T7 glass did not show the increase in the corrosion rate at low pH values observed in tests with CGS glass. This is probably due to differences in the glass compositions, the reaction temperatures, or other test conditions.

A similar pH effect was reported by Barkatt et al. in tests with Pyrex and a high-silica glass conducted at 70°C [136]. In these tests, the dissolution rate of the high-silica glass was about 2.5 orders of magnitude higher in tests at pH 9 than at pH 6, while the dissolution rates were similar at pH 2 and pH 6.

Insight into the pH dependence of waste glass corrosion can be drawn from the study of the release of various components from simple glasses. Tests were conducted by reacting 2 g of crushed glass in 100 mL of various aqueous buffer solutions having pH values between about 1 and 13 at a constant temperature. Different size fractions, test temperatures, and reaction times were used in different studies. The release rates of alkali metals from several binary alkali silicate glasses are shown in Figs. 22a-c as a function of the solution pH [170]. Tests were conducted at 35°C using -30+52 mesh glass. Tests were conducted for up to 4 hours. Over this time period, the release of alkali metal ions was found to increase with the square root of time, which indicates that water diffusion and ion exchange are the dominant reactions. The extent of extraction is generally insensitive to the solution pH for values of about pH 9 or lower, but decreases precipitously as the pH was increased to pH 10 or higher [170]. The decreased extraction with increasing pH can be explained as a shift in the ion exchange equilibrium (see

Fig. 19. Release of B, Na, C, Al, and Si from CSG Glass as a Function of Time at (a) pH 6 and (b) pH 10: (■) Si, (●) Al, (◇) B, (Δ) Na, and (○) Ca (after [112]).

Fig. 20. Corrosion Rate of CSG Glass vs. pH (after [70])

Fig. 21. Corrosion Ratio of R7T7 Glass vs. pH (after [92])

Fig. 22. Alkali Release Rates from Simple Glasses vs. pH: (a) Sodium Release Rate from (●) $15\text{Na}_2\text{O} \cdot 85\text{SiO}_2$, (○) $25\text{Na}_2\text{O} \cdot 5\text{CaO} \cdot 70\text{SiO}_2$, (□) $25\text{Na}_2\text{O} \cdot 10\text{CaO} \cdot 65\text{SiO}_2$, (■) $22\text{Na}_2\text{O} \cdot 6\text{CaO} \cdot 72\text{SiO}_2$; (b) Lithium Release Rate from $33\text{Li}_2\text{O} \cdot 85\text{SiO}_2$; and (c) Potassium Release Rate from $15\text{K}_2\text{O} \cdot 85\text{SiO}_2$ (after [170]).

Eq. 2 in Section 2.1). It is interesting to note that, although the critical pH values for alkali extraction are similar for lithium, sodium, and potassium, the exchange equilibrium constants measured by Dugger et al. [6] suggest that the critical pH should increase slightly as $\text{Li} < \text{Na} < \text{K}$ (see also Fig. 1 from [7]).

The extraction of silicon from fused silica powder and from some simple glasses is shown in Figs. 23a and 23b as a function of the solution pH [170,171]. Tests in Fig. 23a were conducted at 80°C and tests in Fig. 23b were conducted at 35°C using -30+52 mesh glass. Tests were conducted for up to two hours. Over this time period, the release of alkali metal ions was found to increase linearly with time, which indicates that hydrolysis is the dominant reaction. In all cases, the amount extracted is insensitive to the pH for pH values below about 9 but increases as the pH is increased above about 9. The increased extent of extraction is due to the dissociation of silicic acid. While the solubility of nondissociated silicic acid is itself insensitive to changes in the pH, the extent of its dissociation to form H_3SiO_4^- , $\text{H}_2\text{SiO}_4^{2-}$, HSiO_4^{3-} , and SiO_4^{4-} increases with the solution pH.

Figure 24 shows the extraction of sodium, calcium, and silicon from a glass composed of 20 mol% each of soda and lime and 60 mol% silica at 70°C [172]. Both sodium and calcium release increases with decreasing pH, while the silicon release is insensitive to the pH at values below about pH 9.

The extraction of aluminum and sodium from two sodium glasses was measured at different solution pH in tests conducted at 70°C using -25+36 mesh glass [173]. Tests were conducted for 20 minutes. Glass I contained 12.9 mol% Na_2O and 16.1 mol% Al_2O_3 and glass II contained about 13.0 mol% Na_2O and 5.0 mol% Al_2O_3 , with the balance being SiO_2 . Figure 25 shows the extent of extraction of aluminum and sodium from two sodium glasses at different solution pH values. The extraction of aluminum is insensitive to the leachant pH at pH values greater than about 6 but increases as the pH decreases below about 3 for both glasses. The sodium release, while insensitive to pH changes below about 9 in soda lime silicate glasses, shows increased extraction below about 3 in glass I and below about 8 in glass II. The difference in the sodium release behavior of the two glasses can be understood in terms of the relative amounts of sodium and aluminum in the two glasses. Glass I has a sodium-to-aluminum mole ratio less than one, so all sodium are fixed in $[\text{NaAlO}_4/2]$ groups. Glass II has a sodium-to-aluminum mole ratio much greater than one, so most sodium atoms are fixed in nonbridging $\equiv\text{SiO}-\text{Na}$ groups. Release of sodium from $\equiv\text{SiO}-\text{Na}$ groups in glass II occurs preferentially to and at higher pH values than the release of sodium from $[\text{NaAlO}_4/2]$ groups in glass I.

A similar pH-dependence has been observed for the dissolution rate of silicate minerals, for which the slope of the log rate versus pH plot is 0.3 at 70°C and about 0.5 at 90°C [174,175]. The pH-dependence was attributed to the formation of negatively charged silica groups at the surface, and the temperature effect was attributed to the temperature dependence of the dissociation constant of silanol.

The importance of the solution pH to both the rate and mechanism of the corrosion of simple glasses is clearly evidenced by a variety of experiments and has been well known for many years. The similar pH effects seen in tests with simple silicate glasses and with high-level waste glasses demonstrate that similar reactions control their corrosion behaviors, namely ion exchange to release alkali metal and alkaline earth ions and acid-catalyzed hydrolysis to release aluminum and base-catalyzed hydrolysis to release silicon. Based on knowledge gained from tests with simple glasses, the effects of pH on the corrosion of high-level waste glasses can be summarized as follows: Dealkalization occurs at a high rate that is independent of the pH in acidic and near neutral solutions, but is quenched at pH values above about pH 9. Aluminum release increases as the pH decreases below about pH 4, and silicon release is greatly enhanced

Fig. 23. (a) Silicon Release from Fused Silica vs. pH (after [172]) and (b) Silicon Release vs. pH from (○) 33Li₂O•67SiO₂ and (●) 15K₂O•85SiO₂ Glasses (after [170,171]).

Fig. 24. Release of Sodium, Calcium, and Si from $20\text{Na}_2\text{O}\cdot 20\text{CaO}\cdot 60\text{SiO}_2$ vs. pH: (▲) Na, (◆) Ca, and (○) Si (after [172]).

Fig. 25. Release of Aluminum and Sodium vs. pH (a) from Glass 1 and (b) from Glass 2: (O) Na and (●) Al (after [173])

above about pH 9 (see also Section 3.3). Dealkalization of waste glasses generally results in basic conditions, so the waste glass corrosion is controlled by the release of silicon. (Radiolysis may result in acidic leachates, in which case waste glass corrosion will be controlled by the release of aluminum.) The similarity of values for the pH-dependence of silica release that have been measured for simple silicate glasses and for high-level waste glasses support the model that detachment of silica groups controls the glass corrosion rate under alkaline pH values and that the effects of other constituents are negligible.

Models designed to simulate the long-term corrosion behavior of high-level waste glasses have only recently included terms to account for the effects of the solution pH on glass corrosion [176,177]. As will be shown in later sections, the pH effects are very important in understanding the effect of the SA/V ratio.

3.4 Observed Effects of Other Species on Glass Corrosion

Various test methods have been used to assess the effects of specific solution species on the glass corrosion rate. The effects observed in these different tests are related to the reaction that is highlighted in the test method. That is, the effect of a species in a test that generates a highly concentrated leachate solution may be very different than the effect measured in test that maintains a dilute leachate solution. That is, the measured effect may involve different rate-affecting reactions in the different tests, and the effect of a particular species may be very different when measured in the different tests. Thus, it is important to understand the corrosion conditions when discussing the effects of different species.

Flow-through tests with doped leachants have been used to assess the effects of various solution components on the corrosion rate of high-level waste glasses. These tests maintain highly dilute conditions and measure the effects on the forward reaction rate. The measured effects of silicon, aluminum, magnesium, and calcium on the forward reaction rate of CGS glass at different pH values are shown in Fig. 26 [71,112]. The dashed lines show the limiting slopes

Fig. 26. Effects of Si, Al, Mg, and Ca on CSG Dissolution: (O) Undoped Leachant, (●) Si-Doped Leachant, (Δ) Al-Doped Leachant, (■) Mg-Doped Leachant, and (◇) Ca-Doped Leachant (after 71,112]

of the data of Knauss et al. in tests with the same glass [70]. Tests conducted by Bourcier et al. with undoped leachant show good agreement with the results of Knauss et al. The presence of calcium and magnesium in the leachant did not affect the corrosion rate of the glass. The presence of boron in the solution also did not have an effect. The presence of silicon in the leachant solution decreased the corrosion rate at solution pH values greater than about pH 6, while the presence of aluminum decreased the corrosion rate at pH values less than about pH 6. The data in Fig. 26 suggest that the pH dependence of glass corrosion in silicon- and aluminum-containing solutions follows the limiting slopes found by Knauss et al. This suggests that glass corrosion in acidic solutions is dominated by reactions affected by the aluminum concentration and corrosion in alkaline solutions is dominated by reactions affected by the silicon concentration. As shown in Fig. 25, the pH at which the aluminum release increases depends on the glass composition. (This is probably why R7T7 glass did not show increased corrosion at low pH values [166].)

The lack of effect of magnesium determined in the flow-through tests by Bourcier et al. is contrary to the severe quenching effect of magnesium in brine solutions observed by Barkatt et al. in highly saturated environments [14,163,178]. Barkatt et al. attributed the observed effect of magnesium to its attachment to silicate sites and its tendency to reduce the solubility of silica. These effects are not observed in flow-through tests. Therefore, while a solution species may not directly affect the reaction leading to the release of glass components, it may indirectly affect the glass corrosion rate through its effect on solubilities or interactions with the glass surface. The test method used to assess the effects of species controls which effects may or may not be observed, so that several tests may be required to fully characterize the effects of a species over a range of conditions.

The above results indicate that in alkaline solutions, it is valid to model the forward corrosion rate to depend primarily on the pH and silicic acid concentrations. In acidic solutions, however, the effects of aluminum may also have to be taken into account. Acidic conditions may exist when large volumes of brines contact the glass [179] or when small volumes of water undergo radiolysis [114]. The validity of representing the corrosion of a multicomponent glass based only on the solution concentration of silicic acid rests primarily on the general success of the model when applied to waste glasses. (See [92,94,154,164,165,180,181-184] for some applications.) It is likely that any error due to omission of the possible minor effects of other glass components on the corrosion rate in the analytical equation is corrected by the fact that experimental data are used to define various model parameters (see Section 6.1). That is, even though the effects of species other than silicic acid are not specifically accounted for in the model, their influence is included by the use of experimentally derived model parameters.

3.5 Glass Corrosion under Silicon Saturated Conditions: The Residual Rate

Many workers have emphasized the importance of knowing the long-term corrosion behavior of waste glasses in order to model waste glass performance in disposal sites [185,186]. McGrail et al. state that "it is impossible to accurately predict the solubility-limited, mass-transfer rate without accurate knowledge of the kinetics of glass reaction" [185]. Their model calculations show that the transmissivity of radionuclides through a geologic site is proportional to the rate of glass corrosion.

In contrast to Eq. 13, in which the hydrolysis rate is predicted to become nil as silicic acid approaches its saturation concentration and the affinity approaches zero, experiments show the glass corrosion rate to approach a very low, yet non zero value after reaction times of about six months. It is important to note that the residual rate may not represent the long-term corrosion rate (see below). For example, Fig. 27 shows the release of boron and silicon from C-31-3EC

Fig. 27. Normalized Mass Loss of (▲) Boron and (●) Silicon from C-31-3EC Glass in NaCl Solutions at 200°C (after [165])

glass in static leach tests (SA/V from 10 to 100 m⁻¹) conducted in saturated NaCl solutions at 200°C. The constant increase in the normalized boron mass loss at a slope of one on the log-log plot indicates that boron is being released to solution at a constant rate (see also Section 6).

Fruede et al. report the boron release rate to be 1 g/m²/d after several months [168]. The normalized boron mass loss data at short reaction times lie above the line, indicating a high corrosion rate occurs at short time periods. Fruede et al. report the forward rate to be 6 g/m²/d [168]. In contrast, the normalized silicon mass loss remains essentially constant between about 5 and 16 g/m². This indicates that additional silicon is not being released to solution as the reaction progresses, and that the solution has become saturated with silica after short reaction times. An *ad hoc*, constant "residual rate" or "contextual affinity" term is sometimes added to the rate expression to account for the continued reaction under "saturated" conditions [153,166,168,181]. This is shown in Eq. 14, where the first term on the right hand side is from Eqn. 13 and R_{∞} is the residual rate.

$$\frac{d \left(n(\text{H}_4\text{SiO}_4) \right)}{dt} = k_+ \left(1 - \frac{[\text{H}_4\text{SiO}_4]}{[\text{H}_4\text{SiO}_4]_{\text{sat}}} \right) + R_{\infty} \quad (14)$$

Values of the R_{∞} term are generally derived from experimental results, and often from tests conducted at high SA/V. High SA/V conditions are used to generate highly concentrated leachates such that the silicic acid concentration approaches the saturation limit and the affinity term approaches zero within short test periods. Under these conditions, the instantaneous rate measured after long reaction times corresponds to the residual rate. However, the corrosion rate measured and the value of R_{∞} have been found to depend on the SA/V of the test [67,167], probably because of differences in the solution pH in tests at different SA/V. Vernaz and

coworkers have suggested that the stability of the phase controlling the silicic acid concentration may vary with the SA/V, or other, more stable phases may form under different test conditions. The attainment of high solution pH values occurs through the same SA/V effect of a low degree of dilution as the attainment of high silicic acid concentrations, and is a natural consequence of using SA/V to attain concentrated solutions. The high solution pH values result in an increase in the forward rate coefficient, k_+ , and increased silicon concentrations due to dissociation of silicic acid. Both the increase in k_+ and the pH-induced increase in the measured silicic acid concentration must be accounted for when computing the residual rate. The residual rate should be evaluated based on the results of tests conducted at several different SA/V to account for the effects of pH on corrosion and on the residual rate. Estimates of the residual rate made to date are based on corrosion under a single test condition [165,180] (see also Section 6). Modeling the long-term glass corrosion behavior requires an understanding of the effects of test conditions and solution chemistry on the corrosion behavior of glass in general, and on the residual rate, or contextual affinity, in particular.

The concept of a "residual reaction affinity" for glass corrosion based on silicon saturation has been questioned by some researchers who point out that other glass components besides silicon continue to be released from the glass at measurable rates even when the solution is "saturated" with silicon [24]. They suggest that the concept of residual affinity should not be limited to silicon, but extended to include effects of other glass components.

Although the process responsible for continued glass corrosion remains uncertain, continued corrosion in solutions that are saturated with silicic acid has recently been attributed to diffusion-limited ion exchange [187] or water diffusion into the glass [161]. While these reactions are known to occur during glass corrosion, they are generally assumed not to be involved in the rate controlling reaction. Although diffusion through surface layers is known not to significantly affect the corrosion rate of borosilicate waste glasses at early stages of corrosion (see Section 3.1), these tests do show a minor influence of the layer. The effects of the layer may become more significant as the solution effects become negligible. That is to say, the tests conducted to distinguish the effects of the solution and the layer were conducted under conditions relevant to the early stages of corrosion. Other tests relevant to the long-term stages of corrosion must be used to distinguish the relative importance of the alteration layers on the long-term corrosion rate.

The uncertainty surrounding the "residual rate" is due to the fact that the tests used to derive the expression for the corrosion rate focus on the approach of the solution chemistry to saturation, while the residual rate occurs under saturated conditions. Grambow et al. have speculated that residual rate is controlled by diffusion based on their observation that the corrosion rate appears to be proportional to the square root of the reaction time in tests conducted in brines at high SA/V [161]. They also show that the apparent diffusion coefficient for the residual rate is similar to those of several obsidians. Grambow suggested both affinity terms and diffusion terms be utilized in the corrosion rate expression, where one or the other term dominates the corrosion rate, depending on the reaction conditions.

The conclusion of Grambow et al. that the long-term corrosion rate is controlled by the diffusion of water through an alteration layer is contrary to several experimental findings regarding the effects of alteration layers on the glass corrosion rate. First, thicker layers are known to form in tests at lower SA/V than in tests at high SA/V, so that the long-term rate would be expected to be lower in tests at lower SA/V than in tests at higher SA/V. The opposite is observed [167]. Characterization of the glass/layer interfaces developed on some high-level waste glasses shows the layer to crystallize and separate from the glass, such that the corroding solution directly contacts the glass surface [87]. No diffusion layers exist on these glasses under saturated conditions. (Other glasses may behave differently.) Inspection of the data of

Grambow et al. does not show an obvious root time dependence. Their interpretation would be considerably strengthened by tests at common $(SA/V)t^{1/2}$. Finally, the same corrosion behavior is not observed by Grambow et al. in different brine solutions. The behavior is seen to be affected by the solution pH.

Inagaki et al. have included such a term in their reaction path calculations to describe test results with a borosilicate glass identified as P0798 [188]. Tests were conducted with crushed glass (-100+200 mesh) in deionized water at a SA/V of about 1200 m^{-1} at 90°C for up to 130 days. Modeling the test results with the reaction path model and without the diffusion term resulted in an underestimation of the release rates by a factor of two. Apparently, Inagaki et al. used the forward rate, saturation concentration and residual affinity terms determined in tests with other glasses. Certainly, determination of values for these constants using test data for P0798 would improve the fit. Instead, the authors fit the data using a diffusion term and observed a much improved fit to the data. (The contribution of the reaction path term was significantly reduced by halving the forward rate coefficient.) This is due to the fact that the data being fitted were used to determine the fitting coefficients. We note that the test data are constrained to the initial stages of corrosion which can be fit equally well by a diffusion equation and by an affinity-based equation, if appropriate model parameters are used [143].

The need to understand the long-term reaction mechanism and rate-determining reaction is underscored by the importance of the long-term rate to projecting glass corrosion behavior. The time dependence of corrosion under saturated conditions, whether it is linear or root time, will have a significant impact on projections of glass corrosion behavior to long time periods.

Experimental evidence suggests that most high-level waste glasses do not continue to corrode at the "residual rate." Rather, the corrosion accelerates because of the effects other phases that form during glass corrosion have on the silicic acid activity. Attainment of the "residual rate" is known to be prevented in the presence of silicon-consuming phases that may be present as part of the test system, such as clay [189-192] and iron [193,194], and upon the formation of secondary phases from the leachate solution as the glass corrodes [65,81,116]. The effect of secondary phase formation were discussed by Grambow and Strachan when accounting for corrosion in Mg-brines in MCC-1 type tests [152] (see Sec. 3.1). Increased corrosion rates have been observed for several glasses upon formation of secondary phases in tests conducted at high SA/V ratios under PCT conditions [65] and under vapor conditions [81,105,111].

The formation of secondary phases is an unavoidable result of glass corrosion. They form as the glass converts to a more stable assemblage of phases to reduce the free energy of the system. However, because the material for these phases originates in the glass, the opportunity exists to formulate waste forms to reduce the impact of secondary phase formation on glass corrosion by controlling which phases form. For example, analcime ($\text{NaAlSi}_3\text{O}_8$) is a silicon-consuming phase that is typically formed early in the corrosion process. However, its formation has less effect on the corrosion rate in tests with glasses having low aluminum contents, such as PNL 76-68, than in tests with glasses having high aluminum contents, such as SRL 131, because much higher silicic acid concentrations can be maintained in the presence of analcime if the aluminum concentration is low [104]. In other words, the solution concentration of aluminum limits the formation of analcime. The response of the glass corrosion process to the formation of secondary phases such as analcime also depends on the solution chemistry: under the alkaline conditions common during waste glass corrosion, the glass corrosion rate can accelerate to supply silicon. On the other hand, the glass corrosion rate does accelerate under alkaline conditions to supply aluminum for secondary phase formation. Therefore, the effects of

analime formation on the glass corrosion rate are less in glasses with low aluminum concentrations, and waste forms could be formulated to reduce the aluminum concentration to improve the long-term durability.

However, the aluminum content is not the only factor affecting the durability of waste glasses. The formation of secondary phases in vapor hydration tests conducted at ANL using the R7T7 glass, which contains about 5 wt% Al_2O_3 , has been found not to greatly increase the corrosion rate. The durability demonstrated in vapor hydration tests and the durability under acidic conditions observed by Advocat et al. [92] (see Fig. 21) suggests that aluminum is quite stable in the R7T7 glass structure. Of course, the formation of kinetically favored phases, such as clays, will also affect the evolution of the solution chemistry and the formation of precipitated phases.

3.6 Summary

Laboratory tests with high-level waste glasses have shown the solution chemistry to strongly affect the corrosion rate. Mass transport of water species and corrosion products through alteration layers has only a minor affect on the corrosion rate during the early stages of glass corrosion, although mass transport may affect the corrosion rates of some glasses under saturated solution conditions. The aspects of the solution chemistry that have the most effect on the corrosion rate are the silicic acid concentration and the pH. The pH affects the reactions responsible for the release of glass components: the release of alkali metals and alkaline earths are promoted at low pH values (below about pH 9), while the release of silicon is promoted at high pH (above about pH 9). The release of silicon is slowed by the presence of silicic acid in the solution. The glass corrosion rate is well-represented by an equation describing the silicic acid release rate that accounts for both the pH and silicic acid solution effects. This equation also accounts for the effects of other phases on the silicic acid concentration and pH. These phases may be present in the system initially or may be formed as secondary phases as the glass corrodes. An additional term may be required to describe the nonzero corrosion rate observed under saturated conditions prior to secondary phases formation. Such a term can presently only be determined experimentally, and the effects of test conditions on the value of the term are not clear. The physical meaning of the term is likewise unclear, and remains a shortcoming of our ability to model and predict long-term glass behavior.

4.0 OBSERVED EFFECTS OF THE GLASS SURFACE AREA-TO-SOLUTION VOLUME RATIO IN STATIC TESTS

Most studies of the effects of the SA/V used in a test on the corrosion rate of high-level waste glasses have focused on changes in the solution concentration. While the release of glass components into solution is of prime interest, since transport of waste components requires their initial release into the leachant, the effects of the SA/V on the alteration of the glass surface are important to understanding the effects on the corrosion mechanism and the long term corrosion behavior. Knowledge regarding the effects of SA/V on the alteration of the glass surface is needed to determine if tests conducted at high SA/V can be used to characterize advanced stages of corrosion. Thus, both the effects of SA/V on the leachate chemistry and the effects of SA/V on the alteration of the glass surface must be understood to support modeling of the long-term corrosion behavior. Laboratory tests have shown the chemical and physical alteration of the glass surface to have only a minor effect on the corrosion rate (see Section 3.1). However, the alteration phases that accumulate at the glass surface do interact with the leachate solution and affect the corrosion behavior at advanced stages of corrosion (Stage III in Fig. 6). The model used by Bourcier utilizes the composition of the layer to determine the saturation concentration in the affinity term [143].

4.1 Effects of SA/V on the Solution Chemistry

The current model of glass corrosion considers the glass surface to be a reservoir for reactants, and the glass surface area can be assumed to be proportional to the amount of each glass constituent (including alkali metals, silicon, and radioelements) that is available for reaction. The solution volume acts both as a reservoir for dissolved glass components and as a source of reactants, including molecular water and hydroxide involved in hydrolysis reactions, alkali metal ions involved in ion exchange, and silicic acid. The glass surface area and the solution volume are not phenomenologically related. Rather, their coupling into a single test parameter is derived from the rate expression describing the hydrolysis of silicon-oxygen bonds at the glass surface. If all silicic acid released from the glass remains in solution, then the number of moles of silicic acid released from the glass, $n(\text{H}_4\text{SiO}_4)$ in Eq. 13, can be replaced with the molar concentration of silicic acid as $n(\text{H}_4\text{SiO}_4) = [\text{H}_4\text{SiO}_4] \cdot V$, where V is the solution volume, and Eq. 13 can be written as

$$\frac{d[\text{H}_4\text{SiO}_4]}{dt} = \left(\frac{\text{SA}}{V} \right) k_+ \left(1 - \frac{[\text{H}_4\text{SiO}_4]}{[\text{H}_4\text{SiO}_4]_{\text{sat}}} \right) \quad (15)$$

The observed "effect" of the glass surface area/solution volume ratio (SA/V) used in a static test on the glass corrosion rate is the net effect of the glass surface area, which establishes the amount of glass-phase reactant initially available, and of the solution volume, which establishes the extent to which reaction products are diluted. The solution concentrations of key glass components then affect the rates of reactions critical to glass dissolution, including ion exchange and hydrolysis reactions. As discussed earlier, the left hand side of Eq. 15 no longer represents the rate at which silicic acid is released from the glass (i.e., the corrosion rate); rather, it represents the rate at which the silicic acid concentration in the leachate changes. The rate of change is proportional to the SA/V of the test.

Equation 15 predicts that the SA/V used in a corrosion test can be increased to accelerate the saturation of the solution, that is, to drive $[\text{H}_4\text{SiO}_4]$ to $[\text{H}_4\text{SiO}_4]_{\text{sat}}$. Several series of tests have been conducted to verify the predicted effect of SA/V for various glass composition. The results of a few representative tests are discussed below to assess the utility of Eq. 15. In the

presented results, the solution concentrations of glass components other than silicon are sometimes used to quantify the extent of corrosion. The qualitative effect of the SA/V is the same, regardless of which glass component is used, and, as discussed in Section 2.2, highly soluble components, such as boron and alkali metals, provide the best measure of corrosion as the solution becomes saturated.

Clark et al. performed tests at SA/V ratios of about 7.7, 77, and 770 m⁻¹ for up to ten days at 100°C [135]. They used deionized water and various masses of the -45+60 mesh fraction of crushed glass and deionized water to attain the different SA/V ratios. The glass used contained 14 mol% Na₂O, 10 mol% CaO, 72 mol% SiO₂, 1.8 mol% Al₂O₃, 0.7 mol% MgO, 0.5 mol% K₂O, and about 1 mol% "other" oxides. The normalized sodium mass losses and the measured pH values for tests at different SA/V are plotted in Figs. 28a and 28b, which show that tests at lower SA/V attain higher sodium releases and higher corrosion rates, based on sodium release. Tests at higher SA/V generate solutions with higher pH values and higher sodium concentrations, but normalization to the SA/V shows tests at higher SA/V to have lower glass corrosion rates when expressed as mass glass dissolved per unit area.

Shade and Strachan studied the corrosion of PNL 76-68 glass in deionized water and in a silicate/bicarbonate solution at 90°C. They used different size fractions of crushed glass to attain SA/V of about 670, 1780, and >1780 m⁻¹ (calculated on the assumption that the grains were cubes with an edge length equal to the average mesh opening) [132]. Figure 29a shows the pH values to be slightly higher for the tests conducted at higher SA/V. (Similar pH values were measured in tests conducted in the silicate/bicarbonate solution under the same test conditions.) This indicates that the glass corrosion products overwhelm the initial leachant chemistry in these tests, and that the initial leachant does not significantly affect the corrosion rates of these tests. The pH increase occurs over the initial few days and reaches a nearly constant value at longer times. The leveling off of the pH occurs sooner for tests at higher SA/V ratios.

Figure 29b shows the leachate concentrations of silicon in the same tests in deionized water. The silicon concentrations increases initially, more so for tests at higher SA/V, and then increases at a constant rate for longer reaction times. Notice that the results of tests conducted for 1 year are above the trend shown by the shorter term tests. Similar behavior has been seen in tests with other glasses, which has been attributed to the effects of secondary phase formation (see below). Filtration of the leachate revealed that the amount of dissolved silicon (that passing through a 1.8-nm filter) was similar at all SA/V and that colloidal silicon accounted for the differences in the leachate concentrations at different SA/V beyond about 90 days. (This is surprising, in light of the different pH values in tests at different SA/V.) The silicon concentrations measured in tests in the silicate/bicarbonate solution were similar to those measured in corresponding tests in deionized water. This behavior demonstrates that the glass corrosion products (rather than the initial leachant chemistry) dominate the solution chemistry in these tests.

Tests with the same glass were conducted at 150°C to accelerate the glass corrosion [132]. Similar solution pH values (between about 9.9 and 10.1) and similar silicon concentrations (about 500 ppm) were measured in the leachates of tests conducted at all SA/V. The pH increased slightly with time, while the silicon concentration did not. The measured boron concentrations increased slightly with time, and the concentrations were consistently higher in tests at higher SA/V for all times tested. The continued boron release indicates that the glass continues to corrode even after silicon becomes saturated. The silicon concentration was likely set by the solution pH, which was similar at all SA/V. The glass components that have saturated the solution become incorporated in secondary phases, while those that have not saturated the solution continue to dissolve (see Section 2.2).

Fig. 28. (a) Normalized Sodium Mass Loss and (b) pH vs. Time at Different SA/V Ratios: ($\square$) 7.7 m^{-1} , ($\blacksquare$) 77 m^{-1} , and ($\circ$) 770 m^{-1} (after [135])

Fig. 29. Measured (a) pH and (b) Si in Tests with PNL 76-68 Glass Conducted in Deionized Water at Different SA/V Ratios: ($\blacktriangle$) $>1780 \text{ m}^{-1}$, ($\blacksquare$) 1780 m^{-1} , and ($\bullet$) 670 m^{-1} (after [132])

The tests conducted by Clark et al. and by Shade and Strachan show that solution pH and the solution concentrations of glass components increase with the SA/V used in a static leach test, and that high SA/V tests may result in the saturation of silicic acid. The latter observation suggests that results from high SA/V tests will provide insight into the advanced stages of glass corrosion.

4.2 Applicability of (SA/V)t as a Scaling Factor

Before test at high SA/V can be used to characterize advanced stages of glass corrosion, the relationship between tests at different SA/V and between tests using monolithic and crushed glass samples must be established. The relationship between tests at different SA/V can be determined by integration of Eq. 15, which leads to the form [153]

$$[\text{H}_4\text{SiO}_4] = [\text{H}_4\text{SiO}_4]_{\text{sat}} \left(1 - \exp \left\{ \frac{-(\text{SA}/\text{V}) t k_+}{[\text{H}_4\text{SiO}_4]_{\text{sat}}} \right\} \right) \quad (16)$$

Eq. 16 predicts that the silicic acid concentration will approach the limiting value $[\text{H}_4\text{SiO}_4]_{\text{sat}}$ at a rate that is a function of the product $(\text{SA}/\text{V})tk_+$. If k_+ remains constant for tests at all SA/V, then tests performed for the same $(\text{SA}/\text{V})t$ are expected to attain the same silicic acid concentrations in the solution. Therefore, the silicic acid concentration will approach the limiting value $[\text{H}_4\text{SiO}_4]_{\text{sat}}$ as the product $(\text{SA}/\text{V})t$ increases. Tests conducted for larger products of $(\text{SA}/\text{V})t$ will attain a more advanced stage of corrosion.

The effects of SA/V on the dissolution of PNL 76-68 glass have been studied in tests conducted at 90°C with monolithic samples run at SA/V of 1, 10, and 100 m⁻¹ [72,73]. In Fig. 30, the boron and silicon concentrations are shown plotted against the product of the SA/V and the reaction time. Both the boron and silicon concentrations increase linearly with the product $(\text{SA}/\text{V})t$, and data from tests run at different SA/V can be represented by a single line. As evidenced by the linearity of data at each SA/V over the times tested, these test conditions maintain rather dilute solutions and corrosion is dominated by ion exchange. These tests were conducted for a maximum of $(\text{SA}/\text{V})t = 2800 \text{ d/m}$, and are not relevant to the use of the SA/V to attain saturated solution conditions.

Static leach tests were conducted using monolithic and crushed SRL 165 glass at 90°C [195]. Tests with monolithic samples were conducted at SA/V of 10, 30, and 50 m⁻¹, and tests with crushed glass (-40+80 mesh) were conducted at SA/V of 30, 50, and 100 m⁻¹. The surface area of the crushed glass was calculated assuming the grains were spheres with a diameter of 298.5 µm and the surface area of the monoliths were measured geometrically. The measured boron concentrations are plotted against the product $(\text{SA}/\text{V})t$ in Fig. 31. The open symbols represent tests with crushed glass samples and the filled symbols represent tests with monolithic samples. Notice that the tests with monolithic samples have lower measured boron concentrations than tests with crushed samples at common $(\text{SA}/\text{V})t$. This may be due to the higher reactivity of the crushed glass or to the actual surface area of the crushed glass being greater than estimated. Instead of the measured solution concentrations, the authors plotted the normalized leach rates against the product $(\text{SA}/\text{V})t$. This has the effect of canceling the estimated surface area of the samples from the plot.

Chandler et al. conducted tests to assess the applicability of $(\text{SA}/\text{V})t$ as a scaling factor to relate tests with monolithic samples and tests with crushed glass [196]. Tests were conducted in deionized water at 90°C for times up to 28 days using SRP black frit waste glass. Monolithic samples were tested at SA/V ratios of 10 and 100 m⁻¹, and crushed samples of the -40+60 mesh

Fig. 30. Concentration vs. (SA/V)t for Release of Boron and Silicon from PNL 76-68 Glass at 90°C. (□) Si and (○) B at 1 m⁻¹; (■) Si and (●) B at 10 m⁻¹; (□) Si and (○) B at 100 m⁻¹ (after [72,73]).

Fig. 31. Measured Boron Concentrations in Tests with SRL 165 Glass vs. (SA/V)t: Tests with Monolithic Samples at (●) 10, (■) 30, and (◆) 50 m⁻¹ and Tests with Crushed Samples at (□) 30, (◇) 50, and (○) 100 m⁻¹ (after [195])

size fraction were tested at SA/V ratios of 100 and 1000 m^{-1} . Figure 32 shows the boron concentration plotted against the product (SA/V)t for these test data. Data from both tests with monolithic samples and tests with crushed samples can be described using a single curve. The curve is not a straight line, as was seen in the data of Pederson et al. This indicates that the affinity term affected the corrosion to a small degree, but the data are not relevant to near-saturated conditions. These tests were conducted for a maximum of $(\text{SA}/\text{V})t = 2.8 \times 10^4 \text{ d/m}$.

Note in Fig. 32 that tests with monolithic samples and with crushed samples at 100 m^{-1} give almost identical results. This implies that the estimated surface area of the crushed glass is accurate and that the monolithic and crushed glass reacts at a similar rate. Of course, the similarity depends on the surface area assigned to the crushed glass. The authors mentioned that the surface area of the crushed glass was measured using gas adsorption, but did not report the measured area. Neither were the mass of crushed glass or the mass of leachant solution reported. It is possible that the surface area of the crushed glass was determined by matching the data to that from tests with monolithic samples. The greatest difference between tests with monolithic samples and those with crushed samples occurs after 1 day of reaction, when the boron concentration in the test with the monolith is 2.54 ppm and in the test with the crushed glass is 1.50 ppm. The boron concentration is higher in tests with crushed glass than in corresponding tests with monoliths at all other test times, however. The authors ascribe the difference in the 1-day tests to differences in surface morphology, which are expected to become less significant as the glasses corrode. However, while the morphologies may become similar, changes in the solution chemistry due to the initial corrosion will significantly affect the continued corrosion behavior. This is evidenced in tests with crushed samples, which consistently had slightly higher pH values than corresponding tests with monoliths, which indicates that more dealcalization occurred early in the tests with crushed glass.

Fig. 32. Boron Release vs. (SA/V)t in Tests of SRP Black Frit Glass at Various SA/V Ratios: (●) 10 m^{-1} and (▲) 100 m^{-1} with Monolithic Samples and (■) 100 m^{-1} and (◆) 1000 m^{-1} with Powdered Samples (after [196])

Monolithic samples of SRL 165 glass were corroded in deionized water at 90°C at SA/V of 7.9 and 85 m⁻¹ for times up to 56 days [197]. The silica concentrations are plotted against the product (SA/V)t in Fig. 33. Although the authors fit all the data using the single curve shown in the figure, it is obvious that test results at the lower SA/V deviate from the curve as (SA/V)t increases. (The tests at the lower SA/V were only progressed to a (SA/V)t value of 442.4 d/m.) The deviation can be attributed to the difference in the pH values attained in the tests at different SA/V: the pH values in tests at 85 m⁻¹ were consistently about one half unit higher than those in tests at 7.9 m⁻¹.

Hench and coworkers have studied the corrosion of simple alkali silicate glasses to determine the kinetics of the dealcalization process [198,199]. As a part of their investigations, the corrosion of monolithic and crushed glass samples was studied. Figure 34 reproduces the measured silica concentrations in tests conducted with monolithic and crushed Li₂O•2SiO₂ glass at 50°C [198]. Tests with monolithic samples were conducted at a SA/V of 77 m⁻¹, while tests with crushed glass samples were conducted at a reported SA/V of 43 m⁻¹. The authors fit the data for tests with monolithic samples to two lines, as shown in Fig. 34, and interpreted the change in slope after about 300 days to indicate a change in the kinetics from t^{1/2} to t¹ dependence. They drew a single curve through the data for tests with crushed glass and implied that the tests with powders required longer reaction times to change kinetics. However, as shown in Fig. 34, the results of tests with crushed glass can be fit by lines having the same slope and the same change in slope as those drawn through the data from tests with monolithic samples. Thus, the monolithic and crushed glass react identically. The difference in measured silica concentrations is due to the different SA/V of the tests. The differences suggest that the SA/V of the tests with crushed glass is about one third that of the monolithic tests.

Fig. 33. Measured Silicic Acid Concentration vs. (SA/V)t for Tests with SRL 165 Glass at SA/V Ratios of (Δ) 7.9 and (○) 85 m⁻¹ (after [197])

Fig. 34. Measured Silica Concentration vs. Log Time for Tests with $\text{Li}_2\text{O} \cdot 2\text{SiO}_2$ Glass:
 (○) Monolithic Samples at 77 m^{-1} and (Δ) Crushed Samples at 43 m^{-1} (after [198])

Another study to determine the usefulness of $(\text{SA}/\text{V})t$ as a scaling factor for tests with monolithic samples and crushed glass samples was carried out by S. Bates et al., who performed static leach tests in deionized water at 90°C using HW-39 glass at SA/V ratios of 10, 530, 2000, and $20,000 \text{ m}^{-1}$ [133]. Tests at 10 m^{-1} were performed using monolithic glass samples following the MCC-1 protocol, while tests at 530, 2000 and $20,000 \text{ m}^{-1}$ were performed using crushed glass in the -100+200 mesh size fraction following the PCT protocol. The surface area of the crushed glass was approximated by assuming the grains were spheres with a diameter equal to the average mesh size. The leachates from tests with crushed glass were filtered using 450 nm filters to remove suspended glass particles, while the leachates from tests with monolithic samples were not filtered.

The boron concentrations in the leachates and the solution pH values are plotted against the product $(\text{SA}/\text{V})t$ in Figs. 35a and b. The data from tests at 10, 530, and 2000 m^{-1} may be represented by a common curve, within experimental error, but the tests at $20,000 \text{ m}^{-1}$ clearly deviate from this curve. Again, the curvature of data from tests at common SA/V conducted for various time periods indicates that the silicic acid released to solution is affecting the continued glass corrosion rate. These tests were conducted for a maximum of $(\text{SA}/\text{V})t = 5.6 \times 10^5 \text{ d/m}$. The test matrix use by Bates et al. includes tests at different SA/V but overlapping $(\text{SA}/\text{V})t$ which allow direct comparison of the reaction progress. Previously discussed tests did not have overlapping data. Although the data from tests conducted at SA/V of 2000 m^{-1} or less show the same general trend, the tests conducted at higher SA/V consistently have a higher boron release than tests at lower SA/V but at the same $(\text{SA}/\text{V})t$. The difference is greatest between tests at 2000 and $20,000 \text{ m}^{-1}$. These data suggest that the deviations from $(\text{SA}/\text{V})t$ scaling increases with the SA/V and with the product $(\text{SA}/\text{V})t$, and show that tests should be conducted for large values

Fig. 35. Corrosion of HW-39 Glass at Various SA/V Ratios: (a) B, ppm, vs. (SA/V)t and (b) pH vs. Time: (●) 10 m⁻¹, (▲) 530 m⁻¹, (■) 2000 m⁻¹, and (◆) 20,000 m⁻¹ (after [133])

of $(SA/V)t$ to determine the effects of SA/V on the corrosion behavior and to determine if tests at high SA/V can be used to accelerate the corrosion and to characterize advanced corrosion behavior. Note that the boron concentrations in tests with monolithic samples at 530 m^{-1} and the tests with crushed glass at 2000 m^{-1} differ by an extent similar to the difference between the tests at 10 m^{-1} and 530 m^{-1} . This indicates that the surface area of the crushed glass is well approximated by assuming the grains are spheres with a diameter equal to the average mesh size.

Some insight into the effects of SA/V is gained by considering the solution pH values attained in the tests conducted by S. Bates et al. The pH values measured at room temperature for these tests are seen to increase with the SA/V of the test and with the reaction time, shown in Fig. 35b. (The pH values shown in Fig. 35b were tabulated by S. Bates et al., although values that were calculated for 90°C are plotted in [133]. The trends of the data for tests at different SA/V are the same, with the 90°C pH values being about 2 pH units lower than those measured at room temperature.) Both the leachate pH and the boron concentration are consistently higher in the tests at higher SA/V at all $(SA/V)t$ tested. The difference is greatest at high SA/V . S. Bates et al. recognized that the higher solution pH values promote glass dissolution and suggested that test data may scale better under buffered conditions.

The effects of the SA/V on the corrosion of R7T7 glass was investigated by Vernaz et al. by reacting R7T7 glass in deionized water for reaction times up to 364 days [167]. Tests at 400 m^{-1} were performed using monolithic samples, while tests at higher SA/V were attained by using crushed glass of various size fractions: 2000 m^{-1} using the $40\text{-}50\text{ }\mu\text{m}$ size fraction, 8000 m^{-1} using the $100\text{-}125\text{ }\mu\text{m}$ size fraction, and $20,000\text{ m}^{-1}$ using the $400\text{-}500\text{ }\mu\text{m}$ size fraction. The SA/V of tests with crushed glass was based on assuming the grains to be spheres with a diameter equal to the average mesh size. The results are plotted in Figs. 36a-36d. The lines drawn through the boron data were interpreted by Vernaz et al. to represent the residual rates at each SA/V .

In tests at 400 m^{-1} , the silicon concentration increases at the same rate as the boron concentration through about 273 days, as seen by drawing a line through the silicon data having the same slope as the line drawn by Vernaz et al. through the boron data. This indicates that corrosion is not occurring under silica saturation conditions and that the rate quoted is not the residual rate. Note that the data at 364 days suggest a slowing of the corrosion rate. Longer reaction times are needed to characterize the corrosion rate at 400 m^{-1} .

Tests at 2000 m^{-1} also show the silicon concentration to increase at the same rate as the boron concentration through the longest times tested, as seen by drawing a line through the silicon data having the same slope as the line drawn by Vernaz et al. through the boron data. This indicates that corrosion is not occurring under silica saturation conditions in these tests either, and that the rate quoted is not the residual rate. Longer reaction times are needed to characterize the corrosion rate at 2000 m^{-1} .

The silicon concentration increases at a lower rate than the boron concentration in the tests conducted at 8000 m^{-1} also show, as seen by drawing a line through the silicon data. The observation that the silicon concentration continues to increase, albeit slower than the boron concentration, indicates that corrosion is not occurring under silica saturation conditions in these tests, and, again, the rate quoted is not the residual rate. Longer reaction times are needed to characterize the corrosion rate at 8000 m^{-1} .

Fig. 36. Measured Concentrations of ($\diamond$) Boron and ($\blacklozenge$) Silica in Tests with R7T7 Glass: (a) at 400 m^{-1} with Monolithic Samples, (b) at 2000 m^{-1} with Crushed Samples, (c) at 8000 m^{-1} with Crushed Samples, and (d) at $20,000 \text{ m}^{-1}$ with Crushed Samples (after [167])

Tests conducted at $20,000 \text{ m}^{-1}$ show the boron concentration to be essentially constant beyond about 112 days. The total silicon concentration increases through about 28 days, then decreases at longer time periods. Vernaz et al. showed that the decrease was due to the settling of iron silicate colloids from solution. They also filtered the leachate solutions through an approximately 1.8 nm filter to remove colloidal material, and found that the concentration of dissolved silicon (i.e., that which is not removed by filtration) to remain constant at about 120 ppm beyond 28 days. The boron concentrations in these tests do represent the residual rate under silicon saturation, and suggest that the rate is immeasurably low. Vernaz et al. reported the rate to be $<1.5 \times 10^{-4} \text{ g/m}^2/\text{d}$.

Although the authors concluded that the residual rate decreased with the SA/V of the test, consideration of their data strongly suggests that tests conducted at SA/V less than $20,000 \text{ m}^{-1}$ did not attain silica saturation and do not represent the residual rate. The observation that higher rates were measured at lower SA/V simply indicates that the corrosion is occurring under unsaturated conditions. The authors do note that higher solution pH values are attained in tests at higher SA/V. The "mean leachate pH" values measured at 25°C are: 8.91 in tests at 400 m^{-1} , 9.28 in tests at 2000 m^{-1} , 9.61 in tests at 8000 m^{-1} and 9.77 in tests at $20,000 \text{ m}^{-1}$. They also conclude that "the significant slowdown of the final rate is not due to diffusion of leached species through a growing surface layer, since the altered glass thickness diminishes as the SA/V ratio increases" [167], although they do not present measured layer thicknesses in the paper.

Short-term corrosion tests conducted by Feng et al. using several different glasses have shown various correlations between the extent of glass corrosion and the product $(\text{SA/V})t$ [159]. Glasses were reacted using a modified MCC-3 procedure (similar to the PCT procedure) wherein a small fraction of the solution was sampled periodically and replaced with fresh leachant. As discussed in Section 2.5.2.3, such a periodic replenishment of the leachate solution will greatly complicate interpretation of the test results. Continued dilution of the leachate solution will cause a cycling of the corrosion rate between a high value after replenishment and a much lower value after the solution reconcentrates. As the key effect of the SA/V on test results is through dilution of corrosion products, test results will be affected by both the effects of the SA/V and the effects of the leachate replacement. Despite the inappropriateness of the test method, departure from $(\text{SA/V})t$ scaling was most evident in tests conducted at large values of SA/V, as seen in static leach tests. The authors suggested that the observed deviation from $(\text{SA/V})t$ scaling may be due to pH effects and to mass transport effects or may arise from the formation of protective alteration layers. Although tests at higher SA/V attain higher solution concentrations, the release rates per unit area of glass are lower in tests at higher SA/V than in tests at lower SA/V. Therefore, the differences in the layer thickness generated in tests at high and low SA/V cannot account for observed effects of SA/V.

Long-term tests were conducted at ANL that attained larger values of $(\text{SA/V})t$ than had been attained previously [67]. (The testing program is summarized in Section 9.) Tests were performed using glasses SRL 131 and SRL 202 that were doped with technetium and transuranics (and are referred to as SRL 131A and SRL 202A) at 10, 340, 2000, and $20,000 \text{ m}^{-1}$. Tests were conducted in both deionized water and in a tuff groundwater. Tests at 10 and 340 m^{-1} were performed following a modified MCC-1 procedure and tests at 2000, and $20,000 \text{ m}^{-1}$ were performed following a modified PCT procedure. Differences in the extent of corrosion of SRL 202 glass in deionized water and tuff groundwater were negligible at reaction times beyond a few days [66]. Figure 37a shows the boron concentrations plotted against the product $(\text{SA/V})t$ for tests with SRL 202A glass in a tuff groundwater solution. These data clearly cannot be fitted by a single curve. Note that the deviations from a common curve are greater at larger values of $(\text{SA/V})t$, as seen earlier in the data of S. Bates et al. [133]. Figure 37b shows the measured pH values plotted against the reaction time for tests conducted at the different SA/V ratios. In these tests, the increase in the solution pH is a consequence of the natural corrosion process.

In other tests, radiolysis of the air in the test vessel by radioactive glass has been seen to affect the glass corrosion behavior. A sudden increase in the corrosion rate of a glass made from SRL 200 frit (referred to as SRL 200S glass) similar to that observed by Ebert and Bates was observed in static tests performed by Feng et al. [200]. Tests were conducted at 90°C following the PCT method B protocol in tuff groundwater and at a SA/V ratios of 2000 and $20,000 \text{ m}^{-1}$. Tests were performed using the -100+200 mesh size fraction. Tests conducted under analogous conditions using a glass made from the same frit plus added radioactive sludge (referred to as SRL 200R glass) did not show a corresponding sudden increase, however. The two glasses reacted

Fig. 37. (a) Measured Boron Concentration vs. $(SA/V)t$ and (b) Measured pH vs. Time for Tests with SRL 202 Glass at SA/V Ratios of (●) 10, (■) 2000, and (◆) 20,000 m^{-1} (after [67])

identically in all tests at 2000 m^{-1} (results were presented through 364 days) and in through about 182 days in tests at $20,000 \text{ m}^{-1}$. Beyond 182 days in tests at $20,000 \text{ m}^{-1}$, tests with the nonradioactive glass showed a significant increase in the release rate of soluble glass components. The normalized mass losses of boron, lithium, sodium, and silicon in tests conducted at $20,000 \text{ m}^{-1}$ with SRL 200R and SRL 200S glasses are shown in Figs. 38a and 38b. (Data for the release of sodium from the SRL 200R glass are not distinguishable in the plots published by Feng et al.)

All four elements are released suddenly (lithium, boron, and sodium after 330 days and silicon after 364 days), but that the release at the next longest test period is different for the different elements: lithium is released to a slightly greater extent after 364 days than it is after 330 days, silicon is released to a similar extent after 390 days as before the increased release, slightly less sodium and boron is released after 390 days than after 364 days. Also, the release is not stoichiometric. The normalized mass losses decrease as $B > Na \gg L \gg Si$; this shows the increase is not a testing artifact (such as including glass grains in the analyzed solutions). The boron mass loss after 330 days is very near the maximum normalized mass loss for tests with the $-100+200$ mesh size fraction, which is 50 m^{-1} . This indicates that very little unreacted glass remains and that mass losses at longer reaction times are not expected to show much increase. The slight decrease in the boron and sodium mass losses after 364 days may indicate that the corrosion rate was not accelerated at the same time in tests at 330 and 364 days, or may be due to analytical uncertainties. The decreased silicon mass loss after 390 days may indicate a settling of colloidal silicon.

The authors attributed the rate increase to the formation of clinoptilolite, $(\text{Na,K})_6(\text{Al}_6\text{Si}_{30}\text{O}_{72}) \cdot 20\text{H}_2\text{O}$, which was abundant in the tests with the nonradioactive glass but not found in the tests with the radioactive glass. The authors concluded that the lower solution pH values in the tests with the radioactive glass prevented the formation of clinoptilolite and the concomitant increase in the corrosion rate. Radiolysis of the air in the test vessel resulted in the lowering of the pH value in tests with the radioactive glass. Figure 38c shows the solution pH values measured in the tests at $20,000 \text{ m}^{-1}$ with SRL 200R and SRL 200S glasses. The pH values (which were measured at room temperature) remain below about 11.65 in tests with SRL 200R glass, but rise above 12 in tests with SRL 200S glass. The authors speculated that the formation of clinoptilolite occurred in solutions having high pH values under the test conditions. Thus, the key effect of the pH in these tests was on the formation of secondary phases, which then affected the glass corrosion rate.

4.3 Modeling the Effects of the Solution pH

The differences in the solution concentrations attained in tests run at different SA/V can be attributed to the effect of the pH on the corrosion rate. Tests under conditions where the affinity term is maintained near one have clearly shown that the corrosion rate is sensitive to the solution pH (see Figs. 16 and 17 [70,92]). The rate coefficient k_+ has a pH dependency that can be written explicitly as $k_+ = k_f (\text{H}^+)^{-0.4}$, where k_f is the pH-independent reaction coefficient and the exponent was determined experimentally and is valid for pH values greater than about pH 7. Substituting this expression for k_+ into Eq. 16 yields

$$[\text{H}_4\text{SiO}_4] = [\text{H}_4\text{SiO}_4]_{\text{sat}} \left(1 - \exp \left\{ \frac{-(\text{SA/V})tk_f(\text{H}^+)^{-0.4}}{[\text{H}_4\text{SiO}_4]_{\text{sat}}} \right\} \right) \quad (17)$$

Fig. 38. Normalized Mass Loss vs. Reaction Time for Tests with SRL 200R and SRL 200S Glasses at $20,000 \text{ m}^{-1}$: (a) (●) B from SRL 200R, (○) B from 200S, (◇) Na from 200S; (b) (◆) Li from SRL 200R, (◇) Li from 200S, (▲) Si from 200R, (Δ) Si from 200S; and (c) Measured pH for Tests with (●) SRL 200R and (○) SRL 200S Glass (after [200])

From this expression, the coefficient for scaling static test results is $(SA/V)t(H^+)^{-0.4}$ (k_f and $[H_4SiO_4]_{sat}$ are constant with respect to time and solution pH). Therefore, tests performed for common values of $(SA/V)t$ will generate the same silicic acid concentrations only if the solution pH values are the same. Since the SA/V of a static test also determines the extent of dilution of hydroxide generated during dealkalization, tests at different SA/V will generally attain different pH values. Since dealkalization affects the solution chemistry primarily during the initial stage of corrosion, the solution pH usually approaches a constant value as corrosion proceeds (see Fig. 37b, for example). Of course, the change in the pH depends strongly on the amounts of alkali metals in the glass and the buffering capacity of silicic and boric (and perhaps other) acids released from the glass as it corrodes. The pH of the solution is expected to scale with the SA/V of the test better at short times and at small values of SA/V, for which the corrosion rate is not strongly affected by the affinity term.

The use of tests at high SA/V to evaluate advanced stages of corrosion requires solution to a rate equation that depends on both the silicic acid concentration and the pH, such as that given by Eq. 15, where $k_+ = (H^+)^h$. The results of tests conducted at different SA/V can only be directly compared when the effects of either the silicic acid concentration or the pH on the rate can be isolated. That is, the pH must be constant to compare the effects of the silicic acid concentration on the rate and the silicic acid concentration must be constant to compare the effects of the pH. Tests in which the pH and silicic acid concentrations are free to vary cannot be compared directly. Instead, a rate equation that accounts for both effects must be used to compare the data. For example, the results of tests being conducted at ANL have been utilized to demonstrate how the pH can be incorporated into the model being developed at LLNL [176]. Therefore, the deviations observed to occur in plots of solution concentrations against the product $(SA/V)t$ can be attributed to differences in the pH, and these differences can be accounted for in the rate equation used to describe glass corrosion. Tests at high SA/V can be used to characterize advanced stages of corrosion only if differences in the solution chemistry are accounted for by modeling.

First, the differences in pH must be accounted for when comparing the results of tests conducted at different SA/V. The high pH values attained in tests at high SA/V are not an artifact of the test method, they are due to the limited volume of water available to dilute hydroxide generated during glass corrosion. We also note that solution pH values are usually measured at room temperature while tests are conducted at much higher temperatures. Cooling the solution from the test temperature to room temperature can change the solution pH significantly. The pH values at the test temperature can be calculated from the measured solution chemistry using thermodynamic codes. The pH values at the test temperature should be used in the rate equation. The solution concentrations of dissolved components are not expected to be affected by cooling the solution, although in concentrated solutions some components may be removed from solution by precipitation of saturated phases. Solutions are generally acidified to maintain components in solution.

Second, the alteration layers formed on the glass surface may vary with the SA/V of the test. The differences may affect the disposition of some glass components and waste components as the glass corrodes. The amount of glass that must dissolve to saturate the solution decreases as the SA/V increases, so thicker layers form at lower SA/V. While the layers do not significantly affect the glass corrosion rate, they do interact with released materials, including radionuclides, to affect their solution concentrations. Thus, the behavior of waste components may not track the behavior of the glass matrix. (For example, the release of transuranic elements and other components from SRL 202A glass does not follow the increased release of boron and alkali metals shown in Fig. 37a. See also Section 7.)

Third, utilization of the results obtained in tests at high SA/V for predicting advanced corrosion behavior should be related to anticipated disposal conditions, and other environmental effects on the solution chemistry should be taken into account. For example, radiolysis of the air and groundwater to generate nitric acid will lower the pH but not affect the silicic acid concentration. Thus, the pH and silicic acid levels attained in tests at high SA/V will not accurately simulate the corrosion environment. Such effects may greatly affect the glass corrosion behavior [200,201].

4.4 Effects of SA/V on Alteration Layers

Differences in the alteration layers generated when a glass is corroded in tests at different SA/V occur because (1) more glass must react in a test at low SA/V than in a test at high SA/V to attain the same level of solution saturation, and (2) the quicker approach to saturation in tests at high SA/V means the surface is exposed to more concentrated solution chemistries during the test period. For glasses that dissolve nonstoichiometrically, thicker layers are generated in tests at low SA/V than are generated in tests at high SA/V. However, the layers formed on glasses reacted in tests at high SA/V mature faster because they are exposed to more concentrated solutions. Tests at low SA/V can be used to characterize the initial development of the alteration layers because the layers are relatively thick and form in the presence of a solution that remains relatively dilute for long test periods. Interactions between the developing layer and the solution can therefore be tracked as a function of time. Interactions between the layers formed at high SA/V occur more rapidly due to the quick saturation of the solution. While tests conducted at low SA/V can be used to characterize the early stages of layer formation and tests conducted at high SA/V can be used to characterize the later stages of layer formation, differences in the solution chemistry must be accounted for. Specifically, the solution pH will be significantly higher in tests conducted at high SA/V than in tests conducted at low SA/V, so that the layer will develop under different conditions. Alteration phases formed in highly basic solutions in tests at high SA/V may not form in less basic conditions in tests at low SA/V.

Houser et al. analyzed the films formed on monolithic samples of three different waste glasses after corrosion at SA/V of 10 and 0.1 m^{-1} (10^{-1} and 10^{-3} cm^{-1}) for up to 100 hours at 90°C [202]. While the film thicknesses formed on all glasses increased with time, the films formed in tests conducted at SA/V of 10 m^{-1} were slightly thicker than those formed in tests at 0.1 m^{-1} after the longest test time. Although solution concentrations were not reported, the observation of thinner layers in tests at lower SA/V indicates that more surface dissolution occurred in tests at 0.1 m^{-1} , which maintained more dilute solutions.

Oversby and Phinney characterized the layers that formed on SRL 165 glass in MCC-1 tests using secondary ion mass spectrometry. Tests were conducted for up to 91 days at 90°C in either deionized water or a solution containing 60 ppm silica, 183 ppm sodium, 200 ppm bicarbonate, and 250 ppm nitrate [88]. The layers formed on the "as cast" faces were analyzed to avoid any effects of mechanical preparation on the layer development. Analysis showed the alkali metals and boron to be depleted in the layer, and alkaline earths, iron and manganese to be enriched in the layer, relative to the glass. Layers were found to develop in "an orderly process" in the silicate leachant, while the results of layer formation in deionized water were "more complex and less reproducible" [88]. The authors attributed the difference to the faster dissolution that occurs in the deionized water. The corrosion in the silicate solution decreased with the reaction time, as evidenced by both solution results and the layer thickness as defined by the depletion depth of lithium. A layer about $1.3 \mu\text{m}$ thick had formed after 91 days. Oversby and Phinney noted that the structure of the layer developed over time as more stable alteration phases formed.

Tests conducted using the same SRL 165 glass as used by Oversby and Phinney had been conducted earlier under modified MCC-1 conditions (monolithic samples reacted in J-13 groundwater at 90°C and at a SA/V of 30 m⁻¹) [95]. The groundwater solution had slightly less silica than the solution used by Oversby and Phinney. Analysis by transmission electron microscopy of the layers formed as a function of the reaction time reveal how the layer structure evolves [87]. Tests conducted for short times resulted in the formation of an amorphous layer that was depleted of soluble glass components, such as alkali metals and boron, consistent with the composition determined by Oversby. After longer corrosion times, the layer was seen to crystallize and separate from the underlying glass. The layer is comprised of small laths of clay that have formed on a "backbone" of an iron-rich amorphous phase. The layer is seen to have completely crystallized and separated from the glass after about 278 days. The underlying glass continues to dissolve nearly congruently beneath the layer. Soluble material released from the glass is presumed to enter solution and insoluble material to be incorporated into the clay.

Analysis of the alteration layers formed in modified MCC-1 tests with SRL 165 glass by Kamizono et al. by secondary ion mass spectrometry showed boron to be depleted to a depth of about 1.5 µm in tests at 8.5 m⁻¹ and about 3 µm in tests at 7.9 m⁻¹ after 28 days of reaction [197]. The normalized mass loss of boron after 28 days was about 3 times higher in tests at 7.9 m⁻¹ than in tests at 85 m⁻¹. The difference between the thickness predicted based on boron release and that measured is due to dissolution of the glass surface, which is expected to be greater in the tests at 7.9 m⁻¹ than in tests at 85 m⁻¹.

Modified MCC-1 tests conducted in deionized water at 90°C using monolithic samples of SRL 131 glass at SA/V of 10, 50, and 100 m⁻¹ for 100, 20, and 10 days, respectively [203]. All tests were conducted for the same product (SA/V)t, namely 1000 d/m. The solution pH and the solution concentrations of most glass components increased slightly with the SA/V (results were about 10% higher in tests at 100 m⁻¹ than those at 10 m⁻¹). The thickness of the layers were measured to be 40, 3, and 1 µm for tests conducted at 10, 50, and 100 m⁻¹. Theoretical maximum layer thicknesses calculated from the measured boron concentrations, assuming the weight fraction of boron in the glass is 0.04 (see Appendix B) and the glass to have a density of 2.7 g/cm³, are 22.5, 4.6, and 2.6 µm for tests conducted at 10, 50, and 100 m⁻¹, respectively. The measured values are in good agreement at the higher SA/V, but about a factor of 2 too large at 10 m⁻¹. The error is likely due to the layer not being cross-sectioned normal to the glass surface. The layers formed at 50 and 100 m⁻¹ may be thinner than predicted due to dissolution of the outer surface.

The layer formed at 10 m⁻¹ was found to be more complex than those formed in tests at the higher SA/V. It included more phases and sublayers than the layers formed at 50 m⁻¹ and 100 m⁻¹, which were found to be structurally similar. The similarity of the solution chemistries imply that the differences are due to the different reaction times used in the tests. The authors conclude that phases observed in tests conducted at 10 m⁻¹ were absent from layers formed at 50 m⁻¹ and 100 m⁻¹ due to insufficient time for the phases to be precipitated [203]. However, the thinness of the layers formed at 50 and 100 m⁻¹ may have prevented resolution of similar sublayers. The alteration phases observed on the sample reacted for 100 days at 10 m⁻¹ are consistent with those formed on SRL 131 glass under the same test conditions after longer reaction times [62]. Analysis of samples reacted for shorter time periods showed the layer to evolve from a single clay phase to complex layer containing several distinct phases.

Tests conducted with SRL 165 glass under modified MCC-1 conditions (monolithic samples reacted in J-13 groundwater at 90°C and at a SA/V of 30 m⁻¹) show a less complicated layer to form [87]. Tests conducted for short times resulted in the formation of an amorphous layer that was depleted of soluble glass components, such as alkali metals and boron. After longer corrosion times, the layer was seen to crystallize and separate from the underlying glass. The layer formed on SRL 165 glass at 30 m⁻¹ is different than that formed on SRL 131 glass in tests at 10, 50, or 100 m⁻¹ in that the layer formed on SRL 165 glass completely crystallizes and separates from the glass. The underlying glass continues to dissolve nearly congruently beneath the layer. In contrast, the layer formed on SRL 131 glass does not completely crystallize and does not separate from the glass. While the compositions of SRL 131 and SRL 165 glasses are quite similar (see Appendix B), the alteration layers formed are quite different.

The alteration layers formed on SRL 202 glass in tests at 340 m⁻¹ using monolithic samples and in tests at 2000 m⁻¹ using crushed glass were found to develop similarly [64]. (See Sec. 9.3 for a more detailed discussion of layer formed during these tests.) The layers formed in tests at 20,000 m⁻¹ through 182 days were the same as those formed in tests at 2000 m⁻¹. The layers are comprised of laths of clay formed parallel to the glass surface. In tests at 20,000 m⁻¹ conducted for 364 days or longer, a thick layer of the same clay material forms between the layer of clay laths and the glass. The inner layer contains smaller crystallites of the same clay that forms the outer layer. The sudden increase in the layer thickness corresponds to a sudden increase in the release of soluble glass components from the glass [65]. The increased corrosion rate has been attributed to the formation of secondary phases from the leachate solutions. The thickness of the outer layer of clay is about the same after 182 days (before the corrosion is accelerated) and after 364 days (after the corrosion has been accelerated). This indicates that the outer clay layer is unaffected by the significant changes in the solution chemistry that lead to a significant increase in the glass corrosion rate. The clay layer formed on the glass surface does not affect either the solution chemistry or the glass corrosion rate. It is also interesting to note that the highest corrosion rate occurs when the alteration layer is the thickest. This indicates that the layer does not act as a diffusion barrier, and that sloughing of the layer off the glass is not likely to affect the corrosion rate of SRL 202 glass.

The appearance of the layers formed after acceleration of the corrosion rate is very similar to that formed in tests with SRL 131 at 100 m⁻¹ in the work of Mazer et al. [203], though the layer formed on SRL 202 glass is much thicker than that formed on SRL 131. As SRL 131 glass is known to be less durable than SRL 202 glass [67], this implies that the evolution of the alteration layer is affected by the reaction time and the glass composition. While tests conducted with SRL 202 glass at different SA/V attained very different solution chemistries, similar layers developed at all SA/V. This indicates that the development of the layers on SRL 202 glass is not affected by the solution chemistry.

4.5 Tests Conducted to Attain Highly Concentrated Solutions

Because the solution composition controls the glass reaction rate and because the SA/V ratio of a static test determines the extent of dilution of the solution, the SA/V used in a test provides a valuable means of attaining highly concentrated solution conditions. As discussed earlier, the reaction affinity can be kept near one, and the forward reaction rate can be determined by maintaining dilute solutions at very low SA/V in static tests or in dynamic tests. Conversely, the reactivities of glasses in highly concentrated leachate solutions where the reaction affinity is very small can be characterized by performing tests at very high SA/V. Such tests provide valuable insight into glass behavior in highly concentrated solutions, such as those that might eventually contact the waste form in a repository.

Crushed glass is used routinely to achieve high SA/V ratios. Such tests provide for analyses of both the leachates and corroded solids, although the very large number of reacted grains complicates the cataloging of all secondary phases that may be generated. Very high SA/V ratios have also been attained by reacting monolithic samples in vapor at elevated temperatures [104,106-108,111,139]. In a vapor environment, reaction occurs in a thin film of condensed water, and highly concentrated solutions are generated after very little glass has reacted [111]. Secondary phases that form in the leachate film remain on the sample surface, where they can be readily identified and cataloged. Leachate analyses cannot be performed with this method because the water evaporates during test termination; however, residual salts can be rinsed from the sample or redissolved in fresh condensate and analyzed [81].

The results of tests at very high SA/V ratios are consistent with the effects of SA/V predicted by Eq. 15, namely, that highly concentrated solutions are quickly attained and the reaction rate quickly becomes very low. Because glass is metastable and cannot equilibrate with the leachate, the glass corrodes at a "final reaction rate" even after the solution becomes saturated [151,168]; the continued corrosion has been attributed to a "residual" or "contextual" reaction affinity [166]. Continued glass reaction under saturated conditions (where the reaction affinity becomes small), even though the reaction rate may be immeasurably low [24,161,166,168].

Tests performed in highly concentrated solutions have shown that the low residual rate attained under saturated conditions does not necessarily represent the long-term reaction rate. An increase in the glass reaction rate has been observed in tests where secondary phases have precipitated from the fluid after long reaction times [65,67,116]. For example, in Fig. 37a, the boron concentration suddenly increases after 182 days of reaction in tests conducted at $20,000 \text{ m}^{-1}$. This change is indicative of an increase in the reaction rate coincident with secondary phase formation. Such increases can be explained by the effects of secondary phase formation on the solution chemistry, which will result in an increased reaction rate if the solubility limit of silicic acid is maintained at a value less than the saturation concentration, or if the pH increases [143]. Enhanced reaction has also been observed in the presence of clays [204]: silicon is removed from solution as more clay is formed, so the solution chemistry is affected. The effect of added clays is different from that of clays which form during glass corrosion. Clays formed as alteration layers on the glass surfaces during corrosion do not affect the corrosion rate. Although silicic acid has been identified as the dominant species affecting the reaction [153,154,205], recent results suggest that other solution species may also influence the observed reaction rates under some conditions [24,71]. The relative importance of the silicic acid concentration on the glass corrosion rate may depend on the glass composition and the reaction conditions. Thus, the effect of secondary phase formation on other solution components may also affect the glass corrosion rate.

4.6 Summary

The observed effects of the SA/V ratio used in static leach tests can be accounted for by the differences in the extent of dilution of corrosion products, especially the pH and the silicic acid concentration. While the pH and silicic acid concentrations will be affected at all SA/V, the effects of the pH on the corrosion rate become experimentally measurable only when the pH exceeds about pH 9. Below pH 9, the SA/V primarily affects only the silicic acid concentration. The results of tests conducted at different SA/V can be directly related through the product of SA/V and the reaction time only if the pH remains below about pH 9. If the pH exceeds about pH 9, then the difference in pH values of the tests being related must be accounted for analytically.

5.0 OBSERVED EFFECTS OF THE FLOW RATES IN DYNAMIC LEACH TESTS

The difference between static and dynamic tests is that in dynamic tests, the solution concentrations of dissolved glass components do not become concentrated in the solution in contact with the corroding glass as quickly (or as completely) as in static tests because the leachate solution is continuously or periodically replaced with fresh leachant. The effect of the leachant flow rate in dynamic tests is like that of the SA/V in static tests: it primarily affects the silicic acid concentration and the pH at the glass surface. As described in Section 2.5.2, dynamic conditions can be achieved by either pumping solution through a reaction cell or periodically removing a portion of the leachate solution and replacing it with an equal volume of fresh leachant. The rate at which solution is pumped through the cell or the extent of leachant replenishment, which can be described as an effective flow, determines how the solution chemistry changes as the glass dissolves. Higher flow rates and more frequent and more complete replenishments maintain lower concentrations of all released glass components. Solutions contacting the glass in tests at high flow rates maintain the initial leachant conditions, while tests at low flow rates allow reaction products to accumulate in the solution. The effect of flow rate on glass corrosion is well illustrated by the results of tests performed using SRL 131 glass [206]. Figure 39 shows the measured mass loss of the sample as a function of reaction time for tests conducted using deionized water at various flow rates. Tests run using flow rates less than 1.0 mL/h show reaction rates that continually decrease with the reaction time, similar to static tests, while tests at higher flow rates show reaction rates that are linear in time and increase with the flow rate up to a rate of about 50 mL/h. Tests at 100 mL/h react at the same rate as tests 50 mL/h, which is the maximum rate under these temperature and pH conditions (see below). From this data, the forward rate is about 2.5 g/m²/day.

The glass corrosion rate can be related to the leachant flow rate by a simplified mass balance equation written in terms of the flux of silicic acid from the glass surface. Consider the reaction of a glass in a well-mixed reactor having a cell volume V with a solution that passes through the cell at a flow rate ϕ . The change in the silicic acid concentration in the cell over time depends on the amount added to the cell as it is released from the glass and the amount removed from the cell due to solution flow. These amounts are given by Eq. 18

$$\frac{d[\text{H}_4\text{SiO}_4]}{dt} = \frac{\text{SA}}{V} k_+ \left(1 - \frac{[\text{H}_4\text{SiO}_4]}{[\text{H}_4\text{SiO}_4]_{\text{sat}}} \right) - \frac{\phi [\text{H}_4\text{SiO}_4]}{V} \quad (18)$$

where the first term on the right-hand side represents the release of silicic acid from the glass (from Eq. 14), and the second term represents the removal of silicic acid from the reaction cell via flow. An additional term is needed if the leachant contains silicic acid. Equation 18 will not apply under very low flow conditions, in which mass transport through the solution will become important. Under constant flow conditions, a steady state may be attained, in which the silicic acid concentration within the cell remains constant. The steady-state silicic acid concentration will depend on the flow rate, and can be determined from Eq. 18 by setting $d[\text{H}_4\text{SiO}_4]/dt = 0$ and solving for $[\text{H}_4\text{SiO}_4]$. In this context, "steady state" refers to the condition where the rate at which silicic acid is released from the glass is equal to the rate at which it is removed from the reaction cell. The expression for the steady-state silicic acid concentration, which is represented by the term $[\text{H}_4\text{SiO}_4]_{\text{ss}}$, can then be substituted into Eq. 13 to obtain

Fig. 39. Mass Loss vs. Corrosion Time for SRL 131 Glass at 90°C at Various Flow Rates: (a) (Δ) Static, ($\bullet$) 0.1 mL/h, ($\square$) 0.5 mL/h, ($\circ$) 1.0 mL/h; and (b) ($\diamond$) 10 mL/h, ($\blacklozenge$) 25 mL/h, (+) 50 mL/h, (x) 100 mL/h (after [206])

$$\frac{d n[\text{H}_4\text{SiO}_4]_{ss}}{dt} = \frac{SA k_+ \phi}{\left(\frac{SA}{[\text{H}_4\text{SiO}_4]_{sat}} \right) k_+ + \phi} \quad (19)$$

where $n(\text{H}_4\text{SiO}_4)_{ss}$ is the number of moles of silicic acid released from the glass at steady state. Under sufficiently high flow rate conditions such that $\phi \gg (SA/[\text{H}_4\text{SiO}_4]_{sat}) k_+$, the glass corrosion rate becomes

$$\frac{d n[\text{H}_4\text{SiO}_4]}{dt} = SA k_+ \quad (20)$$

and is independent of the flow rate. Under low flow rate conditions, such that $\phi \ll (S/[\text{H}_4\text{SiO}_4]_{sat}) k_+$, the glass corrosion rate becomes

$$\frac{d n[\text{H}_4\text{SiO}_4]}{dt} = [\text{H}_4\text{SiO}_4]_{sat} \phi \quad (21)$$

and is proportional to the flow rate. At very low flow rates, the glass corrodes faster than the flow rate, a steady-state concentration is not maintained, and Eq. 18 becomes equivalent to Eq. 14. It is important to note that it is not the actual flow rate that determines whether a system is in the low or high flow limit, but the difference between the solution flow and the intrinsic corrosion rate. This means that the response of a glass to changes in the solution flow will depend on its composition.

The results obtained by Adiga et al. [206] are replotted in Fig. 40 as the glass reaction rate (i.e., the slopes of the lines in Figs. 39a and 39b) against the leachant flow rate. This plot shows the behavior predicted by Eqs. 20 and 21: at high flow rates the glass corrodes at a constant rate, and at lower flow rates it corrodes at a rate that depends on the flow rate. The constant rate achieved at high flow rates is characteristic of the glass composition, the leachant composition, and the reaction temperature. From Eq. 20, the maximum rate is proportional to the rate coefficient for glass dissolution. From further analysis of these test results, Apted and Adiga concluded that all flow rates tested were high relative to the glass dissolution rate and that the test conditions did not simulate expected repository conditions [142]. They also pointed out that the crossover from transport control to surface reaction control occurs at flows higher than expected in a repository and that radionuclide release rates would be overestimated at high flows. Finally, Apted and Adiga noted that the thickness of a static layer of solution at the glass surface is expected to decrease at higher flow rates. Because this layer provides a diffusion barrier to the transport of released glass components away from the glass surface, they suggested that a decrease in the layer thickness is expected to result in and increase the glass dissolution rate. The effect of such a static fluid layer on the glass corrosion rate is analogous to the effect of the flow rate: thicker barriers would lead to increased concentrations of glass corrosion products at the glass surface and are expected to have the same effect as lower flow rate, namely they are expected to slow the corrosion.

The high flow rates (relative to glass corrosion rates) attained in continuous flow apparatuses are not relevant to any anticipated repository environments, which are usually engineered to prevent water from contacting waste packages. Difficulties associated with attaining low flow rates with liquid pumps in laboratory tests have led to the use of partial-replenishment tests (sometimes called pulsed-flow tests), in which some or all of the leachate is periodically removed from the reaction vessel and replaced with fresh leachant. Of course, periodic replenishment only becomes strictly equivalent to continuous flow in the limit where the

Fig. 40. Results Obtained by Adiga et al. Plotted as Glass Corrosion Rate vs. Flow (after [164])

replacement period becomes infinitesimal. The frequency and extent of replacement can be related to an effective flow rate through the number of exchanged volumes in the reaction cell per year [207,208]. (The fact that an effective flow can be defined for periodic replenishment does not address the issue of whether or not the glass corrodes in the same manner in flow tests and in periodic replenishment tests.) If the volume of the reaction cell is V , and the amount of leachate removed and replaced with an equal amount of fresh leachant is ΔV , and the time between replacements is Δt , then a residence time can be defined as $T_r = \Delta t(\Delta V/V)$. The residence time is the time required for complete replacement of the vessel volume. For example, if one-fourth of the volume is replaced every 28 days, then the entire volume will be replaced after four exchanges and a residence time of 112 days. Tests have been conducted to determine the dependence of the extent of corrosion on Δt and $\Delta V/V$ at a constant value of T_r [127]. The results, which are shown in Fig. 41, show that larger replacement volumes and longer replacement times lead to greater corrosion up to about 168 days. The scatter in the data beyond 168 days is similar to the differences measured at shorter times. Barkatt et al. interpreted these results as demonstrating that the corrosion rate depends on T_r after long reaction times, but not on the exchange period Δt or the exchange fraction $\Delta V/V$ independently [127]. However, the scatter in the data weakens this conclusion. The assumed equivalence of common products of T_r from different Δt and ΔV is tantamount to assuming the equivalence of common products of $(SA/V)t$, an assumption that was demonstrated earlier to fail in most cases because it neglects a pH-dependence of the glass corrosion. In these tests, t is replaced with T_r and V with ΔV . The same arguments against using $(SA/V)t$ as a scaling factor apply to $(SA/\Delta V)T_r$ as a scaling factor. In practice, the effects of pH may be inconsequential at near-neutral values and under dilute conditions, as seen in the results of Pederson et al. [72]. It may be expected that the effects of variations in the exchange volume fraction and time that are observed in partial replenishment tests will depend on the SA/V of the test system and will become more important as the SA/V increases. The test results shown in Fig. 7 were obtained at an SA/V of about 970 m^{-1} . The pH

Fig. 41. Measured Boron Concentration in Partial Replenishment Tests Conducted at Constant $T_r = 112$ Days: (●) $\Delta t = 28$ days, $\Delta V/V = 0.25$; (■) $\Delta t = 7$ days, $\Delta V/V = 0.0625$; (▲) $\Delta t = 3.5$ days, $\Delta V/V = 0.0312$ (after [127])

values were not reported, although the measured differences in the boron concentrations as a function of ΔV suggest the pH was greater than 9. It should be pointed out that the volume of leachate that reacts with the sample is different than the volume of leachate in the test cell at any time. This distinction is not accounted for in the treatment of the data. Likewise, the dilution of the leachate that occurs upon every replenishment cycle is not accounted for. This change may be important when the solution is near saturation.

The cycling of the solution between dilute and near saturation conditions may affect the alteration of the glass surface. While the alteration layers formed on nuclear waste glasses do not significantly retard the glass corrosion, they do interact with the solution and may affect the solution chemistry, which does affect the glass corrosion rate. The impact of cycling the solution chemistry may affect the evolution of the surface alteration phases and continued glass corrosion rate.

Despite questions of the equivalence of different replacement volumes and periods, the results of partial-replenishment tests can be described by the effects of flow predicted in Eqs. 20 and 21. Figure 42 shows the results of partial-replenishment tests with SRL 131 glass conducted in deionized water at 90°C [124]. These tests show the corrosion rate to increase with the flow until a maximum rate of about $2.5 \text{ g/m}^2/\text{day}$ occurs at sufficiently high flow rates. This maximum rate is the same as that found in continuous flow tests conducted with SRL 131 glass [206]. Appropriate selection of exchange volumes and periods is seen to mimic the expected solution behavior at very low effective flow rates not attainable in continuous flow apparatuses. The results show the corrosion rate to continually decrease as the flow decreases.

Fig. 42. Boron Release from SRL 131 Glass as a Function of Flow Rate as Simulated by Periodic Replenishment of the Solution (after [124])

While the expected trend toward Stage II of corrosion (apparent saturation) is observed in these tests, uncertainties remain regarding the effects of the cycling of the leachate between saturated and diluted conditions on the corrosion behavior. That is, while the average solution volume over the test period can be related to that of a test conducted at a low flow rate the sample has been subjected to very different solution chemistries than those of a low flow rate test. Probably as important as comparing the results of periodic replacement tests at high effective flow rates to tests using a constant flow are their comparison at low effective flow rates to static tests. The effects of varying the solution chemistry through periodic replenishments are more significant under these corrosion conditions, where the periodic dilution hinders the nucleation of secondary phases from the saturated solution by periodically diluting the solution and lowering the pH. These effects prevent the natural evolution of the corrosion process and result in a misrepresentation of the long-term corrosion rate. That is, the corrosion rate prior to the formation of secondary phases does not represent the long-term corrosion rate.

The complexities of periodic dilution of the solution have also prevented the use of test data in corrosion models. The primary usefulness of the test results appears to be verification of the effects of the flow rate and the solution chemistry on glass corrosion behavior at intermediate flow rates. While the forward rate determined at high effective flows is consistent with that measured using a constant flow device, the test is not well-suited for measuring the forward rate. In the limit of replenishment schedules the test reduces to an MCC-1 or PCT type test, depending on if monolithic or crushed glass is used. Replenishment of the solution would not benefit the measurement of the forward rate. The forward rate is better measured using single-pass flow through or Soxhlet tests.

Although static tests may most faithfully simulate repository environments where groundwater flow will be extremely low, tests performed at high leachant flows provide conditions in which waste forms corrode free from interactions with corrosion products. These tests allow much-needed characterization of the effects of various solution species and the solution pH on the corrosion process. A knowledge of the forward rate coefficient and how it depends on the solution chemistry is of key importance to defining the corrosion behavior of waste glasses and predicting their long-term behavior. Tests in flowing leachants used to determine the effects of solution chemistry on glass corrosion have been discussed previously (see Sections 3.2-3.4).

6.0 DETERMINATION OF MODEL PARAMETERS FROM LABORATORY TESTS

The observed effects of the solution chemistry, the results of static tests conducted at different SA/V, and the flow rates used in dynamic tests on glass corrosion have lead to a mechanistic description of the processes that result in glass corrosion. By utilizing different tests and different experimental conditions, various reactions have been highlighted and characterized. Analytical models based on descriptions of these processes can be used to predict corrosion behavior of high-level waste glasses over a wide range of environmental conditions and over long time periods. Models are needed because it is not possible to measure experimentally the durabilities of high-level waste glasses for the many tens of thousands of years that they must control the release of radionuclides or under the range of environments that may exist in different disposal systems over time.

The corrosion of most high-level waste glasses can be described as occurring in three stages. These stages correspond to limiting conditions in the analytical description of the glass corrosion rate given in Eq. 14.

$$\frac{d [H_4SiO_4]}{dt} = \left(\frac{SA}{V} \right) k_+ \left(1 - \frac{[H_4SiO_4]}{[H_4SiO_4]_{sat}} \right) \quad (14)$$

The first stage occurs when the solution concentrations of glass components (primarily silicic acid for borosilicate waste glasses) are so low that they do not affect the corrosion reactions. The glass corrodes at its forward rate under these conditions, which depends only on the solution pH and the temperature. Experiments have shown that alkali metals and boron are preferentially leached from the glass under these conditions and that the solution pH increases. The increased pH results in a higher corrosion rate. The second stage occurs as the solution concentration of silicic acid increases and begins to affect the glass corrosion rate. The increased silicic acid concentration slows the glass corrosion rate through the affinity term $(1 - [H_4SiO_4]/[H_4SiO_4]_{sat})$. The rate continually slows during the second stage and approaches an apparent minimum rate as the affinity term approaches zero. The minimum rate depends on the glass composition, temperature, and the solution pH. The third stage of corrosion occurs when secondary phases precipitate from the leachate solution to establish solubility limits for important glass components (primarily silicic acid for borosilicate waste glasses). Changes in the solution chemistry due to secondary phase formation then affect the glass corrosion rate through the affinity term as $[H_4SiO_4]$ is maintained at a value less than $[H_4SiO_4]_{sat}$ by the secondary phases. In some systems, materials that are present throughout the test may affect the solution chemistry in such a way that the minimum rate is never reached. This situation occurs in the presence of clays that may be present in backfill and ductile iron, which consume silicon from the solution as it is released from the glass.

Given this description of the solution control of the corrosion rate, corrosion models must account for the intrinsic corrosion rate in highly dilute solutions, the effects of the solution pH, the decrease in the corrosion rate as the silicic acid solution concentration increases, and the effects of secondary phases and other materials on the corrosion rate. Most current models of waste glass corrosion are based on variations of the model developed by Grambow [153,154] (see Section 3). Implementation of these models requires the determination of several model parameters through the regression of experimental data. It is important to understand how information obtained from the experiments or tests performed relate to the corrosion mechanism and to the corrosion model to ensure that the data are being used correctly.

A simple form of the rate equation is used in current simulations of glass corrosion; it can be written in a form similar to that of Eq. 14:

$$\frac{d[B]}{dt} = (SA/V)k_+ \left(1 - \frac{[H_4SiO_4]}{[H_4SiO_4]_{sat}} \right) + R_{\infty} \quad (22)$$

where the rate is expressed in terms of the solution concentration of boron rather than silicic acid and the term R_{∞} has been added to account for the nonzero reaction rate under "saturated" conditions. The rate is expressed in terms of the measured solution concentration of boron rather than the measured solution concentration of silicon because silicon is commonly incorporated into secondary phases so that the solution concentration of silicon often does not represent the extent of glass corrosion (see also Section 2.2). The affinity term is nevertheless determined by the solution concentration of silicon, regardless of whether or not secondary phases form. Note that this is the rate of change of the solution concentration, and the rate of glass corrosion is given by this rate divided by the surface area. If other phases are present in the system or if secondary phases form as the glass corrodes that limit the silicic acid concentration in solution such that the affinity term remains nonzero, then the rate will likely be determined by the first term on the right side. Otherwise, the first term will approach zero as the solution becomes saturated with respect to silicon, and the long-term rate will be given by R_{∞} .

To solve Eq. 22 as the glass corrodes, one must know k_+ , $[H_4SiO_4]_{sat}$, and R_{∞} . The values of these parameters are determined experimentally either by regression to the test data or by conducting separate experiments designed to highlight and measure the values of specific parameters. Separate tests are required to determine the parametric values for different glass compositions, and under different temperature and pH conditions. The pH dependence of the rate coefficient can be written as $k_+ = k_f [OH^-]^h$, where k_f and h must be determined in separate tests (see Section 3.3). The term R_{∞} is an ad hoc corrective value to account for the difference between the long-term rate predicted in Eq. 22 and experimental observations, and is defined so as to improve the fit of the equation to specific long-term data. Tests at high SA/V are used to characterize R_{∞} [167]. The values of model parameters determined by using experimental results include both measurement errors and errors in the model. Thus, the value determined for R_{∞} includes error in the experimental measurement of the corrosion rate and error in the definition of R_{∞} and expressing the rate in terms of Eq. 22. The values of $[H_4SiO_4]_{sat}$ and R_{∞} are usually found to depend on the test conditions and the pH.

Two limiting cases with regard to Eq. 22 can be defined: highly dilute solutions for which the affinity is near one (i.e., where $[H_4SiO_4]/[H_4SiO_4]_{sat}$ is near zero), and highly concentrated or "saturated" solutions, for which the affinity is near zero (i.e., where $[H_4SiO_4]/[H_4SiO_4]_{sat}$ is near one). Under the limit where $[H_4SiO_4]$ approaches zero, the rate equation can be written as

$$\frac{d[B]}{dt} = (SA/V)k_+ + R_{\infty} \quad (23)$$

Dynamic flow-through tests provide an excellent measure of the forward reaction rate. The use of these tests to measure the corrosion rates under various leachant chemistries has been discussed previously (Sections 3.2 and 3.3). Tests performed at very low values of $(SA/V)t$, which maintain dilute solutions, provide another measure of the forward reaction rate. These tests approach a maximum rate as $(SA/V)t$ decreases, which represents the forward reaction rate. (The value of R_{∞} is generally several orders of magnitude less than k_+ and can be ignored under the test conditions represented by Eq. 23.)

Under the limit where $[H_4SiO_4]$ approaches $[H_4SiO_4]_{sat}$, the rate equation can be written as

$$\frac{d[B]}{dt} = R_{\infty} \quad (24)$$

Solutions can be saturated by doping the leachant with silicic acid or by conducting experiments under conditions in which the solution rapidly approaches "saturation" conditions. The latter method will generate solutions with high concentrations of other glass components that may also affect the corrosion rate. The increase in solution concentrations can be accelerated by conducting tests at high SA/V ratios, although higher solution pH values will also occur in tests at high SA/V that will also accelerate the corrosion. The effects of the pH and the silicic acid concentration on the measured reaction rate cannot be separated based on static test data. Tests conducted in silicic acid-doped solutions likely will not provide a representative measure of the long-term corrosion rate because the secondary phases which control the long-term rate require other glass components be present in solution as well as silicic acid. Unnatural solution chemistries may result in secondary phase formation, but the resulting solution chemistries will be different than those that will result in dissolution of the glass and will affect the glass corrosion rates differently. That is, the silicic acid concentration established in the silicic acid-doped leachant will be different than that established in the leachate generated by dissolution of the glass. The different silicic acid concentrations will result in different long-term corrosion rates.

Reaction parameters can be determined from the results of static leach tests by plotting the corrosion rate against the silicic acid activity (or concentration). In such a plot, k_+ is the rate when the silicic acid activity is zero, and the saturation concentration and residual rate are estimated from the solution chemistry and corrosion rate measured at high silicic acid activities. These limits are illustrated in Fig. 43 [154,164]: the rate at a silicic acid activity of zero gives the forward rate, which is about 1.5 g/m²/day, and the constant rate at silicic acid activities greater than about 10⁻⁴ gives the residual rate, which is about 0.0017 g/m²/day. Scatter in test data leads to large uncertainties in the regressed values in this approach. The values determined by this method are probably not sufficiently reliable for use in performance assessment calculations, which are highly sensitive to the long-term corrosion rate [185].

Determination of the rate coefficient, k_+ , is straightforward from static leach tests conducted for short time periods, since these are likely to have the lowest silicic acid concentrations and so are likely to have affinity terms nearest to one. The rate coefficient can be obtained from static leach test data by combining the forward rate expression in Eq. 23 and the normalized mass loss function in Eq. 10 as

$$k_+ = \text{rate} = \frac{NL_i}{t} = \frac{c_i}{(SA/V) f_i t} \quad (25)$$

where t is the reaction time and the other variables have the same meaning as in Eq. 10. Tests should be conducted for several time periods to ensure that the solution remains dilute enough that the affinity term remains near one. Separate tests in pH-buffered solutions are required to ascertain the pH dependence of the rate coefficient k_+ .

The regression of residual rate from test data is well described by Curti [180], and the data and methodology are reproduced here. Data shown in Fig. 44 were obtained in static leach tests using a British glass referred to as MW and deionized water at 90°C and at an SA/V ratio of 1320 m⁻¹ [180]. The residual rate is determined from the measured release of highly soluble

Fig. 43. Corrosion Rate (as Measured by Boron Release) vs. Ion Activity Product of Silicon (after [154])

Fig. 44. Normalized Boron Mass Loss from MW Glass vs. Reaction Time (after [180])

glass components after silicic acid has become saturated. This is best approximated using tests conducted for the longest reaction times. Curti used test data from tests conducted longer than 28 days to determine the residual rate for corrosion of MW glass, as shown in Fig. 44. The slope of the fitted line gives the residual rate, which is reported to be 0.006 g/m²/day from Fig. 44. However, as reported by Curti, this value represents the mean value while the data include a range of between 0.0014 and 0.0105 g/m²/day at the 95% confidence level. Clearly, a much lower residual rate would be obtained by considering only data from tests conducted for 91 days or longer (the three longest time periods tested). In fact, the residual rate according to these data is essentially zero. This variation demonstrates the high degree of subjectivity in this approach. Again, the wide range of rates is probably not acceptable for conducting performance assessments. The silicic acid saturation concentration that is needed in the rate equation is given by the measured silicic acid concentration in the tests used to determine the residual rate, corrected for dissociation via

$$[\text{H}_4\text{SiO}_4]^\circ = \frac{[\text{Si}]}{(1 + 10^{\text{pH}-\text{pK1}} + 10^{2\text{pH}-\text{pK2}} + 10^{3\text{pH}-\text{pK3}})} \quad (25)$$

where $[\text{H}_4\text{SiO}_4]^\circ$ is the concentration of undissociated silicic acid, $[\text{Si}]$ is the measured amount of silica in the solution, pH is the measured pH corrected to the test temperature, and pK1, pK2, and pK3 are the dissociation constants for silicic acid at the test temperature.

For conditions under which the glass corrodes at a constant rate, Eq. 22 predicts that a plot of $\log[B]$ versus $\log\{(SA/V)t\}$ will be a line with a slope equal to one and a y-intercept that is proportional to the rate [67,209,210]. The glass is expected to react at a constant rate in the limits (1) when the silicic acid concentration is near zero, where the y-intercept corresponds to the forward rate, and (2) when silicic acid has attained its saturation concentration, where the y-intercept corresponds to the residual rate. Experimental data are expected to converge to these lines at both low and high values of $(SA/V)t$. Note that the values of k_+ and R_∞ determined from the data will both depend on the solution pH of the tests under the limiting conditions of low and high $(SA/V)t$, respectively. The solution pH is expected to be different in the two limits for most alkali borosilicate waste glasses. Hence, the differences in the rates will be due to both the different solution concentrations of silicic acid and the different pH values at the two limits.

The results of MCC-1 (at 10 m⁻¹) and MCC-3 (at 2000 m⁻¹) tests with four glasses produced during development of a waste glass for West Valley Demonstration Project and the DWPF glass SRL 165 were plotted as $\log[B]$ vs $\log[(SA/V)t]$ by Strachan [211]. Strachan actually used $(SA/V)t/10$ for the x-axis so that the data were plotted as "equivalent to 10 m⁻¹". The plot is reproduced in Fig. 45. The forward rate coefficient k_+ determined from limiting slope at small equivalent time, and the "saturated rate," R_∞ (represented as k_+ in [211]) determined from the limiting slope at large equivalent time are included in the plot. The forward rates of the five different glasses are the same, within a factor of two, while the "saturated rates" differ by more than a factor of 15. Strachan pointed out that the "saturation rates" of all glasses were higher than those needed to conform to the maximum allowable release according to criterion of the Nuclear Regulatory Commission 10 CFR 60.

Note that Strachan ignored the data point for WV205 glass at the longest equivalent time tested (3920 d equivalent), which had a significantly higher boron concentration than the previous test. While tests at longer equivalent times are needed to verify the long-term behavior of WV205 glass, tests with SRL 202 glass have shown an identical sudden increase at long

Fig. 45. Log of Normalized Boron Mass Loss vs. $\log(SA/V)t$ for Tests with (○) WV205A.01, (Δ) ECM85-13, (◇) WV205, and (●) SRL 165 Glasses (after [211])

reaction progress (see below). Long-term tests with West Valley Reference glass 6, which is compositionally similar to WV205, are currently in progress at ANL. These tests will clarify the long-term behavior of that glass.

Application of the log-log plot to determine the forward and "saturation" rates is demonstrated using the data presented in Fig. 46, where lines with slopes of one have been drawn through data for SRL 202A glass reacted at $20,000 \text{ m}^{-1}$ [67]. The line with a slope of one drawn through the data point from the test run for the shortest time, which was three days, yields a y-intercept of $\log[B] = -3$. Note that these tests are expected to react at a rate significantly lower than the forward rate because of the very high SA/V used. Tests at very low SA/V are appropriate for measuring the forward rate. These data are used only to demonstrate application of the technique. About $1 \times 10^{-3} \text{ g}$ of boron per cubic meter of solution (units of $[B]$ are mg/L) has been released after a reaction progress of $(SA/V)t = 1 \text{ day/m}$. The rate calculated by this method is the integrated rate, that is, the average rate over the entire time tested to that point. The average release rate over the first three days is therefore $1 \times 10^{-3} \text{ g}_{\text{boron}}/\text{m}^2/\text{day}$. Since the weight fraction of boron in SRL 202 glass is $0.025 \text{ g}_{\text{glass}}/\text{g}_{\text{glass}}$, the rate of glass corrosion given in terms of the mass of glass dissolved is about $0.04 \text{ g}_{\text{glass}}/\text{m}^2/\text{day}$. Note that this value is the average rate over three days and that the solution pH has increased from about 8.2 to about 10.5 over this period. Similar calculations using data obtained in static tests run at 2000 m^{-1} gives a forward rate of $0.025 \text{ g}_{\text{glass}}/\text{m}^2/\text{day}$ on the basis of data for 3 days of reaction. The lower rate measured in tests at 2000 m^{-1} probably results from the lower pH attained in these tests. The rate is known to be very sensitive to the pH [70]. Measurement of the solution pH value is usually reliable to within about 0.02 units at room temperature with aqueous solutions, though measurement of the pH values of solutions having high ionic strength is less reliable. The pH

values measured at room temperature can be used to estimate the pH values at the test temperature using solution equilibration codes. The pH values at 90°C are typically about 1 pH unit lower than those measured at room temperature due to the differences in the dissociation constant of water.

The minimum rate attained in the experiments with SRL 202 glass at 20,000 m⁻¹ occurred after about 182 days (at $\log[(SA/V)t] = 6.56$). The line of slope one drawn through this point gave a rate of 0.0025 g_{glass}/m²/day. This is 16 times lower than the rate measured after 3 days of reaction. The observed decrease in the rate between 3 and 180 days is due to the approach of the solution to saturation, although the higher solution pH after 180 days of reaction compared to the pH attained after 3 days of reaction (about 11.2 compared to about 10.5) probably accelerated the reaction. It is important to remember that the solution conditions change continuously with the reaction time prior to secondary phase formation, and that the evolution of the solution chemistry is different for tests conducted at different SA/V. This is clearly demonstrated by the difference in boron concentrations in tests conducted at 2000 and 20,000 m⁻¹ at the same values of (SA/V)t.

Tests at 20,000 m⁻¹ run longer than 182 days yield reaction rates that are higher than that measured after 182 days. The increase is due in part to the higher pH values (about 12) that are reached in tests run longer than 182 days. The increase in pH is expected to increase the value of k_+ and accelerate the reaction. The cause of the pH increase is uncertain; it may be due to the formation of secondary phases or a resurgence of ion exchange as the glass corrosion rate increases. The observed increase in the corrosion rate indicates that the "saturation rate" does not represent the long-term corrosion rate for this glass under these conditions and that the long-term rate may be significantly higher than the "saturation rate." Notice that the lowest rate observed in tests performed at 2000 m⁻¹ can be seen in Fig. 46 to be lower than the lowest rate

Fig. 46. Release of Boron from SRL 202A Glass as $\log[B]$ vs. $(SA/V)t$ for Tests at SA/V: (●) 10, (■) 2000, and (◆) 20,000 m⁻¹. Lines having a slope of one are drawn through data showing the maximum and minimum corrosion rates in tests at 20,000 m⁻¹ (after [67]).

attained in tests performed at $20,000 \text{ m}^{-1}$. (Data points for tests at 2000 m^{-1} conducted for large $(\text{SA/V})t$ lie to the right of the line of slope one drawn through the data point showing the lowest rate in tests at $20,000 \text{ m}^{-1}$.) Because of the different pH values in tests at 2000 and $20,000 \text{ m}^{-1}$, the saturation concentrations will differ in tests conducted at different SA/V. Differences in the solution pH affect both the forward rate and the solution saturation conditions.

A similar increase in the corrosion rate at long reaction times has been observed in other tests upon formation of clinoptilolite [200,201]. The acceleration can be interpreted based on the effects of clinoptilolite (and possibly other secondary phases) on the dissolution affinity and the effects of the solution pH on the precipitation of secondary phases. That is, secondary phase formation controls the glass corrosion rate and the solution pH controls the formation of the secondary phases.

The long-term behavior measured in tests conducted at different SA/V and in tests with radioactive and nonradioactive glasses is very different. The corrosion rate in tests conducted at 2000 m^{-1} are seen to continually decrease as corrosion proceeds, while the rates in tests conducted at $20,000 \text{ m}^{-1}$ decrease initially then increase. The difference in behavior can be attributed to the different solution chemistries attained under the different test conditions: higher pH values and higher solution concentrations are attained in tests conducted at higher SA/V, and the higher pH values support precipitation of secondary phases. Faced with two very different long-term corrosion rates from tests at different SA/V, one must relate the anticipated disposal conditions to the test conditions to determine which rate is relevant. In most cases, the tests at higher SA/V are more relevant to conditions where the amount of water contacting the glass is minimized.

The value of $[\text{H}_4\text{SiO}_4]_{\text{sat}}$ can also be determined from experimental data when the minimum rate occurs. Values are usually found to vary slightly for different glass and leachant compositions, but they are typically on the order of $1 \times 10^{-3} \text{ M}$ and are intermediate between the solubilities of quartz and amorphous silica [161,166,167,205,212,213].

Reaction rates determined by using the methods described above are presented in Table 5 for several glass compositions. The forward reaction rates are generally between 1 and $2 \text{ g}_{\text{glass}}/\text{m}^2/\text{day}$ at 90°C , although some measurements yield much lower rates. The forward rates measured under dynamic conditions are probably more accurate than those measured under static conditions because of the build up of reaction products in the leachate affects the results of static tests. The build up of reaction products in static tests and in dynamic tests at low flow rates severely reduces the reaction rate, such that the average rates measured over even short time periods represent lower limits of the forward rates.

The rates measured under "saturated" conditions are typically two to three orders of magnitude smaller than the forward reaction rates. However, "saturated" rates measured for the same glass reacted in different leachants or at different SA/V ratios may indicate that a unique "saturation" rate may not exist for different glass compositions [167] or that the solution has not become "saturated" in the tests, perhaps because secondary phases have formed. In addition, the corrosion rate may increase as more stable secondary phases form over very long reaction times [64,200,201]. For example, the instantaneous long-term rates of SRL 202 and SRL 131 glasses in tests at $20,000 \text{ m}^{-1}$ are about 0.03 and $0.04 \text{ g}/\text{m}^2/\text{day}$ after secondary phases form [67].

Table 5. Experimentally Determined Reaction Rates at 90°C, in g glass/m²/day

Glass/Leachant	SA/V(m ⁻¹)	Forward Rate	Saturation Rate	Reference
Static Tests				
PNL 76-69/DIW	2000	1.6	0.08 ^a	209
SRL 165/DIW	2000	0.80	0.024 ^a	211
EMS-11/DIW	2000	0.083	0.0016 ^a	211
SRL 165/DIW		1.7	0.016 ^a	211
WV205		3.4	0.035 ^a	211
JSS-A/DIW	10 ^b	1.5	0.0025	165
PNL 76-68/DIW	10 ^b	1.8	0.0075	165
SRL 131/DIW	10 ^b	3.0	0.033	165
SRL 131/J-13 ^c	10	0.14		67
SRL 131/J-13	2000	0.27	0.021	67
SRL 131/J-13	20000	0.84	0.053	67
SRL 202/J-13	10	0.099		67
SRL 202/J-13	2000	0.025	0.0016	67
SRL 202/J-13	20000	0.040	0.0025	67
R7T7/DIW	5	4.9 (100°C)		166
R7T7/DIW	50		0.0083	167
R7T7/Volvic ^d	50		0.0133	167
R7T7/DIW	400		0.0045	167
R7T7/Volvic	400		0.025	167
R7T7/Volvic	2000		0.0006	167
R7T7/Volvic	8000		0.0006	167
R7T7/Volvic	20000		<0.0001	167
MW/DIW	1320	1.1	0.01	180
MW/DIW	10	1.2	0.009	164
MW/DIW	10	1.1		214
Dynamic Tests				
SRL 202/pH 7		0.28 (80°C)		215
SRL 165 ^e /pH 10.5		0.05 ^a		70
SRL 165 ^e /pH 10		0.08 (70°C)		112
R7T7/DIW		1.03		69
SRL 131/DIW		2.5		206

^aEstimated.^bValues determined from results of both static and dynamic tests.^cTuff groundwater. Major components are 45 ppm Si, 55 ppm Na, and 120 ppm HCO₃⁻.^dGranite groundwater. Major components are 11 ppm Si, 9.8 ppm Ca, 9.2 ppm Na, and 66 ppm HCO₃⁻.^eAnalog glass without iron.

7.0 EFFECTS OF SA/V ON RADIONUCLIDE DISPOSITION

The vast majority of laboratory tests have focused on the corrosion behavior of the glass matrix rather than the release of radionuclides because the glass corrosion rate is usually assumed to provide an upper limit of radionuclide release, because of difficulties associated with handling radioactive materials, and difficulties in monitoring radionuclides present in low concentrations. While the chemical behavior of radionuclides in well-constrained environments is known, very few tests have been conducted to characterize their behavior in the presence of glass corrosion products. The few results available regarding the chemistries of actinides and long-lived fission products have not yet been incorporated into glass corrosion models. While the intrinsic resistance of borosilicate waste glasses towards corrosion is important in stabilizing the radionuclides and establishes a bounding upper limit on their release rates, several factors affect the subsequent mobilization and transport of radionuclides away from the waste form. These include the solubilities of the radionuclides and their complexes, colloid formation, and sorption onto other phases.

Knowledge of the behavior of radioactive waste components under various conditions over long time periods will allow realistic source terms representing the release of specific radionuclides to be incorporated into performance assessment calculations. While the solubilities of many radionuclides are known to be very low in neutral and alkaline groundwaters, laboratory tests are required to determine the effects of complexation, sorption, and colloid and secondary phase formation on the behavior of radioelements in a geologic environment.

All radionuclides are stabilized when incorporated into a glass matrix because their mobilization can only occur as the silicate network of the glass corrodes, and so their release from the waste form is limited by the glass corrosion rate. Actinides exist in borosilicate high-level waste glass as U(VI), Np(IV), Pu(IV), and Am(III) [2], where the oxidation states are controlled primarily by redox couples within the melt. Technetium has been found to be present as Tc(IV) in borosilicate glass produced under oxidizing conditions [216]. (Both Tc(IV) and metallic Tc was found in glasses produced under reducing conditions.) The oxidation states of these radionuclides in the leachate will be determined by other redox couples in the surrounding geology and will be more strongly affected by the atmosphere above the solution [217]. For example, the dominant oxidation states in tuff groundwater under slightly oxidizing conditions have been determined to be Np(V), Pu(V and VI), and Am(III) [218,219]. Uranium will be present as U(VI) and technetium as Tc(VII) under these conditions. The oxidation states in the solution will determine the disposition of the radionuclides.

While the intrinsic resistance of borosilicate waste glasses towards corrosion is important in stabilizing the radionuclides and establishes a bounding upper limit on their release rates, several factors affect the subsequent mobilization and transport of radionuclides away from the waste form. These include the solubilities of the radionuclides and their complexes, colloid formation, and sorption onto other phases. The solubility of a particular element is given by the sum of the concentrations of all species of that element which exist in solution, including free ions, hydrolyzed species, and complexes. Solubility products have been measured for many actinide-bearing solids, although the phases which control actinide solubilities during glass corrosion are often difficult to identify. Maximum concentrations of complexed radionuclides are established by their individual equilibria. In the absence of complexation, the solubilities of most actinides are generally quite low. Complexation generally leads to increased actinide solubilities, but may also result in decreased solubilities, depending on the solubility controlling phase. Since the extent of hydrolysis and the concentrations of several ligands depend on the pH, the solubility of radioelements will also be pH-dependent. Complexation by ligands which are the conjugate bases

of weak acids is not significant at pH values below the pK_a values of those acids. For example, carbonate is expected to be present in appreciable quantities only at pH values greater than about 10, bicarbonate above about pH 6, biphosphate above about pH 7, etc. The solubility may be affected by changes in the ionic strength, composition, and temperature of the solution [220,221]. Most laboratory investigations of actinide and technetium hydrolysis, complexation, and solubility have been performed at or near room temperature. Radionuclide behavior at elevated temperatures must currently be extrapolated from room temperature data or estimated from the behavior of nonradioactive homologues. Much thermodynamic information is still needed before the solubilities and complexation of transuranium elements can be utilized to reliably model long-term geologic interactions [222,223].

Technetium and the actinide elements may exist in solution in different oxidation states depending on the chemistry of the contacting solution. The effect of the redox potential of the solution (i.e., whether the groundwater is oxidizing or reducing) on the preferred oxidation state(s) of an individual actinide is related to the reduction potential of that actinide, which depends on the temperature and ionic strength of the solution. The few studies conducted at different temperatures show that the solubilities are sometimes affected by temperature changes [e.g., 224] and sometimes not affected [e.g., 218,219]. Certainly, more experimental work is needed to properly account for temperature changes in performance assessment of potential repositories.

The chemistry of natural groundwaters and aquatic systems that may eventually contact high-level waste glasses can vary significantly depending on the surrounding environment. The redox potentials of various groundwaters are established by the presence of redox pairs, such as Fe^{3+}/Fe^{2+} and Mn^{4+}/Mn^{2+} , and by dissolved oxygen. Measured potentials of shallow groundwaters, mine waters, and fresh waters are generally found to range between about -200 and 600 mV [141]. The maximum Eh of oxic (oxidizing) groundwaters may be limited by the oxygen-hydrogen peroxide equilibrium [141,225], while the lower limit of anoxic (reducing) groundwaters may be established by the hematite-magnetite equilibrium [225,226]. The actual Eh of the groundwater will reflect its interaction with the surrounding mineralogy, primarily with iron-bearing phases and carbonate minerals. The partial pressure of carbon dioxide in equilibrium with the groundwater also affects the pH and Eh. Typical CO_2 partial pressures in contact with groundwaters may range from 0.03% up to 10%, and pH values typically vary between about 4 and 9 [141,217]. Acidic groundwaters typically have higher Eh values than basic groundwaters.

The chemistry of the fluids contacting the waste glass in the near-field environment will differ from the initial groundwater due to corrosion of waste glass and other materials in the engineered barrier system and radiolysis of the air and groundwater [212,227]. The pH of the fluid contacting the waste glass is expected to increase as the glass corrodes due to the release of alkali (although the pH of some brine solutions have been found to decrease during corrosion due to secondary phase formation [179]), but decrease due to the generation of nitric and nitrous acids through the radiolysis of moist air. The pH of the fluid contacting the waste glass will depend on the contributions of several simultaneous interactions. Likewise, the redox potential of the solution will be affected by the release of redox-sensitive elements such as iron and the radioelements themselves from the glass and other materials during corrosion, and radiolysis of the air and groundwater. Therefore, the stabilities of actinide species in the near-field environment must be considered under a range of conditions much wider than those defined by potential groundwaters to interpret laboratory experiments and to model the long-term repository behavior in computer simulations.

While the concept of thermodynamically controlled radionuclide solubilities is useful in discussing actinide release, other interactions may significantly affect the distribution of radionuclides as the glass corrodes. Radionuclides may also sorb onto colloids occurring naturally in the groundwater or colloids generated during glass corrosion. These colloids may

remain mobile and lead to solution concentrations that far exceed the thermodynamic solubility limitations, or they may flocculate and settle out of solution. In addition, radionuclides that are released from the glass into the groundwater may become incorporated in secondary phases that are generated as the glass corrodes, including altered layers at the glass surface and precipitated mineral phases, or they may sorb onto these phases or other materials in the vicinity of the waste form.

The potential distribution of radionuclides between mobile and immobile phases in the immediate vicinity of the corroding waste form is discussed below. Because the solubility and sorptive properties of the radioelements are strongly dependent on their oxidation states and complexation, the chemistry of technetium, uranium, neptunium, plutonium, and americium is first summarized to assist in the interpretation of various test results. Experimental observations of the interactions of these radionuclides with the altered glass layers, secondary mineral phases, and colloids near the surface of the corroding glass are then discussed.

Uranium, neptunium, plutonium, and americium readily undergo hydrolysis as the solution pH increases, with the ease of hydrolysis increasing with the atomic number. Most are complexed by a variety of ligands that are commonly present in groundwaters, including carbonate, fluoride, chloride, sulfate, and phosphate. Other ligands may be generated by radiolysis of air, such as nitrate and nitrite. In general, complexation decreases in strength as $\text{OH}^- > \text{CO}_3^{2-} > \text{PO}_4^{3-} > \text{F}^- > \text{SO}_4^{2-} > \text{HPO}_4^{2-} > \text{Cl}^-$, NO_3^- , and NO_2^- [228]. Because of its high complexing strength and natural occurrence in most groundwaters, carbonate is usually the dominant ligand for actinides dissolved in natural groundwaters [217]. Other metals released during waste form corrosion will also be complexed by these ligands.

Anions present in the waste form that are released as it corrodes will also be available to complex actinides. The anion contents of high-level waste glasses are seldom reported. Sulfur, nitrogen, chlorine, and fluorine are poorly soluble in borosilicate glasses and are usually assumed to be lost as off-gasses during vitrification. Phosphorus is poorly soluble in silicate glasses and phosphorus-bearing phases have been found to precipitate in borosilicate waste glasses [229,230]. Except in highly reducing solutions, technetium exists as the pertechnetate ion, Tc_2O_7^- , and is not complexed.

7.1 Summary of the Chemistries of Tc, U, Np, Pu, and Am

The chemistry of the fluid in contact with the waste form, including the redox state, pH, and available complexants, strongly affects the solubility of multivalent glass components, including the radioelements Tc, U, Np, Pu, and Am. The fluid chemistry will be significantly altered due to corrosion of the glass and other materials in the near-field environment. Changes in the Eh, pH, and ionic strength due to the release of glass components into the solution during corrosion will affect the solubilities and sorptive properties of the radioelements.

The dominant oxidation states in solution under oxidizing conditions are Tc(VII), U(VI), Np(V), Pu(V), and Am(III), while under reducing conditions Tc(IV), U(IV), Np(IV), Pu(III,IV), and Am(III) are dominant. Other oxidation states may be stabilized by complexation. Important complexants include naturally occurring carbonate and humic acids in the groundwater, and may include phosphate, sulfate, and fluoride released from the waste form during corrosion. Complexation may increase or decrease the soluble amounts of radionuclides, depending on what solid phase controls the solubility.

The extent of sorption of actinides onto mineral surfaces generally decreases as the oxidation state increases, as $\text{An(III, IV)} > \text{An(V)} > \text{An(VI)}$. Uranium in the hexavalent state and neptunium in the pentavalent state are poorly sorbed onto most mineral surfaces, while tetravalent plutonium and trivalent americium are strongly adsorbed onto most geologic minerals. Cationic complexes are sorbed much more strongly than are anionic complexes.

The behavior of uranium is expected to be dominated by the chemistry of the uranyl ion, UO_2^{2+} . It is strongly hydrolyzed and complexed by several ligands which may increase the uranium solubility. Carbonate and phosphate complexes are expected to be present. Reduction to U^{4+} may occur under mildly reducing conditions. U^{4+} is more strongly complexed than UO_2^{2+} , and is more strongly sorbed by mineral surfaces. Uranium is sparingly soluble under reducing conditions with the species U(OH)_4 being the dominant solution species [231]. Uranium is much more soluble under oxidizing conditions, where the species $\text{UO}_2(\text{OH})_3(\text{CO}_3)^-$ and $\text{UO}_2(\text{CO}_3)_3^{4-}$ are dominant solution species at basic pH values [232-234]. The solubility limiting phase is expected to be $\text{UO}_2(\text{OH})_2$ under oxidizing conditions and UO_2 under reducing conditions [235].

The behavior of neptunium is expected to be controlled by the chemistry of pentavalent neptonyl as NpO_2^+ under mildly oxidizing conditions [236] or Np(OH)_4 under mildly reducing conditions [237,238]. Neptunium(V) as NpO^+ is more soluble and less strongly hydrolyzed and complexed than Np(IV) ; $\text{NpO}_2(\text{CO}_3)_3^{5-}$ and $\text{NpO}_2(\text{CO}_3)_2^{3-}$ become important above about pH 8 in oxidizing solutions [239]. The solubility limiting phase is expected to be NpO_2 under both oxidizing and reducing conditions [235].

The behavior of plutonium may be controlled by the chemistry of Pu^{4+} , PuO_2^+ , or PuO_2^{2+} depending on the redox potential of the solution. Under slightly reducing conditions where Pu(IV, V) dominate, important species will include Pu(OH)_4 or $\text{PuO}_2(\text{CO}_3)_3^{5-}$ and $\text{PuO}_2(\text{CO}_3)_2^{3-}$ at high carbonate contents [217]. Higher solubilities are expected at pH values above about 9 where the $\text{PuO}_2(\text{CO}_3)_3^{5-}$ species dominates the solution. The solubility limiting phase is expected to be $\text{PuO}_2(\text{OH})_2$ or PuO_2CO_3 under oxidizing conditions [235] and PuO_2 , PuO_2CO_3 , or Pu(OH)_4 [240] under reducing conditions.

The behavior of americium is expected to be controlled by the chemistry of the trivalent Am^{3+} species. The Am(OH)_4^+ [241] and AmCO_3^+ species [242] and organic complexes are expected to be important. The solubility limiting phase is expected to be Am(OH)_3 under high pH (>10) conditions and $\text{Am(CO}_3)_3$ under low pH conditions [235].

The behavior of technetium is expected to be dominated by the chemistry of the highly soluble pertechnetate ion, TcO_4^- , which is not hydrolyzed [243,244], complexed, or sorbed onto mineral surfaces. Tc(IV) species may dominate under reducing conditions. The behavior of Tc(IV) species is very different from Tc(VII) , and Tc(IV) may become complexed and strongly sorbed onto mineral surfaces [245]. The solubility limiting phase is expected to be TcO_2 under most conditions [246].

7.2 Experimental Measurement of Radionuclide Release

The disposition of individual radionuclides between mobile and immobile phases as glass corrodes is strongly influenced by the solution chemistry and the presence of complexants and various sorbents. As presented above, technetium, uranium, and neptunium are generally more soluble than plutonium or americium in oxic solutions at the basic pH values expected in most repositories and attained in most tests, and are expected to attain higher solution concentrations in leach tests. Laboratory tests do show technetium, uranium, and neptunium to be released into solution to a greater extent than either plutonium or americium during corrosion of several glass compositions, including PNL 76-68 [247], R7T7 [248,249], ATM-8 [95,250], SRL 131 [67], SRL 165 [95], and SRL 202 [67,112]. Likewise, higher solution concentrations of Cs than the less soluble Pu or Am were observed during leaching of SRP glass [251], JSS-A glass [252], and TOKAI glass [253]. In other tests, technetium was found to be released congruently with boron under oxidizing conditions [3]. Actinides which do not enter solution due to low solubilities may be retained in the surface layers of the altered glass, either by sorption or incorporation into secondary phases, or may become sorbed on the surfaces of other materials that may be present in the vicinity of the corroding glass.

The conditions of the test will strongly influence the behavior of the radionuclides observed during the test. Tests which maintain dilute solution conditions highlight the reactions releasing the radionuclides from the glass matrix and their solubilities in the leachant. Tests which result in the accumulation of glass corrosion products in the corroding fluid will show effects of colloids and secondary phases on the disposition of the released radionuclides, in addition to changes in the fluid chemistry resulting from glass corrosion, such as higher pH values.

7.2.1 Tests Conducted under Dilute Solution Conditions

Monolithic samples of the borosilicate glass I 117 were reacted in deionized water at a SA/V of 25 m^{-1} at 90°C for 7, 10, 20, and 40 days in the presence of oxygen/argon, carbon dioxide/oxygen/argon, carbon dioxide/hydrogen/argon, and hydrogen/argon mixtures [254]. The presence of carbonate ions was found not to affect the mass loss of any glass component, including uranium, while the extent of glass corrosion was greater under oxidizing conditions. Uranium was found to be depleted in the alteration layers.

The retention of uranium in the surface layer was greater after leaching in an anoxic atmosphere than in air [254]. The uranium solubility is expected to be greater in the presence of air due to formation of carbonate species [255]. Deaeration of deionized water, a bicarbonate solution, and a salt brine was previously seen to decrease the solution concentrations of Np, Pu, and U in tests with PNL 76-68 glass [256].

The effect of the redox state on the solubility and chemical behavior of released actinides was studied using dynamic leach tests with crushed borosilicate glass I 117 doped with 2.4 wt% Np, 0.01 wt% Pu, and 0.03 wt% Am under oxidizing and reducing conditions [257]. The 0.95-1.4 mm size fraction was used in the tests. The glass was reacted in flowing solutions of bicarbonate groundwater (55.2 ppm Na, 159 ppm HCO_3^-) and in a salt brine (78 ppm K, 164 ppm Ca, 538 ppm SO_4^{2-}) under both oxidizing (air-saturated) and reducing ($E_h \leq -200 \text{ mV}$ measured against Pt electrodes) conditions. The flow rate was $1.5 \text{ cm}^3/\text{h}$ and the tests were conducted at 25°C . The glass corrosion rate (based on the sodium release rate) was about $1 \times 10^{-6} \text{ g/m}^2/\text{d}$. The steady-state solution concentration of Am was found to be the same under both oxidizing and reducing conditions because Am(III) is the dominant oxidation state under both test conditions. The measured release rates were $3 \times 10^{-7} \text{ g/m}^2/\text{d}$ in the bicarbonate water

under both oxidizing and reducing conditions. The release of americium was about an order of magnitude higher in the bicarbonate water than in the brine, under oxic conditions. On the other hand, Np(IV) and Pu(IV) were the dominant solution species under reducing conditions while Np(V) and Pu(V) were the dominant solution species under oxidizing conditions. The release rates of Np were 1×10^{-6} g/m²/d in both the bicarbonate solution and the brine under oxidizing conditions, and 6×10^{-8} g/m²/d in the bicarbonate solution under reducing conditions. The release of neptunium was about an order of magnitude lower in the brine under oxic conditions than under reducing conditions. The release rates of Pu were 4×10^{-7} and 3×10^{-7} g/m²/d in the bicarbonate solution and the brine under oxidizing conditions, and 9×10^{-8} g/m²/d in the bicarbonate solution under reducing conditions. The release of neptunium was about an order of magnitude lower in the brine, under oxic conditions.

The behavior observed in these tests is dominated by the solubilities of the different actinide elements. Because the solubilities of the An(V) species are higher than those of the An(III) and An(IV) species, the solution concentrations of both Np and Pu were found to be higher under oxidizing conditions, where they exist preferentially in the An(V) state, than under reducing conditions, where they exist in the An(III) state. Some reduction of Pu(V) at the glass surface was suggested to occur under oxidizing conditions [257]. Americium exists in the An(III) state except under highly oxidizing conditions, and its release behavior was not affected under either of the conditions tested. The actinides not released from the glass likely accumulated in the alteration layers that formed on the glass surface.

The release of Sr, Cs, Tc, Np, Pu, and Am from glass UK 189 into distilled water at 60°C was measured in flow-through tests at various flow rates [258]. The measured leach rates are given in Table 6 (data converted from g/cm²/d to g/m²/d by multiplying data by 10^4 cm²/m²). The corrosion rate of the glass UK 189 at high flow rates was reported to be 0.65 g/m²/d. The authors interpreted the finding that the technetium release rate was independent of the flow rate as evidence that its release is limited by its rate of escape from the glass rather than its solubility. The release of the other radionuclides decreased as the flow rate decreased, indicating that their release was affected by their solubilities. The release rates of glass matrix components were not reported. It is expected that the corrosion rate of the glass matrix would also decrease with the flow rate due to saturation effects. The results of Hall et al. may indicate that the release of technetium is not affected by the corrosion of the glass matrix.

After 270 days of corrosion, the alteration layers formed on the samples were removed and analyzed. The fractions of each isotope retained in the layer was calculated, and is included in Table 6. The high degree of retention of the actinides is consistent with other test results. The authors pointed out that cesium was retained, while lithium and sodium were readily released, and attributed the difference to the different ionic radii. The lower ion exchange rate of cesium probably also contributes to the observed retention. Notice also that the technetium is not completely depleted in the alteration layers. The retained technetium may have been reduced to the less soluble Tc⁴⁺ state during formation of the glass, though this is only speculation.

The corrosion of R7T7 glasses doped with ²³⁷Np, ²³⁹Pu, or ²⁴¹Am has been measured in leach tests with R7T7 glass run 70 days in deionized water at room temperature [249]. These tests were conducted with daily renewal of the leachant, which prevented accumulation of glass corrosion products. The extent of corrosion was small in these tests, and only thin surface layers were observed. The measured leach rates were 6×10^{-3} g/m²/d Np, 6×10^{-3} g/m²/d Pu, and 3×10^{-3} g/m²/d Am. The corrosion rate of the glass matrix was reported to be 12×10^{-3} g/m²/d. Thus, the actinide elements were released at rates slightly lower than the rate at which the glass was corroding.

Table 6. Leach Rates from UK 208 Glass in Distilled Water at 60°C (g/m²/d) [258]

	Flow Rate			% Retained in Layer
	9 mL/day	1 mL/day	1 mL/week	
⁹⁹ Tc	0.7	0.7	0.6	15
⁹⁰ Sr	0.5	0.2	0.05	62
¹³⁷ Cs	0.6	0.4	0.2	31
²³⁷ Np	0.18	0.1	0.03	91
²³⁹ Pu ^a	0.005	0.0013	0.00015	99.4
²⁴¹ Am	0.00004	0.000006	0.000006	99.8

^aIsotope not reported. Pu data reported as uncertain.

In 28-day Soxhlet tests of the same actinide-doped R7T7 glass at 100°C, release rates layers about 6 g/m²/d Np, 5 x 10⁻³ g/m²/d Pu, and 3 x 10⁻³ g/m²/d Am. The release rate of boron was reported to be 2 g/m²/d, which is taken to be the corrosion rate of the glass matrix. The corrosion rate of the glass increased about 150 times and the release of neptunium increased by a factor of 1000, while the plutonium and americium release rates were similar at 25 and 100°C. The low release rates of plutonium and americium were attributed to their retention in the alteration layer formed on the glass surface during the tests [249]. Layers about 20 mm thick were formed during the Soxhlet tests. These test conditions likewise maintain dilute solutions and minimize the effects of dissolved corrosion products while maintaining a high glass dissolution rate. It was concluded from these tests that the alteration layer formed on R7T7 glass effectively retained Pu, Am, and Cm, although Np was not significantly retained by the layers under these test conditions [249]. This is consistent with the higher solubility of neptunium.

The retention of any element *i* by an alteration layer during glass corrosion can be quantified by comparing the amount of that element in solution to the amount of boron in solution, as $RF(i) = NL(B)/NL(i)$, where $NL(i)$ is the normalized mass loss of element *i* [249,259]. Boron is assumed to be totally released into solution and to represent the extent of glass corrosion. High values of $RF(An)$ indicate that actinides remain largely immobile as the glass corrodes, while low values indicate that mobilization of the actinides may occur as the glass corrodes.

It should be emphasized that the retention factor is a measure of the release of actinides (and other elements) to solution relative to the release of boron and does not indicate the extent of glass corrosion. A low $RF(i)$ value simply means that element *i* is released from the glass at a rate similar to boron, which may be fast or slow. Indeed, changes in the release factor under different experimental conditions is as much a measure of the affect of the test conditions on the boron release as it is a measure of the release of less soluble elements.

Actinide retention factors for test results reported by Vernaz et al. are quite low for the tests at 25°C, ($RF(Np) = 2$, $RF(Pu) = 2$, and $RF(Am) = 4$), but higher for corrosion in the Soxhlet tests ($RF(Np) = 3.7$, $RF(Pu) = 412$, and $RF(Am) = 469$).

The release of uranium, neptunium, plutonium, and americium from R7T7 glasses into distilled water was measured in static leach tests with monolithic samples at SA/V of 50 m^{-1} at 50 and 90°C [167,248]. Both the normalized boron release and the normalized actinide releases increased with the reaction time. Boron, uranium, and neptunium were dissolved in the leachates, while most of the plutonium and americium in the leachate was present primarily as suspended material that was removed using a 25 nm filter. The suspended material was considered to be released from the glass and was included in the released fraction. The normalized release of boron and uranium from R7T7 glass doped with natural uranium and boron and neptunium from R7T7 glass doped with ^{237}Np in tests at 90°C are shown in Fig.47a. The boron release is similar from the R7T7 glasses doped with either natural uranium or with neptunium, and the normalized releases of uranium and neptunium are similar. The boron release is rapid initially, then levels off at longer reaction times. The normalized release of boron and plutonium from R7T7 glasses doped with ^{238}Pu or ^{239}Pu and boron and americium from R7T7 glass doped with ^{241}Am in tests at 90°C are shown in Fig.47b. The boron release is similar from the R7T7 glasses doped with either plutonium or americium through 91 days, but the boron release from glasses doped with ^{238}Pu or ^{241}Am increases significantly after longer reaction times. The normalized releases of plutonium and americium remain low and are not affected by the increased glass corrosion rate at 182 days or longer. Vernaz et al. ascribed the differences in reactivity to radiolysis and the resulting differences in the solution pH. The measured solution pH values for tests at 90°C are shown in Fig. 47c. Tests conducted with the U, ^{237}Np , or ^{239}Pu doped glasses attain higher pH values than the glasses with the ^{238}Pu or ^{241}Am doped glasses, which have higher specific activities. The solution pH reflects the effects of glass corrosion, which raise the pH, and radiolysis of the air in the test vessel, which lower the pH.

The measured releases of the same elements in identical tests conducted at 50°C are shown in Figs.48a and b. Notice that the boron continues to be released from the glasses doped with U, ^{237}Np , and ^{239}Pu through 364 days, but reaches a nearly constant value in tests with glasses doped with ^{238}Pu and ^{241}Am . The normalized mass losses of ^{237}Np increases slightly with the reaction time, but the mass losses of the other actinides remain low at all test times. The lower corrosion rates measured in tests at 50°C (as measured by the boron release) correspond to the much lower solution pH values attained in tests with ^{238}Pu and ^{241}Am compared to tests with U, ^{237}Np , and ^{239}Pu . The measured pH values are shown in Fig. 48c. The relative effects of glass corrosion and radiolysis on the solution pH are different in tests at 50 and 90°C because of the lower glass corrosion rate at 50°C. The effect of radiolysis is less sensitive to the temperature than the glass corrosion rate, and so the effects of radiolysis lower the solution pH more in tests at 50°C than in tests at 90°C.

The logarithms of the measured retention factors for tests conducted at 90°C are plotted in Fig. 49a. The data show that all actinides are retained in the layers to some extent ($\log(\text{RF}) > 0$ for all actinides at all times), with americium and plutonium being retained to the highest degree. The increase in the boron release from the ^{238}Pu and ^{241}Am doped glasses results in a slight decrease in the retention factors of these elements. Overall, the retention factors remain essentially constant over all times tested. As the glass corrodes under these conditions, only about 10% of the uranium and neptunium, 3% of the plutonium, and 0.3% of the americium are released into the solution. The balance accumulates in alteration layers on the glass surfaces.

Fig. 47. Measured Normalized Mass Losses in Tests with Actinide-Doped R7T7 Glasses in Tests at 90°C and 50 m⁻¹: (a) (○) U and (●) B from U-Doped R7T7 Glasses and (□) Np and (■) B from Np-Doped R7T7 Glass; (b) (△) Pu-238; (▲) B from Pu-238-Doped R7T7 Glass, (◇) Pu-239, and (◆) B from Pu-239-Doped R7T7 Glass; and (c) Measured pH Values for R7T7 Glass Doped with (○) U, (□) Np, (△) Pu-238, (◇) Pu-239, and (Δ) Am-241 (after [249])

Fig. 48. Measured Normalized Mass Losses in Tests with Actinide-Doped R7T7 Glasses in Tests at 50°C and 50 m⁻¹: (a) (○) U and (●) B from U-Doped R7T7 Glasses and (□) Np and (■) B from Np-Doped R7T7 Glass; (b) (Δ) Pu-238; (▲) B from Pu-238-Doped R7T7 Glass, (◊) Pu-239, and (◆) B from Pu-239-Doped R7T7 Glass; and (c) Measured pH Values for R7T7 Glass Doped with (○) U, (□) Np, (Δ) Pu-238, (◆) Pu-239, and (Δ) Am-241 (after [249])

Fig. 49. Calculated Retention Factors vs. Reaction Time for Tests at (a) 90°C and (b) 50°C with R7T7 Glass Doped with (○) U, (□) Np, (◇) Pu-238, (◆) Pu-239, and (△) Am-241 (after [249])

The retention factors calculated from the normalized mass losses in tests at 50°C are shown in Fig. 49b. The retention factors measured in tests conducted at 50°C are about half those measured in tests conducted at 90°C. Part of the difference is due to the different reactivities of the glasses at 50 and 90°C and the differences in the solution pH. (The dopant oxides were present at 0.85 w% in all glasses, and probably didn't affect the intrinsic reactivity of the R7T7 glass.)

A Savannah River Laboratory glass generated by mixing SRL 165 with radioactive waste was corroded in a tuff groundwater solution [251]. The glass composition differed from SRL 165 mainly in the aluminum and iron contents, which were measured to be 9.8 wt% Al_2O_3 and 6.0 wt% Fe_2O_3 in the glass. The groundwater solution contained about 42 ppm sodium and 27 ppm silica [260] and typically 120 ppm bicarbonate and 30 ppm sulfate. Tests were conducted using monolithic samples at an SA/V of 100 m^{-1} and at 90°C for up to 134 days. Figure 50 shows the normalized mass loss of ^{238}Pu calculated based on the amount of plutonium measured in the leachate (circles) and based on the total amount found in the leachate plus fixed to the steel vessel (squares). Most of the plutonium becomes fixed to the steel vessel during the test.

Tests were conducted using actinide-doped SRL 165 glass in tuff groundwater at an SA/V of 30 m^{-1} and at 90°C for up to 278 days [95]. Figure 51 shows the average retention factors calculated for uranium, neptunium, plutonium, and americium. All the uranium and most of the neptunium were dissolved or suspended in the leachate solution, while most of the plutonium and americium were fixed to the steel reaction vessel. Material fixed to the vessel

Fig. 50. Normalized Mass Loss of Plutonium-238 from SRL 165 Glass in Tests at 100 m^{-1} Based on (●) ^{238}Pu in Solution and (■) ^{238}Pu in Solution Plus on Vessel Walls (after [251])

Fig. 51. Calculated Retention Factors and Measured pH Values for Tests with Actinide-Doped SRL 165 Glass at Tests at 30 m^{-1} : (●) U, (■) Np, (◆) Pu, (▲) Am, and (+) pH (after [95])

was counted as being released from the glass. The average solution pH values measured at room temperature are included in the plot. The relative retention of the different actinides is consistent with their solubilities: americium is retained to the highest degree and is the least soluble, uranium and neptunium have relatively soluble and are only slightly retained. Surface analyses verified the relative concentrations of the actinides in the surface layers [95]. The relatively low SA/V of these tests maintained sufficiently dilute conditions that the disposition of the actinides was not significantly affected by glass corrosion products in the solution. However, the release was sensitive to the solution pH.

7.2.2 Tests under Concentrated Solution Conditions

The disposition of actinides in tests conducted in flowing solutions and at low SA/V is dominated by the actinide solubilities. While the disposition may be affected by interactions with species in the groundwater, it is only weakly affected by interactions with glass components in the solution. The effects of dissolved glass components have been shown to be very important in understanding the long-term behavior of the glass matrix, and are likely to be just as important to the disposition of released radionuclides. Tests conducted at high SA/V highlight the interactions between the leachate solution and the dissolving glass. The effects of the formation of alteration layers and secondary phases are highlighted in these tests. These effects will be important under disposal conditions, where only small volumes of water are likely to contact the waste glass.

The corrosion of two Savannah River glasses (SRL 165/42 and SRL 200R) was measured using the PCT test. The glasses were crushed and the -100+200 mesh size fractions were reacted in deionized water at 90°C for 7 days [261]. Tests were conducted in two laboratories to assess the reproducibility of the test method. The release of components from the

glass matrix (B, Li, Na, Si) agreed within 10%. The SRL 165/42 glass was measured to be less corroded than the SRL 202R glass (the average normalized concentrations of boron were 0.37 g/L for SRL 165/42 glass and 1.1 g/L for SRL 202R). The release of radioactive components agreed within a factor of about 2. The larger variation in the radionuclide results may be attributed to greater analytical difficulties. The average retention factors of radioactive elements (calculated from the ratio of the normalized concentrations of boron and the radionuclides of interest) measured by the different laboratories are: $RF(^{137}\text{Cs}) = 5$, $RF(^{90}\text{Sr}) = 12$, $RF(^{238}\text{Pu} + ^{239}\text{Pu} + ^{244}\text{Cm}) = 5$ for SRL 165/42, and $RF(^{137}\text{Cs}) = 6$, $RF(^{90}\text{Sr}) = 264$, $RF(^{238}\text{Pu} + ^{239}\text{Pu} + ^{244}\text{Cm}) = 15$ for SRL 200R. By comparison to the results of a nonradioactive glass having a composition similar to SRL 202R, the authors determined that radiolysis did not affect the test results. This is probably due to the relatively high solution concentrations attained in PCT tests compared to those attained in tests at 50 m^{-1} (cf. discussion of [249] above, where radiolysis effects were observed).

Tests conducted at higher SA/V ratios and for longer time periods have shown that behavior of the radioactive components becomes more or less independent of the glass corrosion behavior at advanced stages of the corrosion process. For example, the increased corrosion rate observed in tests with actinide-doped SRL 202 glass (see Fig. 37) did not affect the disposition of released actinides [67]. That is, the amounts of Np, Pu, and Am found either suspended in solution as colloids, dissolved in solution, or fixed to steel vessel were not affected by the increased glass corrosion rate at reaction times longer than 182 days. (These results are discussed in more detail in Section 9.2.3.) These results imply that, while the corrosion rate of the glass matrix provides an upper boundary to the release of actinides, other factors limit their release rates at much lower levels.

7.3 Actinide-Bearing Secondary Phases

Identification of the solid phases which control radionuclide solubilities is required to accurately predict the radionuclide behavior during glass corrosion in repository setting. While phases controlling solubilities in pure systems may be identified by precipitation from supersaturated solutions (e.g., [219]), solubility-controlling phases are not easily identified in tests with waste glasses because most laboratory tests are performed using simulated glasses with low concentrations of radioelements, and the phases formed with radioelements are typically very small and interspersed with several other phases. Identification of phases containing radionuclides is complicated by the difficulties associated with handling radioactive samples and the limited sensitivity of analytical methods. Nevertheless, progress has been made in identifying phases formed which contain radionuclides.

Leach tests with relatively high uranium contents have lead to the identification of several uranium-bearing phases which form during glass corrosion. Weeksite, a uranium silicate, is the most commonly formed uranium-bearing mineral during glass corrosion [61,111,108]. Other uranium silicates have been found to form, including uranophanes and haiweeite [112,113,262,263]. These phases are generally formed separate from the glass, although they may nucleate on the glass surface, and crystallites may remain suspended in the leachate. A uranium- and titanium-bearing phase (suggested to be brannerite) was also observed to form on SRL 131 glass [62] and, with thorium, within the altered layer of WVCN 50 glass [111]; a similar phase was also found suspended in solution [263].

Less is known about the phases affecting the solubilities of Np, Pu, and Am or their incorporation into secondary phases due to the low levels of these elements in glasses used in laboratory tests and limitations of analyzing these elements. In addition, the relatively high solubility of neptunium and the fact that plutonium and americium are preferentially fixed onto metal surfaces results in only small amounts being available for secondary phase formation in

leach tests. Plutonium and actinide-bearing phases are typically very small and may be distributed on the altered glass surface or may be suspended in the leachate as colloids. Characterization of plutonium and actinide-bearing phases has been achieved by filtering solutions through holey carbon grids and analyzing the filtrant in an electron microscope [262-264]. Microcrystallites of brockite $[(Ca,Th)PO_4 \cdot H_2O]$ which have incorporated plutonium and americium have been observed both isolated and supported on colloidal clay particles removed from solution [262]. The similarity between clay colloids removed from solution and clays at the surfaces of reacted glass suggests that material may slough from the reacted glass surface and become suspended in solution in a mobile state [262]. Thus, radionuclides found immobilized during short-term tests may become mobilized due to mechanical separation at longer exposure times.

The chemical similarity of radioelements to nonactive elements will result in the substitution of radioelements into other mineral phases which precipitate out of solution, and minerals may incorporate some radioelements via ion exchange. For example, ^{137}Cs may become incorporated into analcime, a sodium aluminosilicate which commonly forms during glass corrosion [61,112,191]. Other zeolites may also incorporate radionuclides. Crystalline lanthanoid phases such as powellite and cerianite-type materials, which may host Am and Cm, were found during the reaction of R7T7 glass in salt solutions [265]. Incorporation of radioelements into mineral phases or their sorption onto mineral phases may be immobilize for different durations, depending on the stability of the host phase. Aging of the mineral phases formed due to glass corrosion will result in thermodynamically more stable phases, which may have a reduced affinity for radionuclides than previous phases.

Colloid formation will also affect the disposition of radioelements over long time periods. Laboratory tests show that aluminum and iron colloids are formed during glass corrosion [167], and that these may sorb radionuclides [67,266]. Depending on the colloid size, solution chemistry, groundwater flow and the characteristics of the surrounding geology, these colloids may or may not be transportable, and their transportability may change over time. Colloids which settle out of solution during static leach tests may behave differently under dynamic conditions. Likewise, incorporation into alteration phases on the glass surface may only temporarily immobilize the radionuclides. Actinide-bearing colloids may be formed by exfoliation of alteration phases from corroded surfaces under stress or over long time periods [262].

7.4 Effects of Other Materials on Actinide Disposition

The extent of accumulation of actinides in the altered layer will depend on the presence of other materials in the vicinity of the corroding glass onto which released actinides may sorb, such as clay backfills or metal barrier components. These materials may act as sinks to remove dissolved actinides from solution and provide additional barriers against radionuclide migration. Tests with R7T7 glass showed the amounts of Np and Pu retained in surface layers varied when other materials, including sand, granite, bentonite, smectite, illite, Boom clay, and salt were included in the test vessel [249]. The presence of these materials also affects the solution chemistry and the glass corrosion rate [267]. In general, the solutions in tests with the added materials contained more Np than those in tests with distilled water alone, but less Pu. Smectite, illite, and Boom clays sorbed Np(weakly) and Pu(strongly), granite and bentonite sorbed Pu (strongly), and salt sorbed Pu (weakly) [249]. It was also found that the amount of plutonium in the altered layer remained high if the smectite was not in direct contact with the glass ($RF(Pu) = 80$), but was significantly reduced if the clay directly contacted the glass ($RF(Pu) = 6$) [249]. This implies that diffusion of plutonium through the solution limits the amount sorbed onto the clay surface.

Leach tests in a slurry of Boom clay showed retention of Np and Pu in layers formed on R7T7 glass and on a Pamela glass SM513, but a third glass, Pamela SM527, did not generate an alteration layer and did not retain these elements [268]. Technetium was not retained by the layers. The Pu and Am released from the glass was strongly sorbed onto the clay, while the Tc and Np were only partially sorbed onto the clay. The plutonium and americium had attained steady-state concentrations after two years of reaction (both below 2×10^{-9} M), while neptunium was present at about 2×10^{-6} M and technetium at about 4×10^{-7} M. Further, addition of Fe_2O_3 to the tests slowed the corrosion of all three glasses by at least a factor of 68 [268].

Released radionuclides may be immobilized by incorporation into or sorption onto mineral phases present naturally or generated by corrosion of repository materials. Immobilization by sorption onto repository components or geologic minerals may be transient, depending on the stabilities of the host phases. Conversely, sorption onto colloids present in the groundwater or generated during glass corrosion may mobilize radionuclides in excess of their solubilities. Sorption of radionuclides onto colloids generated during glass corrosion will strongly affect the distribution of actinides. Sorption is specific to the host mineral phase and the oxidation state of the radionuclide: sorption of lower oxidation states is generally stronger than that of higher oxidation states. Sorption may result in a change in the oxidation state of the radionuclide. Competition between actinides and other metal ions in complexation and sorption reactions results in complex chemistries in real systems, and will, in general, decrease the amounts of radionuclides removed from solution.

Colloidal materials that occur naturally in the groundwater or are generated during corrosion of engineered barrier materials will be available to sorb radionuclides [193,269]. This material may remain suspended in solution, or may eventually be sorbed by immobile phases, may be filtered from the solution by microprobes, or may flocculate and settle out of solution. Sorption is generally stronger for actinides in lower oxidation states, so that Am(III) and Pu(IV) are expected to be more strongly sorbed to colloids and mineral phases than U(VI) or Np(V) [270]. The highly soluble pertechnetate ion is not expected to be sorbed onto colloids.

Migration of radionuclides released from the glass will be affected by the many materials present in the geologic formations or engineered barriers surrounding the waste form, as well as interactions involving the corrosion products of the waste glass itself. Various clays, zeolites, and other mineral phases are known to form as glass waste forms corrode [112,271], and interactions between released radionuclides and these secondary phases may either enhance or reduce their release from the immediate vicinity of the waste form, depending on the mobilities of the particular phases. Species having low valences, such as Am(III) and Pu(IV), generally interact more strongly with sorbents than species with higher valences, such as Np(V) and U(VI). Technetium (VII) does not sorb onto mineral phases, but sorption may occur on some surfaces after reduction to Tc(IV). Reliable long-term predictions of waste behavior must account for these interactions.

Colloidal material formed by naturally occurring inorganic and/or organic materials or microorganisms in the groundwater will be available to sorb radionuclides from the solution to form pseudocolloids that may remain suspended in the groundwater or may flocculate and settle out of solution [269,272,273]. Sorption may occur through dispersive interactions (physisorption), coulombic interactions (electrostatic adsorption and ion exchange), or electron exchange (chemisorption). The presence of colloids bearing radionuclides may lead to increased aqueous concentrations which far exceed the solubility limits of pure phases. Depending on the size and transportability of these colloids, radionuclides may become available for dispersal away from a repository site by groundwater flow or may become immobilized as the colloids settle out of solution. Colloids initially suspended in the groundwater may eventually interact

with geologic materials after longer times or as they migrate away from the waste form so that the colloids become filtered by micropores or as radioelements become more strongly sorbed onto mineral surfaces present in the host rock [274,275].

The sorption energy of the actinides onto mineral surfaces generally decreases as $An(III,IV) > An(V) > An(VI)$. Uranium in the hexavalent state and neptunium in the pentavalent state are poorly sorbed onto most mineral surfaces, while tetravalent plutonium and trivalent americium are strongly adsorbed onto most geologic minerals. Sorption of radionuclides that exist in high valence states in solution may occur on sites capable of reducing the radionuclide to a lower oxidation state. Several studies have been performed to characterize the sorption of radionuclides onto mineral surfaces, both onto monolithic samples and onto mineral colloids. Some are discussed below.

In addition to actinides, sorption sites on mineral surfaces have an affinity for other solution species, including metal ions, complexants, and dissolved humic materials. Competition between different sorbants for surface sites and complexation of actinides in solution will affect the amount of actinides that may become sorbed onto mineral colloids in solution or minerals fixed in rocks.

7.5 Summary

The disposition of individual radionuclides between mobile and immobile phases as glass corrodes is strongly influenced by the solution chemistry and the presence of complexants and various sorbents. Technetium, uranium, and neptunium are generally more soluble than plutonium or americium in oxic solutions at the basic pH values expected in most repositories and attained in most tests, and are expected to attain higher solution concentrations in leach tests. Laboratory tests show technetium, uranium, and neptunium to be released into solution to a greater extent than either plutonium or americium during corrosion of several glass compositions. Actinides which do not enter solution due to low solubilities may be retained in the surface layers of the altered glass, either by sorption or incorporation into secondary phases, or may become sorbed on the surfaces of other materials that may be present in the vicinity of the corroding glass.

Identification of the phases controlling the solubilities of the actinides remains an important information need for predicting long-term behavior of high-level waste glasses. Tests conducted under conditions leading to saturation of the corroding fluids and the precipitation of phases affecting the solubility of released radionuclides are needed to identify important phases. Also, the effects of colloid formation is a critical uncertainty that must be addressed in performance models.

8.0 SUMMARY OF CRITICAL REVIEW OF EFFECTS OF SA/V ON GLASS CORROSION AND DISCUSSION OF REMAINING UNCERTAINTIES

This report has summarized the present state of knowledge regarding the effects of the glass surface area/solution volume (SA/V) ratio on the corrosion behavior of borosilicate waste glasses. The SA/V ratio affects the rate of glass corrosion through the extent of dilution of corrosion products released from the glass into the leachate solution. The observed effects of the SA/V used in a static leach test were interpreted in terms of differences in solution chemistries attained in tests at different SA/V. A detailed discussion of the effects of the solution chemistry on the glass corrosion rate and mechanism was provided to support interpretations of the effects of the SA/V on a mechanistic scale. Glass corrosion was discussed in some detail by first presenting a brief summary of glass structure and then discussing the processes leading to the alteration and dissolution of the glass structure. Laboratory tests performed to elucidate the corrosion process and the relevance of the information they provide to the corrosion behavior and the effects of the solution chemistry on glass corrosion. The results of static leach tests conducted specifically to characterize the effects of the SA/V on glass corrosion are then discussed based on what is known about the effects of the solution chemistry on glass corrosion. Observed effects of the SA/V, solution chemistry, and test procedure on the disposition of radionuclides during glass corrosion was discussed. Key findings are summarized below.

8.1 Observed Corrosion Behavior

Laboratory tests conducted to distinguish the effects of the fluid chemistry and glass alteration layers on glass dissolution has shown that the effects of the fluid chemistry dominate the glass corrosion rate in tests conducted at relatively low SA/V where the solution does not become saturated with silicic acid. The silicic acid concentration and the pH affect the rate most strongly, with the reaction to release silicic acid usually being modeled as the key corrosion step. The rate of silicic acid release from the glass depends on the temperature, glass composition, silicic acid concentration in solution, and the solution pH. For a given glass composition at a given temperature, any test conditions that affect the silicic acid concentration or the solution pH will affect the glass corrosion rate. The SA/V used in the test affects the corrosion rate through dilution of the silicic acid and pH. That is, tests at low SA/V dilute the silicic acid more than tests at high SA/V and also maintain lower pH values (assuming that corrosion of glass results in an increased pH).

The silicic acid concentration and the solution pH are also affected by the release of other glass components as the glass corrodes, the volume of solution available to dilute released glass components, the refreshment of the leachate solution due to solution flow, if any, and the formation of secondary phases. The relative importance of these effects in a particular experiment will be determined by the glass and initial fluid compositions, and by the test method and test conditions utilized. Glasses having large amounts of alkali metals will generate higher solution pH values as the glass corrodes than glasses with less alkali, and the effect of the SA/V on the pH will be greater in tests generating higher pH values. Less durable glasses will attain more concentrated solutions after shorter corrosion times than more durable glasses, and the effects of the SA/V and flow on the corrosion rate will be greater in tests with less durable glasses. In addition, the extent of corrosion of less durable glasses will be greater than that of more durable glasses in a common laboratory test, so that the effects of SA/V and flow occur at a more advanced stage of corrosion in tests with the less durable glass. Species in the leachant may affect the corrosion through interactions with the glass surface, such as ion exchange and precipitation, or by affecting the solubilities of released glass components, such as the formation of transuranic carbonates. Of course, the silicic acid concentration in the fluid contacting the glass and the solution pH will also affect the glass corrosion. In general, the glass and solution

compositions are determined by the waste form and the disposal site, while the test conditions may be selected to simulate particular environmental conditions, highlight a particular reaction step, or characterize the general corrosion behavior.

Past efforts to describe glass corrosion behavior has been mostly empirical, where sets of data were fit by simple analytic expressions, such as diffusion equations. However, reliable predictions of long-term glass corrosion behavior cannot rely on empirical correlations to short term data due to the complex nature of glass corrosion and changing conditions of temperature, solution chemistry, etc. anticipated to occur over the service life of a waste repository. That rates measured in short-term laboratory tests cannot be extrapolated to long times is clearly demonstrated by the observation that the glass corrosion rate varies by about three orders of magnitude as corrosion proceeds, even under static reaction conditions. Instead a mechanistic model of the corrosion processes is needed to account for the effects of varying solution chemistries and flow rates, as well as varying temperatures over the long time periods that radioactive wastes must be contained.

8.2 Modeling

Glass corrosion is well described by a rate equation that combines a rate coefficient that depends on the glass composition, a pH-dependence, and a reaction affinity term expressed as the difference between the actual silicic acid concentration and a "saturation" value. Use of this equation requires the experimental determination of (1) the rate coefficient and its temperature dependence, (2) the pH dependence of the rate coefficient, and (3) the saturation value. Laboratory testing has shown that, under some conditions, a residual affinity remains under saturated conditions so that a final rate term must be measured experimentally. In other tests, secondary phases that precipitate from highly concentration solutions have shown to control the silicic acid concentration after long corrosion progress. These phases must be identified to predict the long-term corrosion behavior. Other phases present in a disposal system may likewise affect the solution chemistry, particularly the solution pH and silicic acid concentration, and influence the corrosion rate. These effects must also be accounted for.

The corrosion of borosilicate waste glasses can be described as occurring in three stages, which are related to the solution chemistry. The first stage occurs when the solution concentration of silicic acid is very low and the glass corrodes at a relatively high rate that depends on the glass composition and the solution pH. This is sometimes referred to as the "intrinsic" rate, since it depends on the glass composition and is not affected by the affinity term. The second stage occurs as the silicic acid concentration increases to a maximum level, the so-called "saturation" concentration and the glass corrosion rate decreases to a minimum value. The decrease in the corrosion rate is due to a decrease in the reaction affinity. A third stage occurs when secondary phases precipitate from solution to establish a silicic acid solubility limit that may be different than the "saturation" concentration. This change affects the rate through the affinity term, because the presence of secondary phases causes the silicic acid concentration to be maintained below the apparent saturation value imposed by the glass.

8.3 Testing

Different laboratory tests can be used to highlight particular aspects of the corrosion process. An understanding of how different test conditions affect the solution conditions and the glass corrosion process is required to correctly utilize the test results to describe the corrosion behavior and determine values for model parameters. Tests with large solution volumes relative to the exposed glass surface area (low effective SA/V), such as MCC-1 and flow-through tests, will maintain dilute conditions and the glass corrosion rate will be unaffected or only weakly affected by the minor accumulation of silicic acid in solution. These tests are used to determine the intrinsic corrosion rate and the pH-dependence of the corrosion rate. Tests that allow silicic

acid to accumulate in the solution, such as the MCC-3 test and the Product Consistency Test, show the effects of the affinity term on the corrosion rate and the approach to saturation. If these tests are conducted for sufficiently long time periods, secondary phases will precipitate from the saturated solutions and affect the corrosion rate. The corrosion process may be affected by secondary phase formation under some test conditions, although reaction times of several months or years may be required before secondary phases precipitate. The Vapor Hydration Test developed at ANL was designed to quickly attain saturation of the solution and drive the corrosion process to the point where the solution chemistry was controlled by secondary phases within a short time period.

While many laboratory tests have been developed to investigate the corrosion process, these are rarely related to modeling efforts either with regard to elucidating corrosion processes or deriving values of model parameters. This divergence has resulted in a situation where many studies are done on the basis of a test procedure that provides little or no information relevant to describing the corrosion behavior. A need exists to clearly relate test procedures and test results to a mechanistic description of the glass corrosion process that can be used to predict long-term glass durability and conduct performance assessments of disposal systems.

8.4 SA/V

The SA/V ratio used in static leach tests affects the dilution of the reaction products, including the silicic acid concentration and the pH, both of which affect the glass corrosion rate. The SA/V affects how fast the solution chemistry changes as the glass occurs, and a value can be selected to highlight the early stages of the reaction under dilute conditions (at low SA/V) or the more advanced stages of corrosion under more concentrated conditions (at high SA/V). The solution results of tests conducted at different SA/V ratios are conveniently plotted as a function of the product of the SA/V and the corrosion time to include data on a single plot. The product (SA/V)t can be used to estimate the reaction progress. That is, tests conducted for greater (SA/V)t attain a greater extent of reaction, as measured by the release of glass components. Plotting solution concentrations as a function of (SA/V)t is based on the different dilution effects of tests at different SA/V, assuming the glass corrodes at the same rate at both SA/V. However, two caveats exist regarding the comparison of data measured at different SA/V but common (SA/V)t:

- (1) The glass corrosion rates are also affected by the solution pH value. A simple correlation variables such as (SA/V)t cannot be used to quantitatively relate tests run at different SA/V unless the effects of the pH are taken into account. While the solution pH affects the rates of reactions to release most glass components, most important is the increased release of aluminum at pH values below about pH 5 and the increased release of silicon at pH values above about pH 9. Solutions in tests conducted at higher SA/V generally attain higher pH values than tests conducted at lower SA/V. The higher pH values accelerate the corrosion rate, so that the glass reacts faster in tests at higher SA/V than in tests at lower SA/V. Thus, more glass will have reacted and more glass components will have been released into solution in a tests at a higher SA/V than in a test at a lower SA/V conducted for the same product of (SA/V)t. This is observed in tests with high-level waste glasses, where tests at high SA/V consistently have higher solution concentrations of soluble glass components than tests at low SA/V at common (SA/V)t. Higher concentrations of some glass components, including silicon and boron, can occur due to dissociation at high pH values.
- (2) The alteration of the glass surface will not scale as (SA/V)t due to the effects of different solution chemistries and due to the kinetics of secondary phase formation. Thicker layers are formed in tests at low SA/V than at high SA/V

when a glass dissolves nonstoichiometrically, so a larger volume of alteration phases is formed in tests at lower SA/V. While tests conducted at high SA/V are interpreted to represent advanced stages of corrosion, it must be recognized that the alteration phases form in a leachate solution having a higher pH than those formed at lower SA/V. The solution pH strongly affect the stabilities of most secondary phases. For example, the solubility of analcime, a zeolite commonly formed during corrosion of high-level waste glasses, is given as $K_{sp} = [Na^+][Al^{3+}]/[H^+]^4$. Thus, the formation of analcime is expected to occur after less glass has corroded in tests at high SA/V which attain a higher pH value than in tests at lower SA/V which attain lower pH values.

8.5 Flow

The flow rate used in dynamic tests also affects the concentration of silicic acid at the glass surface. The corrosion rate will be proportional to the flow rate at intermediate flow rates, but a maximum corrosion rate is attained at high flow rates. At low flow rates, the corrosion rate is limited by the accumulation of glass corrosion products in the fluid and the resulting low reaction affinity. The glass corrosion behavior under very low flow conditions approaches that in a static test. Tests at higher flow rates are generally used to measure the forward reaction rate. Tests using pH buffered leachants that are performed at high flow rates can be used to measure the forward reaction rate as a function of pH. These tests are also well suited to measuring the effects of glass composition on the corrosion behavior, since they minimize the effects of reaction products on the continued glass corrosion.

8.6 Remaining Issues

Based on this review of the literature regarding the effects of SA/V on glass corrosion, several key issues identified in the initial literature review that impact waste management remain unresolved. These include:

- How can tests at different SA/V be related?

Tests have shown (SA/V)^t scaling of solution concentrations to be affected by the solution pH. Current models of the glass corrosion rate account for the effects of the pH and may be used to improve the scaling approach, although this remains to be done.

- Can tests at high SA/V be used to characterize advanced stages of corrosion with regard to alteration of the glass?

Tests conducted at high SA/V attain saturated solution conditions after shorter reaction times than tests conducted at lower SA/V, but also attain higher solution pH values. The higher solution pH values will likely influence the formation of secondary phases. The effects of solution chemistry on secondary phases formation can be predicted using solution equilibration computer codes.

- Are tests conducted using monolithic and crushed glass samples at the same SA/V equivalent?

Several investigations have compared the corrosion rates of monolithic and crushed glass samples. The inability to accurately measure the effective surface area of crushed glass samples limits the direct comparison of test data. Geometric approximation based on sieve sizes are more realistic than surface areas measured using adsorption techniques. It is generally assumed that the corrosion rates of monolithic and crushed glass are the same, and comparison of

solution concentrations generated by monoliths and crushed glass samples in tests maintaining dilute leachate solutions is often used to determine the effective surface area of the crushed glass. Comparison of tests at different estimated SA/V using the same size fraction of crushed glass is probably more reliable than comparison of tests using different size fractions to attain different SA/V.

- How can the loss of surface area be accounted for in long-term tests with crushed glass?

The reduction in the surface area available for reaction is currently not accounted for when calculating the corrosion rate. An analytical method to correct the calculated normalized mass loss and normalized corrosion rate for changes in the surface area of crushed glass is needed.

- Does the SA/V used in a test affect the disposition of released radionuclides?

Very few tests have been conducted to assess the effects of released glass matrix components on the disposition of radionuclides. Most of these have been conducted at low SA/V or under flowing conditions that are not relevant to repository disposal conditions. Tests at high SA/V are needed to measure the effects of the higher solution pH, secondary phase formation, and colloid formation.

- Which secondary phases control the solubility of released radionuclides?

Little is known regarding the phases controlling the solubilities of radionuclides in the presence of glass corrosion products. Determination of the solubility controlling phases is complicated by difficulties associated with work with radioactive materials, analytical limitations for detecting low concentrations of some elements and the low solubilities of some radionuclides (Np, Pu), difficulties in isolating the small phases that form.

- What is the effect of long corrosion times in tests at high SA/V?

Tests conducted at high SA/V show deviations from the trends observed in tests at lower SA/V, such as resumption of high corrosion rates after secondary phase formation. It needs to be determined if the high corrosion rate is a transient phenomenon or if it represents the long-term corrosion rate. Tests in which a high corrosion rate is observed need to be conducted to determine which rate should be used in the corrosion model.

- What process controls the glass corrosion rate under silica saturation prior to secondary phase formation?

Prior to secondary phase formation, the corrosion slows to a negligible rate as the solution approaches apparent saturation with respect to the glass phase. However, the glass is metastable and continues to corrode, even under apparent saturated conditions. Because the glass may continue to corrode under these conditions for very long times prior to secondary phase formation, the process controlling the corrosion rate must be understood. Experimental measurement of the rate is difficult, due to its low value and the relatively large uncertainties of experimental measurements. Of course, the resumption of higher corrosion rates after secondary phases form will overwhelm the importance of this process to continued glass corrosion.

- Which SA/V ratios are relevant to repository storage?

The SA/V representative of repository storage conditions will vary with time as water may slowly reenter the repository and contact the glass. (In fact, corrosion of glass in the repository and laboratory tests at various SA/V are readily related through the effects of the solution chemistry.) Initial contact will likely occur between glass and condensed water vapor, with small volumes of liquid water eventually collecting on the glass. These conditions are represented by an initially extremely high then slowly decreasing SA/V. Hence, the conditions in the repository are not well represented by commonly performed laboratory tests, which exposed the glass to large volumes of water throughout the test period. (An exception is the Unsaturated Test Method developed at ANL.) These tests are conducted to characterize the corrosion behavior of the glass, but do not directly simulate glass corrosion under service conditions. Glass corrosion models must be correctly integrate both laboratory test results and site characterization results to model glass corrosion under service conditions. Other tests, such as the Unsaturated Test Method, must be developed to simulate changes in environmental conditions, such as the effective SA/V, to verify model predictions.

- What other repository-relevant conditions will impact the glass corrosion behavior, the solution chemistry, and the disposition of radionuclides and should be tested?

Conditions specific to a particular repository may impact the corrosion behavior of waste glasses. For a repository in an unsaturated environment, cycling of wet and dry conditions and episodic contact by relatively large volumes of water may significantly affect the performance of waste glasses. Periodic evaporation of water from the glass may result in cracking and peeling of alteration layers. These layers may be flushed from the glass at later times. Evaporation may also leave hygroscopic residues that will attract water entering at later times. Changing temperatures will affect the solubilities of most glass components and released radionuclides, which may result in the precipitation of secondary phases, formation of colloids, and other effects that will affect the corrosion of the glass and the transportability of radionuclides.

9.0 ANL TASK "RELATIONSHIP BETWEEN HIGH SA/V TESTS AND MCC-1"

The DOE sponsored an experimental task designed to clarify the effects of the SA/V ratio used in static leach tests on the corrosion mechanism and corrosion rate of borosilicate waste glasses relevant to the DWPF, and to assess the effects of the SA/V on the release on disposition of important radionuclides. The task was designed to include MCC-1 tests at SA/V of 10 m^{-1} and PCT tests at SA/V of 2000 m^{-1} , plus tests following the MCC-1 and PCT protocols at SA/V of 340 and $20,000 \text{ m}^{-1}$. Both solutions and corroded solids were analyzed to completely characterize the reacted system. Although several long-term tests remain in progress, enough tests have been completed to address the issues identified regarding the effects of the SA/V used in a static leach test on the solution chemistry, glass alteration, and disposition of actinide components. The results of this task have been published as data has become available [64-67, 112, 276, 277]. Some results have also been discussed in earlier sections of this report. The technical approach and the data collected to date are summarized in this chapter. Spread-sheets containing all available data are also included in Appendix D. Tests being conducted for long time periods are still in progress, and a report describing the task and all test results will be issued upon completion of the task.

9.1 Technical Approach

Frit material for preparing SRL 131 and SRL 202 glasses was supplied by the Savannah River Technology Center and doped with ^{99}Tc , ^{238}U , ^{237}Np , ^{239}Pu , and ^{241}Am at ANL. Glasses doped with the full compliment of radionuclides are referred to as SRL 131A and SRL 202A. Another glass was prepared by doping SRL 202 with natural uranium only. This glass is referred to as SRL 202U. The SRL 202 composition is the current "Design-Basis" glass composition for blended Savannah River waste [278]. The SRL 131 composition was an earlier reference composition known to be less durable than the SRL 202 composition. It is similar to the "Purex" glass, which was a possible "worst-case" composition [278] prior to specification of the "Environmental Assessment" glass for the DWPF (SRL EA) [101]. It was tested to demonstrate the effects of SA/V on a glass having a relatively poor durability, and to assure that the corrosion would advance to a large extent within the testing time. In addition, both the MCC-1 and PCT tests were designed to distinguish the relative durabilities of glasses having different compositions. Testing two glasses provides insight regarding the ability of different tests to distinguish the long-term corrosion behavior. The SRL 131A and SRL 202A compositions are given in Appendix B. The SRL 202U composition is identical to the SRL 202A composition, except for the absence of Tc, Np, Pu, and Am.

Both monolithic disk samples and crushed glass samples were prepared for testing. The faces and circumference edges of the monolithic disks were ground to a 600-grit finish for use in MCC-1 type tests at SA/V ratios of about 10 and 340 m^{-1} . All surfaces on the monoliths were ground to the same finish to assure uniformity in the surfaces of all samples to minimize the effects of the surface finish on corrosion (see Section 2.7). The crushed glass was sieved to isolate the -100+200 mesh fraction and washed for use in PCT type tests at SA/V ratios of about 2000 and $20,000 \text{ m}^{-1}$. Tests with SRL 131A and SRL 202A glass were performed in type 304L stainless steel vessels having a volume of approximately 22 mL. Tests with SRL 202U glass were performed in PFA Teflon vessels having a similar volume to provide data free from any effects of stainless steel for use in computer modeling. All tests were conducted at $90 \pm 2^\circ\text{C}$ in either a constant-temperature oven (tests in steel vessels) or water bath (tests in Teflon vessels). Teflon vessels were placed in a water bath such that the vessel bottoms were submerged but the vessel tops were not submerged to prevent possible leakage of water from the water bath into the

vessel. A water bath was used to minimize loss of leachate from the vessels over long test periods, and to reduce the diffusion of carbon dioxide into the vessels. Other tests have shown carbon dioxide to penetrate teflon vessels and affect the solution pH [102,279].

The leachant was a tuff groundwater solution that was prepared by reacting pulverized caliche-free tuff rock with actual groundwater from well J-13 on the Nevada Test Site at 90°C for about 28 days. This was done to simulate the interaction of groundwater with tuff near the repository where the rock temperature is anticipated to be near 90°C, since the solution concentrations of ions in the groundwater will change as the temperature increases from 25 to 90°C. The resulting groundwater solution was then cooled to room temperature and filtered through 0.1 μm filters to remove any suspended tuff particles. The resulting solution is referred to as EJ-13 water. The composition of the J-13 groundwater and the EJ-13 water are given in Table 7. The major changes in the J-13 chemistry due to reaction with tuff are an increase in the sodium and silicon concentrations, an increase in the pH, and a decrease in the alkaline earth concentrations resulting from their retrograde solubilities. The actual groundwater that might eventually contact the waste glass in a repository will be affected by interaction with the waste package components and by radiolysis, as well as by interaction with the tuff. These interactions may affect the pH and all solute concentrations. Because the engineered barrier system has not yet been specified, only the effects of interactions with the tuff were considered in this Task. The effects of radiation on the corrosion behavior of high-level waste glasses have been measured previously [95,113,114,280-283]. SRL 202U glass was tested in both EJ-13 and in deionized water to assess effects of the leachant composition on the corrosion.

MCC-1 type tests were conducted by placing either one or four monolithic disks in either 16 or 2 mL of leachant to achieve SA/V ratios of 10 and 340 m^{-1} , respectively. PCT type tests were conducted by adding either 1 or 5 g of crushed glass to either 10 or 5 mL of leachant to achieve SA/V ratios of 2000 or 20,000 m^{-1} , respectively. The surface area of the grains was approximated by assuming they were spheres having a diameter equal to the average mesh opening (which was about 112 μm). The surface area of the monolithic disks was measured geometrically. All tests were conducted in duplicate at $90 \pm 2^\circ\text{C}$. Test durations were selected to provide both a temporal description of the corrosion of both glass types at all SA/V. Tests were also scheduled to provide results at different SA/V but common products of (SA/V)t. This was done to compare solution and solids analyses and to assess the usefulness of (SA/V)t as a measure of the corrosion progress. The test matrix is summarized in Table 8, which shows the overlap in (SA/V)t of tests performed at different SA/V.

Table 7. Compositions of Tuff Groundwater from Well J-13^a and EJ-13 Water

	J-13	EJ-13		J-13	EJ-13
Al	0.37	1.1	HCO ₃ ⁻	N/A	120
B	0.15	0.16	F ⁻	N/A	3
Ca	11.7	5.1	Cl ⁻	N/A	9
K	6.6	7.3	NO ₃ ⁻	10.1	16
Li	0.035	0.05	SO ₄ ²⁻	N/A	35
Mg	1.8	0.39			
Na	42	54			
Si	28	46	pH	6.9	8.2

^aFrom [195].

Table 8. Summary of Test Matrix for ANL Task

(SA/V)t (days/m)	Test Length (days)			
	10 m ^{-1a,b}	340 m ^{-1c}	2000 m ^{-1a,b,c}	20,000 m ^{-1a,b,c}
70	7			
280	28			
940	94			
2380	238			
4760		14		
6000	600		3	
9520		28		
14,000	1400		7	
19,040		56		
28,000	(2800) ^d		14	
30,940		91		
60,000			30	3
61,200		180		
122,400		360		
140,000			70	7
183,000		540		
224,800		720		
280,000			140	14
560,000			280	28
1,120,000			560	56
1,960,000			980	98
3,640,000			(1820)	182
7,280,000				364
14,560,000				728
20,000,000				1000
TBD ^e			TBD	TBD

^aDuplicate tests performed with SRL 202A glass.

^bDuplicate tests performed with SRL 131A glass

^cDuplicate tests performed with SRL 202U glass.

^dTime in parentheses indicate tests in progress.

^eTBD = test time "to be determined."

After the predetermined test duration, the vessel was removed from the oven and opened. The leachate solution and reacted glass were removed for analysis. The leachates were removed while still hot to avoid thermal effects, such as precipitation of solids or flocculation of colloids. To isolate suspended solids, some of the leachate solution was filtered through a holey carbon grid used in a transmission electron microscope (TEM). In this procedure, a small volume of leachate is placed on a holey carbon grid and then wicked through the grid using alcohol. Suspended solids are filtered from the solution as it passes through the grid and remain on top of the grid where they can be analyzed using the TEM. An aliquot of the leachate solution was also removed for actinide analysis. The remaining solution was then filtered through a preheated 0.45- μm cartridge filter. A portion of this solution was further filtered through an Amicon model CF50A filter, which is assumed to have an effective filter size of about 6 nm. Solutions were analyzed for pH (using a combination electrode), anions (by ion chromatography), cations [by inductively coupled plasma-atomic emission or inductively coupled plasma-mass spectrometry (ICP-MS)], and actinides (by alpha spectrometry). Additional aliquots of the unfiltered leachate solutions or the solutions produced by filtering through 0.45- μm filters were taken for analysis of the radionuclides using ICP-MS. These aliquots were evaporated to dryness then redissolved and acidified with nitric acid for analysis. The measured pH values are expected to be affected by the temperature (they were measured at room temperature) and the high ionic strengths of the leachate solutions. Nevertheless, the pH values provide a qualitative measure of the effects of the SA/V ratio and reaction time on the solution chemistry.

The reacted glass was gently rinsed with deionized water then removed from the vessel and allowed to dry. The vessel was then filled with a nitric acid solution and placed in a 90°C oven overnight to dissolve any actinides that had become fixed to the stainless steel surface. An aliquot of this acid soak solution was stippled onto stainless steel planchets and analyzed by alpha spectroscopy. Some of the more saline solutions were electroplated onto planchets and then analyzed. The solution results are tabulated in spreadsheets included in Appendix D.

Solids were analyzed by using an optical microscope, scanning electron microscope (SEM), and transmission electron microscope (TEM) to characterize the glass alteration and identify secondary phases formed during glass corrosion. Reacted glass was analyzed by using analytical electron microscopy (AEM), which combines high-resolution TEM with energy-dispersive X-ray spectroscopy (EDS). Representative samples of the reacted glass were fixed in epoxy, and thin-sections were prepared by using an ultramicrotome [284]. Grains of powdered glass were fixed in resin directly, while sample chips were removed from reacted monoliths using a diamond knife. Secondary phases found distributed among the reacted grains were isolated and analyzed separately by using optical microscopy, SEM, TEM, and X-ray diffraction for phase identification.

9.2 Observed Effects of the SA/V on the Leachate Chemistry

9.2.1 Leachate pH

The aliquots of the leachates from tests at 10 and 340 m^{-1} and the filtrates from tests at 2000 and 20,000 m^{-1} taken for pH analysis were allowed to cool to room temperature prior to analysis. The pH measured at room temperature is expected to be about one pH unit higher than the pH at the reaction temperature because of the difference in the ionization constant of water at 90 and 25°C. The measured pH values from duplicate tests with SRL 202A, SRL 202U, and SRL 131A reacted in EJ-13 water or in deionized water are plotted versus the reaction time in Fig. 52a-c. Differences in duplicate tests with crushed glass were generally small, but large differences were measured in some tests with monolithic samples, both at 10 and 340 m^{-1} . (The solution concentrations of glass components also differ significantly in these tests, as shown below.) The measured pH values in tests with all glasses are higher than the pH

Fig. 52. Measured pH Values in ANL Tests with (a) SRL 202A Glass, (b) SRL 202U Glass, and (c) SRL 131A Glass in Tests at 90°C at (●) 10, (▲) 340, (■) 2000, and (◆) 20,000 m⁻¹ in EJ-13 Water and at (Δ) 340, (□) 2000, and (◇) 20,000 m⁻¹ in Deionized Water

value of the initial leachant, which was 8.2 for the EJ-13 leachant and about 5.5 for the deionized water, at all reaction times. Higher pH values were attained in tests conducted at higher SA/V for all glasses and at all reaction times. The pH is increased only about one pH unit above the initial leachant pH in tests at 10 m^{-1} , even after 1400 days. Tests conducted with SRL 202U glass in deionized water had slightly higher pH values than tests conducted in EJ-13 water in tests at $20,000 \text{ m}^{-1}$ (larger differences were measured after 182 and 364 days), but slightly lower pH values than tests conducted in EJ-13 water in tests at 340 m^{-1} at each time tested. Tests with SRL 131A generally had higher leachate pH values than tests with SRL 202A conducted at the same SA/V and for the same reaction time.

9.2.2 Glass Matrix Components

The leachate and filtrate solutions were analyzed by using inductively coupled plasma-atomic emission spectroscopy, inductively coupled plasma-mass spectroscopy, and fluorescence spectroscopy. Figures 53, 54, and 55 show the normalized elemental mass losses of several glass components from SRL 202A and SRL 131A glass at 10, 2000, and $20,000 \text{ m}^{-1}$ and from SRL 202U glass at 340 and $20,000 \text{ m}^{-1}$ in tests conducted in EJ-13 tuff groundwater solution, and from SRL 202U in tests in deionized water. The average values of duplicate tests are plotted. The data for each test is included in Table 2 of Appendix D. Difference in the results of duplicate tests are generally within the size of the symbol for tests with crushed glass, but slightly larger for tests with monolithic samples. The difference in the error ranges of tests with crushed glass or monolithic glass samples is due in large part to the normalization of the solution concentrations by dividing them by the SA/V of the test. Similar analytical errors in the measured concentrations are more significant when divided by 10 or 340 than they are when divided by 2000 or 20,000. The normalized elemental mass losses were computed directly from the leachate results for tests with monolith samples, and from the results of solution filtered through $0.45\text{-}\mu\text{m}$ filters for tests with crushed glass.

The release of different glass components from SRL 202A, SRL 131A, and SRL 202U in tests at different SA/V can be compared directly using the normalized mass loss of each element. Figures 53, 54, and 55 show the release of Li, Na, K, B, Si, and U into solution to be nonstoichiometric (with respect to the glass composition) for all glasses and at all SA/V tested. (Stoichiometric release would result in the data points for all elements falling on the same curve.) The general order of elemental release is $\text{Li} > \text{Na} > \text{B} > \text{K} > \text{Si} > \text{U}$, though at 10 m^{-1} the sodium release is greater than the lithium release from both SRL 202A and SRL 131A glass. The alkali metal and boron concentrations increase at high rates initially, but the release slows as the reaction progresses at intermediate reaction times. As the reaction continues for longer times under several test conditions, the boron and alkali metal release rates suddenly increase and become nearly linear in time. This change occurs in tests with SRL 202A, SRL 202U, and SRL 131A glasses at $20,000 \text{ m}^{-1}$ beyond about 182 days, and in tests with SRL 131A glass at 2000 m^{-1} beyond about 140 days. The time at which the corrosion accelerated can be estimated by extrapolating the constant rate observed in tests conducted for the longest times back to where it intersects the rate curve determined by tests run for shorter times. The acceleration is estimated to occur shortly beyond 182 days in tests with SRL 202A glass at 2000 m^{-1} , after about 182 days in tests with SRL 131A glass at 2000 m^{-1} , and after about 90 days in tests with SRL 131A glass at 2000 m^{-1} . Data are not yet available to determine when tests with SRL 202U glass at 2000 m^{-1} are accelerated, although the similarity between tests with SRL 202U and SRL 202A glasses suggest that they, too, were accelerated shortly after about 182 days. Other tests with SRL 202U glass are in progress that are expected to show the same corrosion rate as tests run for 364 days.

Fig. 53. Measured Cation Concentrations in ANL Tests with SRL 202A Glass Conducted at 90°C at (a) 10, (b) 2000, (c) 20,000, and (d) 20,000 m⁻¹: (○) Si, (◆) K, (●) B, (▲) Na, (■) Li, and (□) U

Fig. 54. Measured Cation Concentrations in ANL Tests with SRL 131A Glass Conducted at $90^\circ C$ at (a) 10, (b) 2000, and (c) 20,000 m^{-1} : (○) Si, (◆) K, (●) B, (▲) Na, (■) Li, and (□) U

Fig. 55.

Measured Cation Concentrations in ANL Tests with SRL 202U Glass Conducted at 90°C at (a) 340 and (b) 20,000 m^{-1} in EJ-13 and at (c) 340, (d) 2000, and (e) 20,000 m^{-1} in Deionized Water: (○) Si, (◆) K, (●) B, (▲) Na, (■) Li, and (□) U

When the corrosion is accelerated, the boron release to solution becomes greater than the release of the alkali metals. The normalized silicon mass loss is only slightly increased when boron and the alkali metals show accelerated release in tests at $20,000 \text{ m}^{-1}$ (refer to Figs. 53d, 54d, and 55c). The normalized uranium mass losses remain well below the normalized silicon mass loss in all tests.

Both suspended and dissolved material were included in the calculation of the normalized mass loss to represent the total amount of glass that had dissolved. Leachate solutions were further filtered through 6-nm filters to remove suspended material and measure the amounts of various glass components that were dissolved. Table 9 shows the measured concentrations of aluminum, iron, silicon, and boron in the 0.45- μm and 6-nm filtrates of tests with SRL 202U glass reacted at $20,000 \text{ m}^{-1}$. The aluminum and iron concentrations in the 0.4- μm filtrates increase with the reaction times through 14 days, but then decrease to low values at longer times. Filtration through 6-nm filters removed most of the aluminum and iron from solution. The silicon and boron concentrations increase with the reaction time at all times tested. Further filtration through 6-nm filters did not significantly reduce these concentrations; this result indicates that these components are dissolved. Large increases in the boron and silicon concentrations are measured in both the 0.45- μm and 6-nm filter solutions after 364 days of reaction. Similar trends were seen upon filtration of the leachates of tests with SRL 202U, SRL 202A, and SRL 131A glass at all SA/V ratios tested. That is, the concentrations of soluble alkali metals, boron, and silicon are not reduced by filtration (the reduction in the silicon concentration is less than the assumed analytical error range of about 15%), and aluminum, iron, and other components with low solubilities are further removed from the leachate by 6 nm filters.

Table 9. Results of Filtering Leachate Solutions from Tests with SRL 202U Glass through 6 nm Filters

Test No.	Time (days)	pH	Al, ppm		Fe, ppm		Si, ppm		B, ppm	
			0.45 μm	6 nm	0.45 μm	6 nm	0.45 μm	6 nm	0.45 μm	6 nm
1	3	10.86	5.27	1.26	25.9	0.38	279	243	75.9	73.5
2	3	10.89	5.42	1.25	26.8	0.38	283	243	77.0	75.0
3	7	11.03	9.18	1.31	44.2	0.30	351	267	93.8	85.9
4	7	11.01	9.33	-- ^a	38.2	--	338	--	92.8	--
5	14	11.23	11.5	--	51.7	--	411	--	111	--
6	14	11.28	12.1	<2.5 ^b	55.0	0.75	417	323	111	107
7	28	11.37	6.05	1.80	18.5	0.28	361	360	119	127
8	28	11.38	3.85	1.93	8.37	<0.19	350	362	121	128
9	56	11.37	1.55	--	0.50	--	379	--	148	--
10	56	11.41	1.52	--	0.37	--	385	--	149	--
11	91	11.4	<1.3	<1.1	0.66	<0.23	429	440	177	182
12	91	11.38	2.33	1.70	0.24	<0.23	436	446	180	181
13	182	11.63	1.12	<1.64	0.173	<0.328	562	532	246	230
14	182	11.66	1.21	<1.72	0.173	<0.345	567	539	244	229
15	364	11.75	<2.86	1.3	1.43	0.602	1060	971	3490	3400

^aSolution not analyzed.

^bBelow detection limit, value given is adjusted for dilution.

The results plotted in Figs. 53, 54, and 55 showed that the amounts of glass components released into solution are also affected by the SA/V ratio of the test. In plotting the normalized mass losses, the measured solution concentrations are divided by the SA/V ratio to account for differences in dilution at different SA/V. If the reaction rates are the same in tests at different SA/V, then the normalized released rates should also be the same. Comparing the results at different SA/V ratios in Figs. 53, 54, and 55, the normalized release rates of all elements plotted are different in tests at different SA/V. For example, Fig. 56 shows the normalized boron mass losses from SRL 202A and SRL 202U glasses in tests at 10, 340, 2000, and 20,000 m^{-1} on the same plot. (Figures 56a and 56b show the same results on different scales.) At short reaction times (<200 days), the normalized mass loss of boron decreases with the SA/V of the test. The long-term tests at different SA/V show very different reaction trends. Tests at 10 and 2000 m^{-1} release boron at a rate that decreases as the reaction progresses, with tests at 2000 m^{-1} reaching a low rate after short reaction times. The release rate of boron is lowest at 20,000 m^{-1} at short reaction times, but the reaction is accelerated after about 182 days to a high rate that remains constant with time thereafter. This implies that the glass reaction rate and/or reaction mechanism is affected differently by the different solution chemistries attained in tests at different SA/V.

Instead of plotting the normalized elemental mass loss against time, as in Fig. 56, the measured concentrations can be plotted against the product of the reaction time and the SA/V of the test. The product $(\text{SA/V})t$ has been suggested as a single parameter that can be used to measure the extent of glass reaction, meaning that tests at different SA/V but equivalent $(\text{SA/V})t$ should yield the same extent of glass reaction [72,285,286]. The boron data from replicate tests with SRL 202A, SRL 131A, and SRL 202U glasses are plotted against $(\text{SA/V})t$ in Fig. 57. Tests at 2000 and 20,000 m^{-1} performed for equivalent $(\text{SA/V})t$ clearly do not generate equivalent solution compositions in tests with either SRL 202A, SRL 131A, or SRL 202U glass. Tests performed at higher SA/V but shorter reaction times consistently have greater boron releases. Tests with SRL 131A show a larger difference than tests with SRL 202A. Although duplicate tests at 10 m^{-1} show significant scatter, some tests at 10 m^{-1} have boron concentrations similar to those in tests at 2000 m^{-1} at equivalent $(\text{SA/V})t$.

The results of these tests show that the reaction is characterized by an initially high forward rate, an intermediate stage where a minimum rate is approached, and a long-term rate that may be greater than the minimum rate. The progression of the reaction from the initial rate to the intermediate rate to the long-term rate is controlled primarily by the leachate solution composition. Under dilute solution compositions, the observed rate will likely depend on the glass composition and the solution pH. The leachate pH will generally increase as the glass reacts to release alkali metals, and the glass reaction rate will increase with the pH. However, the rate decreases as the leachate becomes more concentrated in glass components (primarily in silicic acid). The rate-decreasing effects of the buildup of solution concentrations overwhelms the rate-increasing effect of the higher pH values, and a minimum reaction rate is approached as the leachate approaches saturation. The glass may react at the minimum rate for very long times. The reaction may eventually be accelerated if secondary phases form to affect the solution composition. As the assemblage of secondary phases changes as the reaction progresses, the reaction rate will likely also change. A characteristic "final dissolution rate" may not exist because of the influence of the pH and the secondary phases on the reaction rate.

Fig. 56. Normalized Boron Mass Loss from Glasses vs. Reaction Time in Tests Conducted at ANL: ($\blacklozenge$) 10, ($\blacksquare$) 2000, and ($\bullet$) 20,000 m^{-1} in Tests with SRL 202A in EJ-13 Water; ($\blacktriangle$) 340 and ($\oplus$) 20,000 m^{-1} in Tests with SRL 202U in EJ-13 Water; ($\triangle$) 340, ($\square$) 2000, and ($\circ$) 20,000 m^{-1} in Tests with SRL 202U in Deionized Water. (a) All data plotted and (b) data for $\text{NL(B)} < 3 \text{ g/m}^2$.

Fig. 57. Logarithm of Measured Boron Concentration vs. Logarithm of $(\text{SA}/V)t$ for Tests with (a) SRL 131A, (b) SRL 202A, and (c) SRL 202U Glasses Conducted at SA/V of: (●) 10, (▲) 340, (■) 2000, and (◆) 20,000 m^{-1} in EJ-13 Water, and (Δ) 340, (□) 2000, and (◇) 20,000 m^{-1} in Deionized Water

Tests conducted at different SA/V ratios generate very different solution chemistries which affect the glass corrosion rate and will affect the disposition of radioelements. The pH and anion concentrations are expected to affect the behavior of released actinides. The pH values and anion concentrations measured in the leachate solutions of tests with SRL 131A and SRL 202A glasses at 2000 and 20,000 m⁻¹ are listed in Table 10. High leachate pH values are reached after only a few days tests with SRL 131A and SRL 202A because of alkali release as the glasses corrode. Different pH values are attained in tests at different SA/V ratios because of the different volumes of solution available to dilute the hydroxide and other corrosion products.

Quantification of fluoride in some samples was complicated by the overlap of the fluoride and formate (HCO₂⁻) peaks in the ion exchange column. Most tests showed the fluoride concentration to be approximately the same as its initial concentration in the leachant, about 3 ppm. Fluoride levels were assumed to remain near 3 ppm for all tests, and the formate levels were calculated after subtracting this amount of fluoride from overlapped peaks. At these low concentrations, neither fluoride nor formate is expected to affect the solubilities of the actinides. Relatively high concentrations of oxalate (C₂O₄²⁻) were measured in tests with SRL 202A glass, but most tests with SRL 131A glass had oxalate concentrations below the detection limit of 0.5 ppm. This suggests that the organics were released from the glasses as they corroded, and that the SRL 202 frit used to produce SRL 202A and SRL 202U glasses had more carbon than the SRL 131 frit used to produce SRL 131A glass.

The chloride content remained near its concentration in the EJ-13 leachant for most tests at 2000 m⁻¹, but increased to more than 200 ppm in tests with both glasses at 20,000 m⁻¹. Nitrate concentration in all tests remained near the leachant level of 16 ppm. Both chloride and nitrate are weak complexants and these levels are not expected to significantly increase the actinide solubilities. The hydrogen phosphate level increased in all tests with both glasses: tests with SRL 131A glass attain concentrations exceeding 400 ppm, and tests with SRL 202A glass attain concentrations exceeding 100 ppm after two years at 20,000 m⁻¹. Very high sulfate concentrations were also measured in tests with SRL 131A glass at 20,000 m⁻¹ at all reaction times and with SRL 202A glass in tests beyond two years. Both plutonium and americium form phosphates having lower solubilities than the oxides, and the release of hydrogen phosphate and sulfate is expected to lower the solubilities of plutonium and americium in these leachates.

The organic and inorganic carbon contents of the leachates were also measured, and the results are included in Table 10 as the total organic carbon (TOC) and total inorganic carbon (TIC) concentrations. The EJ-13 leachant contains about 3 ppm organic carbon and 22 ppm inorganic carbon (as the bicarbonate ion). Corrosion of the glass increases both the organic and inorganic carbon levels. In general, tests with SRL 202A glass at 2000 m⁻¹ attained slightly higher inorganic carbon levels than tests with SRL 131A glass at the same SA/V ratio. Tests at 20,000 m⁻¹ had greater amounts of total carbon than tests at 2000 m⁻¹, a result that is consistent with carbon being released from the glass. Also, tests at 20,000 m⁻¹ show a decrease in the inorganic content after 364 days or longer. This may be due to the formation of various carbonate secondary phases (see below). Formation of such phases will decrease the amounts of carbonate available to complex actinides.

Table 10. Anion Concentrations in Tests with SRL 131 and SRL 202 Glasses^a

Time	pH	F ⁻	HCO ₂ ⁻	Cl ⁻	NO ₃ ⁻	HPO ₄ ²⁻	SO ₄ ²⁻	C ₂ O ₂ ²⁻	TOC	TIC
EJ-13	8.20	3	<0.5	9	16	<0.5	25	<0.5	3	22
SRL 202 at 2000 m ⁻¹										
14	9.77	3	<0.5	11	16	<0.5	26	<0.5	11	64
140	10.11	3	<0.5	11	11	6	25	2	13	38
560	10.42	4	<0.5	10	9	6	31	3	32	130
980	10.65	5	<0.5	18	22	10	68	5	27	130
SRL 202 at 20,000 m ⁻¹										
14	11.01	3	17	12	16	12	37	7	70	72
98	11.26	3 ^b	29 ^b	15	22	28	42	14	160	92
182	11.29	3 ^b	23 ^b	18	--	71	39	18	220	120
364	12.03	3 ^b	28 ^b	<10	17	121	95	17	80	83
736	11.98	3 ^b	30 ^b	215	26	103	293	56	98	40
SRL 131 at 2000 m ⁻¹										
14	10.45	3	<0.5	10	16	<0.5	27	<0.5	12	27
140	10.72	3	<0.5	12	11	<2	28	<0.5	14	34
560	11.46	3 ^b	<0.5 ^b	12	9	9	32	1	13	55
980	11.72	3 ^b	3 ^b	50	16	<40	170	<40	24	71
SRL 131 at 20,000 m ⁻¹										
14	12.05	3	<0.5	66	21	66	160	<0.5	91	14
98	12.12	3 ^b	<0.5	90	20	99	190	<0.5	160	20
182	12.19	<3	<5	150	27	190	780	<0.5	29	68
364	12.18	c	c	210	12	244	1300	<1	100	8
729	12.03	<3	<5	240	18	460	1710	<40	120	4

^aUnits given as: S/V (m⁻¹), Time (days), anions (ppm), Total Organic Carbon (TOC, ppm), Total Inorganic Carbon (TIC, ppm).

^bFormate values estimated based on 3 ppm F⁻.

^cValues not reported.

Specific analysis of the formate and oxalate concentrations by ion chromatography shows relatively high formate levels in tests with SRL 202A glass at 2000 m⁻¹ but only small or undetectable levels in other tests. In many instances, the formate peak had to be resolved from the fluoride peak by assuming that only 3 ppm fluoride was present. Significant levels of oxalate were measured only in tests with SRL 202A and SRL 202U glasses at 2000 and 20,000 m⁻¹. (The oxalate peaks are fully resolved in the ion chromatograms. The presence of oxalate supports the values assigned to the formate levels in these same tests. That is, formate and oxalate levels appear to be correlated.) The measured formate and oxalate levels are consistent with the measured TOC concentrations, though the TOC concentrations are significantly higher. Although humic acids are known to complex actinides [269,287], the simple organics presence in these tests are not expected to significantly affect the solubilities of the transuranics.

9.2.3 Observed Effects of SA/V on the Behavior of Radionuclides

Of key interest in this Task is how the SA/V used in a static test affects the behavior of released radionuclides. Tests conducted in this Task were designed to address several questions regarding the behavior of radionuclides as the glass corrodes: Is the amount of radionuclides sensitive to the different solution chemistries attained under different SA/V? Does the presence of other glass components in the leachate affect the release, solubility, and stability of released radionuclides? Does the corrosion rate of the glass matrix affect the disposition of the radionuclides. Are the amounts of radionuclides in solution controlled by their solubilities? Is the disposition of the radionuclides affected by the formation of secondary phases? Is the behavior observed in short-term tests representative of the long-term behavior of radionuclides?

The amounts of technetium and actinides in the leachate solutions were measured using ICP-MS, and the calculated normalized mass losses based on the measured concentrations of technetium, uranium, and neptunium are compiled in Table 11. The normalized mass losses based on the boron release is included for comparison. Analyses were performed on the unfiltered leachates or the 0.45-μm filtrate solutions. The boron release represents the extent of glass corrosion. The results of the test having the highest boron release of the duplicate tests conducted at each SA/V and reaction time are tabulated. In most cases, the results of duplicate tests are within the expected analytical error (see Tables 2 and 4 in Appendix D). The normalized mass losses for tests conducted at 2000 or 20,000 m⁻¹ with both glasses are plotted against the reaction time in Figs. 58a-d. Lines are drawn to show the trends in the data, but do not represent analytical fits to the data.

The boron data show the glass to corrode at a continuously decreasing rate in tests with SRL 202A glass at 2000 m⁻¹. Technetium is released to solution stoichiometrically with boron, while uranium and neptunium are released to lesser extents than boron and technetium. In tests with SRL 202A glass at 2000 m⁻¹, boron is released at a low rate through 182 days, but is released at a high rate after 182 days. Technetium is released at a low rate similar to boron through 182 days, and at a higher rate after 182 days, but the release of technetium after 182 days is less than that of boron. Of course, data from tests run for longer times are needed to determine the long-term behavior of technetium. Analysis of these solutions is in progress. Uranium and neptunium are released at low rates at all times tested, and are not affected by the increased glass corrosion rate after 182 days.

Table 11. Normalized Mass Losses for Radionuclides

Time, days	NL(B)	NL(Tc)	NL(U)	NL(Np)
SRL 202A, 10 m ⁻¹				
7	0.69	2.7	0.045	0.67
28	0.75	<1a	0.04	<0.2
94	4.3	<1	0.18	<0.2
238	0.95	<1	0.11	<0.1
600	15	<1	0.29	0.33
SRL 202A, 2000 m ⁻¹				
3	0.096	0.056	0.046	0.049
7	0.19	0.12	0.11	0.099
14	0.27	0.19	0.11	0.14
30	0.41	0.28	0.077	0.14
140	0.57	0.44	0.095	0.20
280	0.67	0.87	0.12	_b
560	0.83	0.87	0.17	--
980	0.95	1.1	0.16	--
SRL 202A, 20,000 m ⁻¹				
3	0.13	0.074	0.028	0.025
7	0.17	0.095	0.039	0.037
14	0.20	0.12	0.046	0.041
28	0.24	0.17	0.036	0.031
56	0.31	0.26	0.019	0.021
98	0.36	0.29	0.013	0.014
182	0.57	1.5	0.010	0.072
364	5.2	2.7	0.0038	0.0041
484	12	--	0.0023	--
728	18	--	0.0013	--
SRL 131A, 10 m ⁻¹				
7	1.0	<0.8	0.16	0.59
28	1.8	3.2	0.19	2.6
94	5.2	<0.8	0.45	1.2
238	34	7.3	1.6	3.5
601	53	34	7.7	15
SRL 131A, 2000 m ⁻¹				
3	0.76	0.35	0.12	0.80
7	1.1	0.51	0.20	0.12
14	1.3	0.61	0.19	0.12
30	1.5	0.68	0.12	0.11
70	1.7	0.81	0.081	0.041
140	2.2	0.96	0.083	0.041
280	5.4	0.96	0.082	--
560	9.6	5.4	0.036	--
980	13	7.5	--	--
SRL 131A, 20,000 m ⁻¹				
3	2.6	1.3	0.0033	0.00081
7	4.8	2.5	0.0043	0.00096
14	6.0	3.3	0.0036	0.0011
28	7.5	4.5	0.0052	0.0013
56	9.7	5.4	0.0054	0.0012
98	9.5	9.7	0.0050	0.0017
182	11	8.5	0.0055	0.0041
364	20	12	0.0039	0.0016
552	23	--	0.0036	--
728	36	--	0.0068	--

aMaximum value based on analyzed radionuclide concentration below detection limit.

bAnalysis not yet performed.

Fig. 58. Normalized Mass Loss vs. Reaction Time for (a) Tests with SRL 202A Glass at 2000 m⁻¹, (b) Tests with SRL 202A Glass at 20,000 m⁻¹, (c) Tests with SRL 131A Glass at 2000 m⁻¹, and (d) Tests with SRL 131 A Glass at 20,000 m⁻¹: (○) B, (□) Tc, (◇) U, and (Δ) Np.

Tests with SRL 131A glass at 2000 m^{-1} show the glass to corrode at a high rate after about 140 days, as shown by the boron release. Technetium shows a similar constant release rate, but the normalized mass loss of technetium is about half that of boron. Uranium and neptunium are released to much lower extents than boron or technetium. The same behavior is seen in tests at $20,000\text{ m}^{-1}$, where boron and technetium are released at nearly constant rates, with boron being released a little faster than technetium, and uranium and neptunium are released at low rates at all times tested.

The behavior of technetium, uranium, and neptunium in the presence of glass corrosion products is of key interest due to their known high solubilities in oxidizing groundwaters. These tests show that technetium is released nearly congruently with boron as the glass corrodes, and that an increase in the glass corrosion rate due to secondary phase formation likely results in an increase in the release of technetium, although more analyses are needed to confirm this behavior in tests with SRL 202A at $20,000\text{ m}^{-1}$, it is clearly demonstrated in tests with SRL 131A at both 2000 and $20,000\text{ m}^{-1}$. The observation that $NL(B)$ is consistently higher than $NL(Tc)$ in tests with SRL 131A may be attributed to the large difference in the amounts of boron and technetium in the glass, although similar amounts are in SRL 202A. The thick alteration layers formed on SRL 131A and SRL 202A glasses after long reaction times may attenuate the release of technetium slightly.

Other analyses were performed to determine the disposition of the sparingly soluble actinide elements. The amounts of neptunium, plutonium, and americium that were suspended in solution (that fraction which passed through a $0.45\text{-}\mu\text{m}$ filter but did not pass through a 6-nm filter), dissolved in solution (that fraction which passed through a 6-nm filter), or became fixed to the stainless steel reaction vessel (the fraction in the acid soak solution) were measured using alpha spectroscopy. The results of these analyses in terms of the amounts suspended in solution, dissolved in solution, and fixed to the steel are presented in Table 12 for several, representative tests with SRL 202A and SRL 131A glasses under columns labeled "sus," "dis," and "ads." Actinides not found in these fractions are assumed to remain associated with the altered glass or to have settled out of solution during the test.

Although there is scatter in the results, several general trends appear in the distributions of the actinides between various fractions in tests conducted at different SA/V and for different time periods. On average, neptunium was evenly distributed between the suspended and dissolved fractions in tests with both glasses. Neptunium was fixed to the vessel in tests at $20,000\text{ m}^{-1}$ but not in tests at 2000 m^{-1} . About four times more plutonium was present in the suspended fraction than in the dissolved fraction in tests at 2000 m^{-1} , but plutonium was found in only small amounts in the suspended fractions in tests at $20,000\text{ m}^{-1}$. Plutonium was preferentially fixed to the steel vessel in all tests. Only small amounts of dissolved americium are present in all tests. Similar amounts of americium are suspended in the leachate and fixed to the vessel in tests at 2000 m^{-1} , but americium was only detected fixed to the vessel in tests at $20,000\text{ m}^{-1}$.

Between 75 and 90% of the released uranium was dissolved and the balance was suspended in all tests. The uranium distribution between the dissolved and suspended phases was similar for all time periods beyond a few days. Uranium was not found fixed to the stainless steel.

Table 12. Mass of Actinides in Various Fractions. Reaction time in days; fractions that are suspended (sus), dissolved (dis), and fixed to the steel vessel (ads) in ng

Time, d	Np, ng			Pu, ng			Am, ng		
	sus	dis	ads	sus	dis	ads	sus	dis	ads
SRL 202 at 2000 m ⁻¹									
14	420	96	51	34	0.75	8.6	0.87	0.017	0.24
30	440	100	56	50	1.4	23	1.4	0.029	0.51
70	110	290	15	60	12	30	1.3	0.23	1.2
140	0	670	0	68	38	33	1.6	0.61	1.3
280	540	160	0	140	3.1	170	2.1	0.074	3.6
560	0	550	0	130	34	110	3.2	0.79	2.6
980	87	170	0	74	19	150	1.8	0.39	3.0
SRL 202 at 20,000 m ⁻¹									
14	3000	180	120	180	5.5	17	3.7	0.12	0.59
28	380	150	160	65	11	50	1.4	0.28	1.9
56	180	68	270	10	0.97	38	0.22	0.02	1.6
98	75	89	290	1.3	0.28	52	0.026	0.002	1.8
182	21	76	360	0	1.7	58	0.02	0.003	2.1
364	28	6.8	1100	0.11	0	88	0.01	0	2.6
504	0.43	1.5	860	0	0.01	60	0	0	2.7
728	0	69	660	0	0.37	203	0	0.004	6.9
SRL 131 at 2000 m ⁻¹									
14	221	125	580	5.4	0.65	4.5	0.07	0.004	0.097
30	87	130	170	4.2	1.5	41	0.032	0.007	0.35
70	64	230	0	13	4.7	46	0.20	0.036	0.93
140	127	180	0	12	7.0	97	0.14	0.088	2.3
280	41	35	0	2.4	11	92	0.022	0.009	1.0
560	18	17	0	0.25	0.22	109	0.013	0	1.7
980	46	0	0	0	0	122	0	0	1.9
SRL 131 at 20,000 m ⁻¹									
14	2.4	1.9	140	0.78	0.002	15	8.6	0	0.48
28	3.0	1.5	260	0.003	0.001	53	0	0	1.4
56	75	2.2	230	4.6	0.003	32	94	0	1.3
98	1.3	0.95	330	0	0.002	140	0	0	3.9
182	0	7.7	25	0	0.014	33	0	0	1.2
364	0	2.7	383	0	0.012	24	0.05	0	0.78
552	4.6	0	290	0.11	0	81	0.004	0	2.2
728	13	0	330	1.4	0	100	0.006	0	2.8

The amounts of plutonium and americium in the suspended and dissolved fractions generally decreased with the reaction time, most noticeably in tests at 20,000 m⁻¹. The amounts of each found fixed to the vessel generally increased with the reaction time. Temporal changes are probably due to increased glass corrosion, changes in the solution chemistry, and flocculation of colloids.

The highest levels of neptunium, plutonium, and americium measured in the dissolved fractions under each test condition are summarized in Table 13. Also shown are the solubilities of these actinides measured in J-13 groundwater at room temperature at pH 8.5 [221]. These solubility concentrations are several orders of magnitude greater than the amounts measured to be dissolved in these tests. This discrepancy may be due to competing equilibria among dissolved, colloidal, and sorbed or plated phases, or to different solubility-controlling phases. The amounts of actinides released from either SRL 131A or SRL 202A in these tests were much smaller than the amounts released from R7T7 glass in tests conducted at lower SA/V ratios [248,249]. Except for weeksite, which may control the solubility of uranium, solubility-controlling phases remain to be identified.

These tests are relevant to glass performance because they indicate the amounts of each transuranic element that can be transported by groundwater flow. The dissolved fraction is assumed to be fully mobile, while the suspended fraction may or may not be fully mobile, depending on the surroundings. Although the fractions of actinides sorbed to the vessel or remaining with the glass are immobile during the test, actinides may be mobilized as the metal corrodes, as corrosion products slough off the glass, or as the solution chemistry changes. Of course, subsequent interactions may also affect the mobility of the dissolved fraction.

The amounts of actinides that remain associated with the glass during static leach tests can be estimated by comparing the amounts found to be suspended, dissolved, or fixed to the vessel to the extent of glass corrosion in each test. A "retention factor" has been used to describe ability of the corroded glass to mitigate the release of actinides to the solution [249]. The retention factor is defined as the ratio of the normalized boron mass loss (which defines the extent of corrosion) to the normalized actinide mass loss, where the boron release represents the extent of glass corrosion. Retention factors for technetium, uranium, and neptunium were calculated from the ICP-MS results. The sum of the amounts in the suspended, dissolved, and fixed fractions measured by alpha spectroscopy was used to compute the normalized mass losses of neptunium, plutonium, and americium. The amounts adsorbed to the steel vessel were included in the total amount released because the adsorbed material was not retained by the

Table 13. Highest Actinide Concentrations Measured in Leach Tests

Test	Np (M)	Pu (M)	Am (M)
SRL 202 at 2000 m ⁻¹	2 x 10 ⁻⁷	1 x 10 ⁻⁸	1 x 10 ⁻⁹
SRL 202 at 20,000 m ⁻¹	2 x 10 ⁻⁷	5 x 10 ⁻⁹	1 x 10 ⁻¹⁰
SRL 131 at 2000 m ⁻¹	1 x 10 ⁻⁷	5 x 10 ⁻⁹	1 x 10 ⁻¹¹
SRL 131 at 20,000 m ⁻¹	7 x 10 ⁻⁹	6 x 10 ⁻¹²	0
Solubility in J-13 (25°C) ^a	4.4 x 10 ⁻⁵	2.9 x 10 ⁻⁷	2.4 x 10 ⁻⁴

^aFrom Nitsche [221].

altered glass and was available for transport upon its release. The calculated retention factors for each radionuclide, $RF(Rn)$, where Rn is Tc, U, Np, Pu, or Am, are given for the tests in Table 14. A value of $RF(Rn)$ near one indicates that the actinide is released into solution to a similar extent as boron, while large values of $RF(Rn)$ indicate that the actinide is retained by the glass as it corrodes. Retention may result from several phenomena, including secondary phase formation, sorption, flocculation of colloidal material, and simply the limited solubilities of the actinides.

Note that the difference in the $RF(Np)$ calculated from the ICP-MS results and from the alpha spectroscopy results is due to the neptunium sorbed to the vessel walls. This fraction is counted as not being retained in the $RF(Np)$ calculated from the alpha spectroscopy results, but is counted as being retained in the $RF(Np)$ calculated from the ICP-MS results. As seen in Table 12, the neptunium adsorbed onto the vessel walls is the dominant fraction in tests conducted at $20,000\text{ m}^{-1}$ for both glasses. Technetium and uranium are not adsorbed onto the vessel walls.

In general, these tests show (1) actinides are retained by the glass much more than technetium (2) similar technetium retention factors are calculated for both glasses, at all SA/V ratios, and at all test durations, (3) the retention factors of the actinides are greater for the less durable SRL 131A glass than for the SRL 202A glass, increase with the SA/V ratio of the test, and increase with the test durations. The first finding can be explained as a result of the much higher solubility of technetium than any of the actinides. The differences in the solubilities of the actinides are also correlated with their retention: of the actinides, uranium and neptunium are more soluble and have lower retention factors than plutonium and americium. The observation that the retention factor of technetium is independent of the glass composition, SA/V ratio, and duration of the test simply shows that technetium is released congruent with boron, and that the effects of glass composition, SA/V ratio, and time on the glass corrosion rate similarly effect the technetium release. Conversely, this shows that technetium cannot be released any faster than the glass corrodes (as measured by the release of boron to solution).

The observation that the retention of the actinides varies with the glass composition, SA/V ratio, and duration of the test shows that their retention is affected by glass corrosion products, and can be explained as an effect of the formation of alteration layers, secondary phases, and colloids. This is seen in the difference of the dissolved and suspended fractions in Table 12. At short reaction times and at low SA/V ratios, most of the plutonium and americium are found suspended in solution. At longer times, very little is found suspended in solution. The change is attributed to the settling of colloids at longer reaction times. Analysis of the leachate solution before and after filtration through 6 nm filters has indicated that material containing aluminum, iron, and probably silicon becomes suspended in the leachates as the glasses corrode. This material remains suspended in the leachates of tests conducted at 2000 m^{-1} , but settles out of solution after 28 days in tests at $20,000\text{ m}^{-1}$. Actinides that become associated with suspended material during the initial stages of corrosion are removed from solution as the material flocculates and settles out of solution.

The higher retention of actinides by SRL 131 glass is due to its poorer durability. (The $NL(B)$ values for tests with SRL 131 glass are about an order of magnitude higher than those for tests with SRL 202 glass under the same test conditions.) Colloids containing actinides form in, and then settle out of, solution faster in tests with SRL 131A than in tests with SRL 202A glass. In addition, thicker alteration layers form on SRL 131A glass as it corrodes, which provides much more surface area for sorption of actinides than the thinner layers formed on SRL 202A glass.

Table 14. Radionuclide Retention Factors for Tests with SRL 202A and SRL 131A Glasses^a

Time, days	RF(Tc)	RF(U)	RF(Np)	RF(Pu)	RF(Am)
SRL 202 at 10 m ⁻¹					
7	0.45	16	1.2	1.5	0.07
28	<0.8 ^b	18	<4	<4	<0.5
94	<1	31	<3	<3	<0.5
238	<6	9.1	4.5	<27	<3
600	<26	59	42	60	<15
SRL 202 at 2000 m ⁻¹					
14	1.4	2.4	1.9	12	19
30	1.5	5.2	2.9	11	16
70	1.3	7.5	2.4	9.2	14
140	1.3	6.1	2.8	8.5	13
280	0.9	5.9	2.0	4.5	9.7
560	0.95	5.9	3.0	6.1	9.9
980	0.85	6.0	12	8.3	15
SRL 202 at 20,000 m ⁻¹					
14	1.7	4.0	4.8	1.9	3.6
28	1.2	7.0	8.0	3.8	5.3
56	1.1	15	15	13	14
98	1.1	27	25	14	17
182	0.63	58	7.9	18	19
364	2.0	1400	9.0	120	160
484	— ^c	9000	28	400	360
736	—	8000	54	180	210
SRL 131 at 10 m ⁻¹					
7	<0.5	5.5	1.7	<2	<0.2
28	0.75	9.7	1.4	0.7	4.3
94	<2.8	8.5	6.0	<11	<14
238	2.8	7	1.4	30	14
601	1.7	8.5	3.4	<340	<460
SRL 131 at 2000 m ⁻¹					
14	2.3	7.0	10	120	570
30	2.3	13	15	65	310
70	2.1	19	42	120	120
140	2.2	26	52	37	70
280	5.5	63	59	110	380
560	1.8	270	550	180	460
980	1.7	1000	600	220	580
SRL 131 at 20,000 m ⁻¹					
14	1.7	4.0	40	800	1000
28	1.7	3.2	58	280	430
56	1.8	4.0	64	520	600
98	1.1	4.4	55	130	190
182	1.2	5.9	710	670	730
364	1.6	12	100	1700	2100
552	—	12	160	570	840
728	—	15	210	720	1000

^aValues in bold type calculated from alpha-counting analyses. Values in regular type calculated from ICP-MS analyses.

^bMaximum value based on analyzed radionuclide concentration below detection limit.

^cAnalysis not yet performed.

Colloids that have been identified to be suspended in the leachates of these tests include smectite and kaolinite clays, nagelschmidtite, calcite, dolomite, magnesium oxide, titanium oxide, weeksite, uranophane, boltwoodite, and phases rich in uranium-titanium-, iron-, and aluminum [263]. Smectite clays are the most ubiquitous and abundant phases found suspended in the solution. Smectite clays are known to be an abundant alteration phase of the hydrated surfaces of these glasses [64,65]. All the other phases have been found to be either associated with alteration layers formed on individual glass grains or existing as separate phases mixed with the reacted glass. These phases may nucleate both in the solution and on the reacted glass surface, and then settle out of solution as separate solids as they increase in size. The suspended phases allow mobile fractions of actinide elements to exist in the leachate at levels far above their solubilities either through their being sorbed onto or incorporated into the phases. While several uranium-bearing phases have been identified, the low concentrations of transuranic elements used in testing make it difficult to identify the secondary phases which sequester them and/or affect their solubilities using electron beam techniques. Autoradiography has been used to identify particles containing plutonium and americium oxides associated with colloidal clay material in other tests [262]. The same clay colloids have been found to form in the present tests, and it is likely that actinides become associated with them. Work is in progress to identify which actinide phases are formed during glass corrosion.

9.3 Effects of the SA/V on the Alteration of the Glass Surface

The alteration of the glass surface as the glass corrodes is of interest because

- (1) alteration layers provide valuable insight into the corrosion mechanism of the glass, particularly the glass/layer interface. The relation between the processes leading to glass dissolution (i.e., water diffusion, ion exchange, and hydrolysis) will be revealed at this interface. For example, whether or not the interrelation of water diffusion and ion exchange observed in the early stages of corrosion in work by Dran et al. (see Section 2.1.3) persists throughout the corrosion process needs to be known to describe the long-term corrosion behavior. An understanding of the glass corrosion mechanism and rate limiting process is important for characterizing the long-term corrosion rate needed in performance assessment models.
- (2) Alteration layers provide insight regarding the control the solubilities of key glass matrix elements during Stage II of the corrosion (prior to the formation of secondary phases). One approach to projecting the glass corrosion rate requires analysis of the alteration layer to define the saturation conditions [143]. Also, secondary phases formed after the solution becomes saturated may nucleate on or in the alteration layer. These phases must be identified to project long-term glass corrosion behavior.
- (3) Alteration layers commonly retain radionuclides as the glass corrodes, and layer analysis is needed to characterize their disposition.

Alteration layers that formed in tests in this Task that were conducted at different SA/V ratios are being characterized and compared to determine how the SA/V ratio of the test affected the composition, structure, and thickness of the layers. Crystalline layers were observed to form at the glass surface in tests conducted at SA/V ratios of 340, 2000, and 20,000 m⁻¹ at all reaction times tested. (Layers formed in tests at 10 m⁻¹ have not yet been analyzed. Layers formed after short reaction times are expected to be amorphous, as was seen in tests with SRL 165 glass at an SA/V ratio of 30 m⁻¹ conducted for 56 days or less [87].) Fibrous clay crystals, oriented perpendicular to the glass surface, formed at the shortest reaction times tested at all SA/V (3 days in the PCT tests and 7 days in the MCC-1 tests). After longer reaction times, these crystals became consolidated into laths of clay that were generally oriented perpendicular to the glass surface. Figures 59a-f show the TEM images of alteration layers that formed on SRL 202U glass in tests in either deionized water or EJ-13 solution at SA/V ratios of 340, 2000, or 20,000 m⁻¹. The alteration layers are on the top halves of Figs. 59a-e and on the left side of Fig. 59f. The layers have formed separated from the glass in Figs. 59b-e, but not in Figs. 59a and 59f. The

Fig. 59. Photomicrographs of Alteration Layers Formed on SRL 202U Glass: (a) 340 m^{-1} /DIW after 28 Days (bar = 100 nm), (b) 340 m^{-1} /DIW after 360 Days (bar = 100 nm), (c) 340 m^{-1} /DIW after 360 Days (bar = 100 nm), (d) 2000 m^{-1} /EJ-13 after 360 Days (bar = 100 nm), (e) $20,000 \text{ m}^{-1}$ /EJ-13 after 180 Days (bar = 100 nm), and (f) $20,000 \text{ m}^{-1}$ /EJ-13 after 364 Days (bar = 400 nm). Distance represented by bars are given in parentheses (after [64]).

unreacted glass was chattered during sample preparation, but the alteration layers that had formed on the glass during testing were not chattered. The layers remained intact because epoxy used to fix the particles during sample preparation had penetrated the layers but not the glass.

Figure 59a shows the layer formed in a test conducted at an SA/V ratio of 340 m^{-1} after 28 days of corrosion in deionized water. Laths of crystalline material radiate perpendicular to the surface, and would have projected into the leachate solution during the test. A layer of fine-grained material can be seen beneath the layer of coarse crystals. The fine-grain material appears to remain in contact with the underlying glass, though the layer has been mechanically separated from the glass in some areas during sample preparation. Figure 59b shows that, after 360 days in tests in deionized water, small, randomly oriented crystallites have formed within the initially amorphous layer, and discrete laths of crystalline material identical to that formed at shorter times has formed between the amorphous layer and the glass surface. This results in the formation of a twinned layer of coarse-grained crystalline laths that separates from the underlying glass. A thin band of fine-grained material remains between the layers of lath-like crystals in most samples. Notice that another amorphous layer does not form on the glass surface exposed as the layer separates from the glass. The glass beneath the layer appears to dissolve stoichiometrically as corrosion continues, though the erosion of the glass is not uniform and the glass eventually looks pitted.

The layer formed after corrosion in deionized water at 2000 m^{-1} for 360 days is shown in Fig. 59c. This layer is very similar to that formed in tests at 340 m^{-1} for 360 days shown in Fig. 59b. Both have a twinned appearance with coarse-grained crystalline laths protruding inward towards the glass and outward into the resin with an inner band of fine-grained material in the center. The layers in both tests have separated from the underlying glass and over-all thicknesses are similar. The layer in tests in EJ-13 solution at 2000 m^{-1} run for 360 days is shown in Fig. 59d. This layer is similar to that shown in Fig 59c, except the inner band of fine-grain material is thinner in layers formed on glasses reacted in EJ-13 solution than on those formed in deionized water. A similar layer is formed after reaction for 182 days in EJ-13 solution at $20,000 \text{ m}^{-1}$, as shown in Fig. 59e.

The layer formed in a test conducted for 364 days in EJ-13 solution at $20,000 \text{ m}^{-1}$ is shown in Fig. 59f. This test had shown a sudden release of soluble glass components indicative of an increased corrosion rate (see Section 4.1). The increased corrosion rate results in the formation of a much thicker alteration layer than seen prior to the increase in the rate. The outer portion of the layer is identical to that formed previously. It consists of an outer layer of coarse-grained crystals oriented perpendicular to the surface that has a thickness similar to that of the layer formed after 182 days. A thicker inner layer of fine-grain material is present between the outer layer of coarse-grain material and the unreacted glass. The fine-grain material interfaces with both the outer layer and the underlying glass, i.e., it does not show the separation between layer and glass the way the coarse-grain layers did (compare to Figs. 59b-e).

Compositional analysis of the layers by means of EDS and structural analysis by means of selected area electron diffraction (SAED) showed that the outer and inner layers were composed of the same polycrystalline material, which is identified as a smectite clay. Table 15 gives a representative composition of the clay laths in the oriented outer layers and the random inner layer formed in deionized water. The compositions of the inner and outer layers are similar; both are depleted of soluble glass components and enriched in aluminum and chloride. Clays with a similar composition are formed in EJ-13 [64]. Table 16 gives the lattice spacings measured from several representative SAED patterns and those of some smectite clays.

Table 15. Measured Composition of Alteration Layers Formed on SRL 202 Glass Reacted in DIW at 20,000 m⁻¹ for 364 Days^a

Element	Outer Layer	Inner Layer	Unreacted Glass
Al	8.5	10	4.9
Ca	0.1	0.1	2.2
Cl	3.2	3.2	<0.5
Fe	20	16	21
K	0.6	0.9	7.5
Mg	1.1	1.5	0.2
Mn	4.4	4.7	4.3
Na	0.5	0.1	8.2
Ni	2.5	2.9	2.1
Si	57.8	59.5	48
Ti	1.3	1.1	1.6

^aIn elemental weight percent, normalized to 100% total.

Table 16. Measured d-Spacings from Selected Area Electron Diffraction Analysis of SRL 202 Glass Reacted at 20,000 m⁻¹ for 364 Days, in Angstroms

Layer I	Layer II	Layer III	Nontronite (29-1497) ^a	Montmorillonite (12-219)	Saponite (29-1491)
			15.2	17.6	15.5
9.0 ^b				9.00	
			7.44		7.73
				5.99	5.11
4.48	4.48	4.42 ± 0.12 ^c	4.48	4.49	4.57
		3.20 ± 0.07	3.58	3.58	3.834
			3.05	2.99	3.07
2.57	2.53	2.58 ± 0.05	2.564	2.57	2.61
				2.242	
		1.70 ± 0.03		1.699	1.731
1.50	1.50	1.50 ± 0.02	1.51	1.504	1.533
1.29	1.29	1.30 ± 0.02	1.336	1.294	1.321

Layer I: outer layer/EJ-13; II: inner layer/DIW; III: outer layer/DIW.

^aJCPDS file number of representative smectite clay.

^bMeasured from lattice fringes in image.

^cEstimate measurement errors common to all experimental values.

The structure and composition of isolated secondary phases were analyzed by AEM and SEM. Phases identified include the uranium-bearing phases weeksite and boltwoodite, zeolites in the chabazite and heulandite groups, clinoptilolite, and nagelschmidtite. A noncrystalline feature bearing uranium and titanium was also identified within the clay phase or between the clay layer and the underlying glass.

9.4 Summary

A series of static leach tests is being conducted to address remaining issues regarding the effect of the SA/V ratio on the glass corrosion behavior and the disposition of radionuclides during glass corrosion. These results show that the SA/V ratios used in static leach tests affect both the dominant reaction step and the long-term rate of glass reaction. The reaction can be characterized in terms of three stages: reaction at a forward rate characteristic of the glass composition and solution pH, at an intermediate rate that may become very low, and at a long-term rate, which may be the same as the intermediate rate or may be accelerated by the effects of secondary phase formation. The SA/V used in a test influences which reaction stage is observed during the test. Tests at low SA/V (10 and 340 m⁻¹) maintain dilute leachate conditions, and the observed glass reaction proceeds at near the forward reaction rate. The glass reaction is dominated by the leaching of alkali metals and boron from the glass, but the silicon release and dissolution of the glass network is low under these conditions. The release of alkali metals leads to an increase in the solution pH value, which facilitates the silicon release and network dissolution. As the silicic acid concentration increases as the glass reacts, the reaction slows and approaches a minimum rate. The higher pH values attained in tests at 2000 m⁻¹ promote silica dissolution and lead to high solution concentrations of several glass components. The high silicic acid concentrations eventually slow the glass corrosion to a low rate. At an even higher SA/V (20,000 m⁻¹), the glass reaction slows initially as the silicic acid level increases, but then it accelerates as secondary phases form to consume silicic acid and other solution components. Because the reaction step controlling the observed glass dissolution rate may differ in tests at different SA/V, the measured long-term rate is sensitive to the test conditions, including the pH that is attained and secondary phase formation, and the SA/V cannot be used to quantitatively or uniformly accelerate the glass reaction progress.

Actinide elements become distributed between dissolved, suspended, and immobile phases as glass waste forms corrode. The disposition of actinides measured in laboratory tests may be affected by their interaction with glass corrosion products, complexants in the leachate, and the reaction vessel. Specific solubilities and interactions of the actinides are complicated by chemical and physical changes that may occur during the test. These include changes in the solution chemistry as the glass corrodes, especially the ligand concentrations and the pH; in the abundance of colloidal material; and in the reaction vessel. Corresponding effects will be present during storage of waste forms in geologic repositories and must be accounted for when assessing the performance of a disposal site. The present tests at ANL demonstrate that (1) the concentrations of complexants and the disposition and solubilities of actinide elements are sensitive to the SA/V of the test, (2) association of actinides with suspended corrosion products may increase their solution contents, (3) the abundance of suspended material depends on the solution chemistry and the reaction time, and (4) the amounts of actinide materials available for transport are affected by their solubilities and the stabilities of suspended phases with which they become associated.

The release of transuranics increases as the glass reacts during this stage, but their low solubilities limit the solution concentrations to low values. The release of transuranics into solution increases initially, but secondary phase and colloid formation eventually removes them from solution at longer reaction times. The release of transuranics is not affected by the accelerated glass reaction at high SA/V, but is controlled by their low solubilities.

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APPENDIX A. DEFINITIONS

Accelerated Test - a test in which the values of independent test parameters used result in increased alteration rates of the specimen relative to rates under expected service conditions. Any change in the corrosion mechanism must be accounted for in the use of test data.

Alteration - a change in the chemical or physical properties of a material.

Characterization Test - any test performed to provide information to support the mechanistic description of the corrosion process.

Corrosion Model - a set of chemical reactions and/or physical processes that can be used to describe observed phenomena.

Diffusion - the process of mass transfer of a reactant or product species through either a liquid or solid phase as a result of chemical potential gradient to maintain corrosion. A diffusion coefficient is defined in some corrosion models to characterize the release of silicic acid and other glass components during corrosion via partial retention by the altered surface.

Dynamic Test - a laboratory test in which the corroding fluid is continuously or periodically refreshed during the test.

Effective Flow Rate - the equivalent velocity of leachant passing the glass surface simulated by the periodic removal of leachate and addition of fresh leachant in a reaction cell. Associated with the effective flow rate is a characteristic replacement time, which is the period required for complete refreshment of the solution within the sample chamber or corrosion cell.

Empirical Model - a model based exclusively on experimental observations, independent of model or theory.

Extent of Corrosion - a measure of the amount of glass that is chemically or physically altered as a result of interactions with a fluid. Material may be dissolved from the glass and either enter solution or become separated from the glass structure and form a secondary solid phase.

Flow Rate - the velocity of the leachant passing the glass surface. The flow rate affects the corrosion by changing the fluid chemistry at the corroding surface.

Glass Surface Area - the area of glass available for corrosion reactions. The total surface area includes free surfaces as well as surfaces within cracks and pores that are accessible to water.

Glass Surface Area-to-Solution Volume Ratio (SA/V) - the ratio of the surface area of the glass available for reaction and the leachant solution volume available to dilute the reaction products. The SA/V ratio is a system parameter and not a property of either the glass or the fluid. Because they occur in most reaction rate equations as a ratio, the glass surface area and leachant volume are conveniently represented as a ratio and discussed as a single parameter.

Hydrolysis - the reaction in which molecular water is inserted into a bond, resulting in the rupture of that bond. The hydrolysis reaction may be catalyzed by the nucleophilic attack by hydroxide ions (base-catalyzed) or electrophilic attack by hydronium ions (acid-catalyzed) on the glass. Hydrolysis of an oxygen-alkali metal or alkaline earth bond is also referred to as ion exchange.

Instantaneous Corrosion Rate - the rate at a specific time during corrosion, usually determined over a specific time interval.

Integrated Corrosion Rate - the rate determined by dividing the measured extent of corrosion by the total reaction time.

Intrinsic Corrosion Rate - the rate at which a glass reacts in the absence of corrosion products. The intrinsic corrosion rate depends on the glass composition, temperature, and solution pH.

Ion Exchange - the reaction in which an ion fixed in the glass is replaced by an ion previously associated with a mobile species. The hydrolysis reactions responsible for the release of alkali metals and alkaline earths from the glass are often described as occurring through ion exchange reactions, in which a proton from molecular water or hydronium replaces the alkali metal or alkaline earth ion in the glass structure. The alkali or alkaline earth ion is then free to be solvated and migrate into solution.

Leachant - the fluid that has not yet interacted with the glass. In laboratory tests, the leachant is the fluid added to the tests.

Leachate - the fluid that has contacted and interacted with the glass and contains dissolved glass components. The leachate solution may be analyzed to provide a measure of the extent of glass corrosion.

Mechanistic Model - a model based on chemical and physical laws and processes.

Model - a representation of a process as a set of chemical reactions and processes.

Nuclear Waste Glass - the fusion product of radioactive waste and glass-forming additives.

Response Test - any test performed primarily to provide information for comparative purposes independent of a corrosion mechanism.

Simulated Waste Glass - the fusion product of glass-forming additives and chemicals intended to represent the chemical behavior of radioactive elements.

Simulation - an implementation of a mechanistic model described in a mathematical algorithm, usually in a computer code.

Solution Volume - the volume of fluid contacting the glass. In static tests, this is the volume of leachant included in the test less the amount lost to vessel leakage and evaporation. In dynamic tests, the solution volume may refer to the integrated volume contacting the glass throughout the test duration, or to the volume of the corrosion cell.

Static Test - a laboratory test in which the corroding fluid is not refreshed during the test.

Waste Form - the radioactive waste materials and any stabilizing matrix prepared for long-term storage or disposal.

Vitrification - the process of fusing chemicals to generate a glass.

APPENDIX B. GLASS COMPOSITIONS

The compositions of glasses used in tests described in this report are tabulated for reference. Because compositional variations occur between different laboratories and even between different testing programs within a laboratory, the compositions presented should be used as a guide for differences in major components. Compositions were obtained from the following references.

C31-3.....	[152]
DWRG.....	[169]
HW-39.....	[164]
I117.....	[254]
JSS-A.....	[164]
MW.....	[164]
PNL 76-68.....	[152]
R7T7.....	[164]
SM58LW11.....	[156]
SRL 131A.....	[112]
SRL 165A.....	[112]
SRL 200R.....	[200]
SRL 200S.....	[200]
SRL 202A.....	[112]
SRL EA.....	[101]
SRP Black Frit.....	[196]
UK 189.....	[258]
WV 205.....	[288]

OXIDE	C31-3	DWRG	HW-39	I 117	JSS-A	MW	PNL 76-68	R7T7	SM58LW11
AgO					0.03	0.02		0.03	
Al2O3	10.28	6.58	4.3	5	4.87	5.33	0.6	4.91	2.1
B2O3	4.14	6.77	9.6	15	14.4	16.65	8.81	14.02	12.3
BaO	15.28		0.2	0.33	0.06	0.57	0.53	0.06	0.1
CaO	3.84	1.95	2.9		4.01		2.36	4.04	3.8
CdO					0.03	0.02		0.03	
CeO2	1.38				0.96	1.02	0.9	0.98	0.18
CoO		0.02			0.12			0.03	
Cr2O3		0.06	1.3		0.5	0.41		0.51	0.28
Cs2O	1.18	0.31	0.2	0.76	1.41	1.13	1.02	1.42	0.16
CuO		0.01	0.1	0.04					
Eu2O3						0.04			
Fe2O3	0.33	10.05	11.1	3.25	2.89	2.69	9.4	2.91	1.71
FeO									
Gd2O3						0.02			
K2O									0
La2O3	0.67		0.5	0.63	0.89	0.52	4.06	0.9	
Li2O	0.98	4.85	3.8		1.97	3.79	0	1.98	3.7
MgO	1.44	0.074	0.8		0.04	5.85	0.15		2.1
MnO2		3.09		1.35	0.71			0.72	0.67
MoO3	2.36		0.3	0.16	1.69	1.76	1.93	1.7	0.28
Na2O	3.12	9.2	10.4	18.49	9.79	8.1	11.53	9.86	8
Nd2O3	2.18		0.5	0.99	1.58	1.61	1.38	1.59	0.57
NiO		2.01	0.6	0.32	0.74	0.27		0.74	0.43
P2O5					0.48	0.18	0.7	0.28	
PbO									
Pr6O11			0.1	0.31	0.44	1.02		0.44	
PuO2					0.22				
Rb2O						0.13			
RuO2	1.28								
Sb2O3			0.4			0.06			
SeO2					0.01	0.01		0.01	
SiO2	34.75	49.9	51.3	48	45.12	46.2	40.84	45.48	56.8
Sm2O3				1.12		0.32			
SnO2				0.03	0.02	0.02		0.02	
SrO	0.45	0.46		0.22	0.33	0.36	0.42	0.33	0.05
TeO2						0.18		0.23	
Tc2O7					0.16				
ThO2								0.33	
TiO2	2.8	0.17					3.13		4.5
UO2	1.37				0.51			0.52	0.11
U3O8		2.61		1.38					
Y2O3					0.2			0.2	
ZnO	4.9			0.03	2.52	0.21	4.72	2.5	0.02
ZrO2	3.08	0.94	0.6	1.06	2.63	1.71	1.68	2.65	0.81
OXIDE	SRL 131A	SRL 165A	SRL 200R	SRL 200S	SRL 202A	SRP Black	SRL EA	UK 189	WV 205
AgO									
Al2O3	3.27	4.08	5.9	5.5	3.84	4.54	3.6	5.03	3.31
B2O3	9.65	6.76	9.7	9.6	7.97	7.35	11.16	21.87	9.97
BaO	0.16	0.06	0.02	0.03	0.22				0.06
CaO	0.93	1.62	0.9	0.9	1.2		1.23		0.61
CdO									
CeO2									0.18
CoO									
Cr2O3	0.13		0.3	0.2	0.03			0.55	0.22
Cs2O						0.07			0.11
CuO	0.02		0.1	0.1	0.4				
Eu2O3									
Fe2O3	12.66	11.74	9	8.8	11.41	11.4	7.58	2.68	11.83
FeO							1.59		
Gd2O3									
K2O	3.86	0.19	3.5	3.3	3.71		0.04		3.62
La2O3			0.06	0.01			0.28		0.12
Li2O	3	4.18	3.4	3.3	4.23		4.21	3.69	3
MgO	1.31	0.7	1.6	1.7	1.32	0.7	1.79	6.23	1.24
MnO2	2.43	2.79	1.6	1.5	2.21	1.98	1.36		1.73
MoO3			0.01	0.02	0.05				
Na2O	12.08	10.85	15	16	8.92	10.2	16.88	7.68	11.05
Nd2O3			0.1						
NiO	1.24	0.85	0.9	0.9	0.82		0.53	0.36	0.7
P2O5									2.52
PbO			0.03	0.05	0.01				
Pr6O11									
PuO2		0.01							
Rb2O									
RuO2									0.07
Sb2O3									0.14
SeO2									
SiO2	43.76	52.86	45.5	45.4	48.95	53.5	48.76	41.51	45.25
Sm2O3									
SnO2									0.13
SrO	0.01	0.11	0.01	0.02	0.03	0.09			0.13
TeO2									
Tc2O7	0.02	0.02			0.02				
ThO2					0.26				
TiO2	0.65	0.14	0.08	0.1	0.91		0.65		0.99
UO2	2.73	0.92	1.8	1.9	1.93				
U3O8						1.19		0.06	
Y2O3									0.03
ZnO	0.02	0.04	0.02		0.02		0.26	0.44	
ZrO2	0.22	0.66	0.06		0.1		0.48		2.99

APPENDIX C. DERIVATION OF RATE EQUATION

The following derivation is based on that of Aagaard and Helgeson [54]; deviations and comments are noted.

In general, the net rate of a chemical reaction is given as the difference of the forward and reverse rates

$$\text{net rate} = \text{rate}_f - \text{rate}_r \quad (\text{C-1})$$

This expression can be rearranged as

$$\text{net rate} = \text{rate}_f \left(\frac{1 - \text{rate}_r}{\text{rate}_f} \right) \quad (\text{C-2})$$

For a system at equilibrium,

$$\frac{\text{rate}_r}{\text{rate}_f} = 1 \quad (\text{C-3})$$

For a system not at equilibrium,

$$\frac{\text{rate}_r}{\text{rate}_f} = \exp \left(\frac{A}{RT} \right) \quad (\text{C-4})$$

where R is the gas constant, T is the absolute temperature, and A is the reaction affinity, which is defined as the negative of the partial derivative of the Gibbs free energy with respect to the reaction progress at constant temperature and pressure (where d indicates the partial derivative):

$$A = - \left(\frac{dG}{d\xi} \right)_{T,P} \quad (\text{C-5})$$

where G is the Gibbs free energy of the reaction and ξ is the reaction progress variable. The Gibbs free energy is a function of temperature, pressure, and the number of moles of each species, and its derivative can be written

$$dG = \left(\frac{dG}{dT} \right)_{P,N} dT + \left(\frac{dG}{dP} \right)_{T,N} dP + \sum_i \left(\frac{dG}{dN_i} \right)_{T,P,N_{j \neq i}} dN_i \quad (\text{C-6})$$

At constant T and P

$$dG = \sum_i \left(\frac{dG}{dN_i} \right)_{T,P,N_{j \neq i}} dN_i \quad (\text{C-7})$$

The partial derivative of the free energy with respect to the extent of reaction, ξ , is

$$\left(\frac{dG}{d\xi} \right)_{T,P} = \sum_i \mu_i \nu_i \quad (\text{C-8})$$

where μ_i is the chemical potential, and ν_i is the stoichiometric coefficient of species i in the reaction. Substituting

$$\mu_i = \mu_i^\circ + RT \ln a_i \quad (\text{C-9})$$

where R is the gas constant, into Eq. C-8 gives

$$\left(\frac{dG}{d\xi} \right)_{T,P} = \sum_i (\mu_i^\circ + RT \ln a_i) \nu_i \quad (\text{C-10})$$

or

$$\left(\frac{dG}{d\xi} \right)_{T,P} = \sum_i \mu_i^\circ \nu_i + \sum_i RT \ln (a_i)^{\nu_i} \quad (\text{C-11})$$

By definition,

$$\sum_i \mu_i^\circ \nu_i = \Delta G^\circ = -RT \ln K \quad (\text{C-12})$$

where K is the equilibrium constant, so

$$\left(\frac{dG}{d\xi} \right)_{T,P} = -A = -RT \ln K + RT \ln Q = RT \ln \left(\frac{Q}{K} \right) \quad (\text{C-13})$$

where Q is the ion activity product. Substituting this expression for the affinity, A , into Eqs. C-4 and C-2,

$$\text{net rate} = \text{rate}_f \left(1 - \exp \left(\frac{\ln Q}{K} \right) \right) = \text{rate}_f \left(1 - \frac{Q}{K} \right) \quad (\text{C-14})$$

Now consider the application of Eq. C-14 to the release of silicic acid from a glass phase. The net reaction can be written as

where the fact that silicic acid is a weaker base than silanol is accounted for in the reaction. The rate of this reaction can be written as

$$\text{net rate} = \frac{d[\text{H}_4\text{SiO}_4]}{dt} \quad (\text{C-16})$$

We note that when applying Eq. C-14 to a glass, the substitution made in Eq. C-12 is inexact because the standard states of all species i in the glass and in solution are not the same. The inexactness of this substitution may be the reason an ad hoc residual rate is required in the rate expression (see Eq. 21).

The forward rate is conveniently expressed by using activated complex theory. Consider the reaction to release silicic acid by nucleophilic attack of hydroxide at the underscored silicon atom to form the activated complex

Equation C-17 differs from previous derivations, where molecular water was the nucleophile. Hydroxide is indicated as the nucleophile in Eq. C-17 in response to the observed effects of the pH on glass corrosion. If molecular water is the reactant rather than hydroxide, then the activated complex is uncharged and its formation is pH-independent. In addition, molecular water is likely to form a hydrogen bond with the underscored oxygen atom. The activated complex can decompose by breaking the bond between the underscored silicon and the reactant hydroxide, in which case no net reaction occurs, or by breaking the bond between the underscored silicon and the underscored oxygen as

The proton on the nucleophilic hydroxide may also form a hydrogen bond with the underscored oxygen atom. The silicate species (either H_4SiO_4 or H_3SiO_4^-) formed during decomposition is shown to be in the reprotonated form to form silicic acid. In activated complex theory, the forward reaction rate is given by the rate of the decomposition step in Eq. C-18. If the concentration of the activated complex is given as $\text{AC}^\ddagger$, the first-order decomposition reaction rate can be written as

$$\text{rate}_f = \alpha_i f \text{AC}^\ddagger \quad (\text{C-19})$$

where α_i and f are the usual transmission coefficient and frequency factor. Activated complex theory assumes an equilibrium exists between the reactants and the activated complex, so we can write

$$K^\ddagger = \frac{\text{AC}^\ddagger}{(\equiv\text{Si}-\text{O}-\text{Si}(\text{OH})_3)(\text{OH}^-)} \quad (\text{C-20})$$

where $K^\ddagger$ is the equilibrium constant. Note that the (OH^-) term does not occur if molecular water is the nucleophile in the reaction. Solving Eq. C-20 for $\text{AC}^\ddagger$ and substituting into Eq. C-19 gives

$$\text{rate}_f = \alpha_i f (\equiv\text{Si}-\text{O}-\text{Si}(\text{OH})_3)(\text{OH}^-) K^\ddagger \quad (\text{C-21})$$

Now, the concentration of terminal $(\equiv\text{Si}-\text{O}-\text{Si}(\text{OH})_3)$ groups can be considered proportional to the total surface area:

$$(\equiv\text{Si}-\text{O}-\text{Si}(\text{OH})_3) = \beta_i \text{SA} \quad (\text{C-22})$$

where SA is the total surface area and β_i is a proportionality constant. Substituting this expression in Eq. C-19 gives

$$\text{rate}_f = \alpha_i f \beta_i \text{SA} (\text{OH}^-) K^\dagger \quad (\text{C-23})$$

The Gibbs free energy of formation of the activated complex can be expressed as

$$\Delta G^{\ddagger^\circ} = -RT \ln K^\dagger \quad (\text{C-24})$$

Also,

$$\Delta G^{\ddagger^\circ} = \Delta H^{\ddagger^\circ} + T \Delta S^{\ddagger^\circ} \quad (\text{C-25})$$

and

$$\Delta H^{\ddagger^\circ} = E_a - RT \quad (\text{C-26})$$

so the equilibrium constant can be written as

$$K^\dagger = \exp\left(\frac{-E_a}{RT}\right) \exp\left(\frac{RT}{RT}\right) \exp\left(\frac{\Delta S^{\ddagger^\circ}}{R}\right) \quad (\text{C-27})$$

Substituting Eq. C-27 into Eq. C-23 gives

$$\text{rate}_f = \alpha_i f \beta_i \text{SA} (\text{OH}^-) \exp\left(\frac{-E_a}{RT}\right) \exp\left(\frac{1 + \Delta S^{\ddagger^\circ}}{R}\right) \quad (\text{C-28})$$

Substituting Eq. C-28 into Eq. C-14 gives

$$\text{net rate} = \alpha_i f \beta_i \text{SA} (\text{OH}^-) \exp\left(\frac{-E_a}{RT}\right) \exp\left(\frac{1 + \Delta S^{\ddagger^\circ}}{R}\right) \left(1 - \frac{Q}{K}\right) \quad (\text{C-29})$$

Equation C-29 can be simplified by defining the reaction coefficient as

$$k_+ = \alpha_i f \beta_i (\text{OH}^-) \exp\left(\frac{-E_a}{RT}\right) \exp\left(\frac{1 + \Delta S^{\ddagger^\circ}}{R}\right) \quad (\text{C-30})$$

which gives the simplified expression for the corrosion rate as

$$\text{net rate} = \text{SA} k_+ \left(1 - \frac{Q}{K}\right) \quad (\text{C-31})$$

The values of k_+ and K are determined experimentally. Equation C-30 explicitly predicts the Arrhenius behavior of the rate coefficient. The pH dependence is also accounted for by the k_+ term. Equations C-31 and C-16 can be combined as

$$\frac{d[\text{H}_4\text{SiO}_4]}{dt} = SA k_+ \left(1 - \frac{Q}{K}\right) \quad (\text{C-32})$$

APPENDIX D. DATA GENERATED IN THE STUDY OF THE RELATIONSHIP BETWEEN HIGH SA/V TESTS AND MCC-1 TESTS

This appendix contains spreadsheets presenting data generated in tests conducted in the ANL Task "Relationship between High SA/V Tests and MCC-1." Tests within the Task were organized into series having a common glass, common leachant, and common SA/V, as shown in Table D.1 below. The results are presented in four tables: Table D.2, test matrices; Table D.3, cation results; Table D.4, anion results; and Table D.5, actinide results. Each table has a cover sheet that explains the spreadsheet headings. Calculations performed in Tables D.2 through D.5 are summarized in Table D.6, where sample calculations are also presented.

Table D.1. Test Series Description

Test Series	Glass Type	Leachant	SA/V (m ⁻¹)
BX1 - BX16	SRL 202A	EJ-13	10
BW1 - BW16	SRL 202A	EJ-13	340
BY1 - BY22	SRL 202A	EJ-13	2,000
BZ1 - BZ20	SRL 202A	EJ-13	20,000
PX1 - PX16	SRL 131A	EJ-13	10
PW1 - PW16	SRL 131A	EJ-13	340
PY1 - PY22	SRL 131A	EJ-13	2,000
PZ1 - PZ20	SRL 131A	EJ-13	20,000
TW1 - TW18	SRL 202U	DIW	340
TW31 - TW48	SRL 202U	EJ-13	340
TY1 - TY20	SRL 202U	DIW	2,000
TZ1 - TZ18	SRL 202U	DIW	20,000
TZ31 - TZ48	SRL 202U	EJ-13	20,000

Table D.2. Test Matrices

<u>Column Heading</u>	<u>Comment</u>
TEST No.	Test number in task matrix (Table D.1).
TARGET TIME	Scheduled reaction time, in days.
VESSEL No.	Vessel number in which test was performed.
DATE IN	Calendar date test was initiated.
DATE OUT	Calendar date test was terminated.
RXN TIME, d	Actual reaction time, in days.
SA, mm ²	Measured geometric surface area of monolithic sample(s), in square millimeters.
EJ-13, g	Mass of EJ-13 solution added as leachant, in grams.
DIW, g	Mass of deionized water added as leachant, in grams.
SA/V, /m	Calculated glass surface area/leachant volume ratio, in inverse meters.
VESSEL IN	Vessel mass at test initiation, in grams.
VESSEL OUT	Vessel mass at test termination, in grams.
CHANGE, g	Change in vessel mass during test, as VESSEL OUT-IN, in grams.
pH	Leachate pH measured at room temperature.
UF LPE	Mass of empty solution bottle used for unfiltered leachate, in grams.
+UF SOLN	Mass of solution bottle plus unfiltered leachate, in grams.
+DIW+HNO ₃	Mass of deionized water and nitric acid added to leachate, in grams.
UF DF	Dilution factor for unfiltered leachate.
F50 LPE	Mass of empty solution bottle used for filtrate, in grams.
+F50 SOLN	Mass of solution bottle plus filtrate, in grams.
+DIW+HNO ₃	Mass of deionized water and nitric acid added to filtrate, in grams.
F50 DF	Dilution factor for filtrate.

TEST No.	TARGET TIME	VESSEL No.	DATE IN	DATE OUT	RXN TIME, d	SA, mm2	EJ-13, g	SA/V, /m	VESSEL IN	VESSEL OUT	CHANGE, g
BX1	7	351	8/10/90	8/17/90	7	166	16.62	10	339.84	339.8	-0.04
BX2	7	352	8/10/90	8/17/90	7	167	16.66	10	341.21	341.2	-0.01
BX3	28	353	8/10/90	9/7/90	28	174	17.42	9.99	340.69	340.63	-0.06
BX4	28	354	8/10/90	9/7/90	28	169	16.9	10	344.2	344.05	-0.15
BX5	91	355	8/10/90	11/12/90	94	174	17.41	10	341.05	340.95	-0.1
BX6	91	356	8/10/90	11/12/90	94	165	16.48	10	339.11	339.04	-0.07
BX7	238	357	8/10/90	4/5/91	238	164	16.4	9.99	341.16	341.13	-0.03
BX8	238	358	8/10/90	4/5/91	238	162	16.23	10	340.33	340.09	-0.24
BX9	600	360	8/10/90	4/1/92	600	167	16.67	10	339.96	339.69	-0.27
BX10	600	361	8/10/90	4/1/92	600	169	16.93	10	341.03	340.91	-0.12
BX11	1400	362	8/10/90	6/1/94	1402	171	17.06	10	340.87	340.37	-0.5
BX12	1400	363	8/10/90	6/1/94	1402	164	16.44	10	340.38	340.38	0
BX13	2800	364	8/10/90	2/8/98		169	16.87	10	340.15		
BX14	2800	365	8/10/90	2/8/98		166	16.58	9.99	339.59		
BX15	TBD	52	2/7/91			169	16.88	9.99	340.7		
BX16	TBD	53	2/7/91			169	16.94	11.59	341.23		

TEST No.	pH	UF LPE	+ UF SOLN	+ DIW+HN03	UF DF	F50 LPE	+F50 SOLN	+DIW+HN03	F50 DF
BX1	8.21	24.27	38.78	10.07	1.694				
BX2	8.2	24.43	39.06	10.08	1.689				
BX3	8.28	24.36	39.4	10.17	1.6762				
BX4	8.34	24.33	38.63	10.11	1.707				
BX5	8.26	24.32	39.38	10.13	1.6726				
BX6	8.59	24.36	38.48	10.11	1.716				
BX7	8.43	8.04	17.09	0.12	1.0133	7.98	10.8	5.09	2.805
BX8	9.04	8	17.81	0.12	1.0122	7.97	10.99	5.06	2.6755
BX9	8.57	10.87	21.33	5.06	1.4837	10.79	13.33	12.16	5.7874
BX10	8.58	10.88	18.16	7.35	2.0096	10.98	13.91	12.15	5.1468
BX11	8.74	8.17	20.93	4.88	1.3824				
BX12	8.91	8.31	20.7	4.75	1.3834				
BX13									
BX14									
BX15									
BX16									

TEST No.	TARGET TIME	VESSEL No.	DATE IN	DATE OUT	RXN TIME, d	SA, mm2	EJ-13, g	SA/V, /m	VESSEL IN	VESSEL OUT	CHANGE, g
BW1	14		not run			683					
BW2	14		not run			680					
BW3	28		not run			674					
BW4	28		not run			677					
BW5	56		not run			686					
BW6	56		not run			668					
BW7	91		not run			679					
BW8	91		not run			675					
BW9	TBD	0	7/1/91			678	2.05	331	326.84		
BW10	TBD	183	7/1/91			683	2.05	333	324.65		
BW11	360	202	7/1/91	6/25/92	360	672	2.05	328	297.49	297.12	-0.37
BW12	360	242	7/1/91	6/25/92	360	682	2.05	333	294.91	294.89	-0.02
BW13	540	338	7/1/91	12/22/92	540	672	2.05	328	299.76	299.76	0
BW14	540	265	7/1/91	12/22/92	540	679	2.05	331	291.16	289.35	-1.81
BW15	720	357	7/1/91	6/20/93	715	683	2.05	333	293.89	293.56	-0.33
BW16	720	281	7/1/91	6/20/93	715	674	2.05	32	327.37	327.29	-0.08
BW17	180		not run			677					
BW18	180		not run			682					

TEST No.	pH	F.45 LPE	+F.45 SOLN	+DIW+HN03	F.45 DF	F50 LPE	+F50 SOLN	+DIW+HN03	F50 DF
BW1									
BW2									
BW3									
BW4									
BW5									
BW6									
BW7									
BW8									
BW9									
BW10									
BW11	8.03	10.78	11.45	15.28	23.806				
BW12	8.14	11	11.89	15.15	18.022				
BW13	5.83	10.95	11.92	5.01	6.1649				
BW14									
BW15	6.42	10.99	11.91	9.77	11.62				
BW16	3.67	10.94	12.12	9.82	9.322				
BW17									
BW18									

TEST No.	TARGET TIME	VESSEL No.	DATE IN	DATE OUT	RXN TIME	SAMPLE IN	EJ-13, g	SA/V, /m	VESSEL IN	VESSEL OUT	CHANGE, g
BY1	3	343	7/23/90	7/26/90	3	0.9932	10	1986	330.38	330.37	-0.01
BY2	3	344	7/23/90	7/26/90	3	1.0204	9.96	2049	331.64	331.63	-0.01
BY3	7	347	7/27/90	8/3/90	7	0.9734	10	1947	331.37	331.35	-0.02
BY4	7	348	7/27/90	8/3/90	7	1.022	9.99	2046	330.7	330.65	-0.05
BY5	14	339	7/10/90	7/24/90	14	0.9972	9.95	2004	331.07	331.05	-0.02
BY6	14	340	7/10/90	7/24/90	14	0.9886	10.05	1967	330.54	330.54	0
BY7	30	323	6/20/90	7/20/90	30	1.0215	10.01	2041	331.11	331.11	0
BY8	30	324	6/20/90	7/20/90	30	1.0004	10.09	1983	329.31	329.28	-0.03
BY9	70	33	10/8/90	12/17/90	70	1.0254	10.29	1993	329.79	329.72	-0.07
BY10	70	83	10/8/90	12/17/90	70	0.9926	10.02	1981	330.02	330.02	0
BY11	140	154	10/8/90	2/25/91	140	0.9944	9.96	1997	296.76	296.72	-0.04
BY12	140	155	10/8/90	2/25/91	140	1.024	10.22	2004	328.99	328.92	-0.07
BY13	280	325	6/20/90	3/27/91	280	0.9825	10.03	1959	330.53	330.52	-0.01
BY14	280	326	6/20/90	3/27/91	280	0.9982	10	1996	330.63	330.62	-0.01
BY15	560	327	6/20/90	1/1/92	560	1.0076	9.99	2017	330.61	330.46	-0.15
BY16	560	328	6/20/90	1/1/92	560	1.0017	10.02	1999	329.69	329.6	-0.09
BY17	980	329	6/20/90	2/24/93	980	0.9473	10.01	1893	329.29	329.28	-0.01
BY18	980	330	6/20/90	2/24/93	980	0.9949	10	1990	330.95	330.94	-0.01
BY19	1820	331	6/20/90	6/3/95		1.0088	10.02	2014	330.57		
BY20	1820	332	6/20/90	6/3/95		1.0182	10.02	2032	330.26		
BY21	TBD	93	2/7/91			1.0096	10.1	1999	332.07		
BY22	TBD	107	2/7/91			1.0084	10.12	1993	329.16		

TEST No.	pH	F.45 LPE	+F.45 SOLN	+DIW+HN03	F.45 DF	F50 LPE	+F50 SOLN	+DIW+HN03	F.50 DF
BY1	9.27	24.15	31.67	10.05	2.3364				
BY2	9.3	24.2	31.64	10	2.3441				
BY3	9.48	24.42	31.99	10.06	2.3289				
BY4	9.48	24.17	31.49	10.07	2.3757				
BY5	9.77	24.25	31.37	10.02	2.4073				
BY6	9.74	24.16	31.56	10.4	2.4054				
BY7	9.94	24.32	31.72	10.01	2.3527				
BY8	9.87	24.29	31.71	10.03	2.3518				
BY9	10.09	24.27	32.15	10.09	2.2805				
BY10	10.11	24.56	31.89	10.09	2.3765				
BY11	10.02	7.77	10.11	3.27	2.3974	7.57	9.16	3.76	3.3648
BY12	10.11	7.53	9.93	3.14	2.3083	7.65	9.22	3.62	3.3057
BY13	10.47	7.79	14.15	10.2	2.6038				
BY14	10.45	7.69	14.17	10.24	2.5802				
BY15	10.33	10.91	15.69	15.1	4.159	11.09	12.59	15.18	11.12
BY16	10.42	10.83	15.36	15.06	4.3245	10.99	12.53	15.15	10.838
BY17	10.52	11.09	15.66	15.25	4.337	10.99	14.87	15.16	4.9072
BY18	10.65	10.89	15.48	15.09	4.2876	11.06	14.96	15.12	4.8769
BY19									
BY20									
BY21									
BY22									

TEST No.	TARGET TIME	VESSEL No.	DATE IN	DATE OUT	RXN TIME, d	SAMPLE IN	EJ-13, g	SA/V, /m	VESSEL IN	VESSEL OUT	CHANGE, g
BZ1	3	345	7/23/90	7/26/90	3	5.0123	4.99	20089	328.34	328.32	-0.02
BZ2	3	346	7/23/90	7/26/90	3	4.9312	5.03	19607	327.85	327.84	-0.01
BZ3	7	349	7/27/90	8/3/90	7	5.018	5	20072	329.63	329.62	-0.01
BZ4	7	350	7/27/90	8/3/90	7	4.965	5.01	19820	329.08	329.07	-0.01
BZ5	14	341	7/10/90	7/24/90	14	4.9457	4.97	19902	328.51	328.5	-0.01
BZ6	14	342	7/10/90	7/24/90	14	4.9793	4.98	19997	329.64	329.61	-0.03
BZ7	28	337	6/22/90	7/20/90	28	5.0235	4.95	20297	329.24	329.22	-0.02
BZ8	28	338	6/22/90	7/20/90	28	5.0319	5.02	20047	329.62	329.6	-0.02
BZ9	56	250	10/8/90	12/3/90	56	4.9792	4.99	19957	329.31	329.25	-0.06
BZ10	56	257	10/8/90	12/3/90	56	5.0185	5.01	20034	330.5	330.49	-0.01
BZ11	98	270	10/8/90	1/14/91	98	5.0027	4.97	20132	329.63	329.59	-0.04
BZ12	98	281	10/8/90	1/14/91	98	4.9837	4.98	20015	330.21	330.14	-0.07
BZ13	182	283	10/8/90	4/8/91	182	4.9708	4.98	19963	328.59	328.4	-0.19
BZ14	182	318	10/8/90	4/8/91	182	5.0419	5	20168	328.4	328.38	-0.02
BZ15	364	333	6/20/90	6/19/91	364	5.0021	5	20008	330.09	330.03	-0.06
BZ16	364	334	6/20/90	6/19/91	364	5.0221	5	20088	328.48	328.28	-0.2
BZ17	728	335	6/20/90	6/25/92	736	4.9692	5.02	19798	330.07	329.94	-0.13
BZ18	728	336	6/20/90	6/25/92	736	4.9337	5.02	19656	329.53	329.47	-0.06
BZ19	TBD	135	2/7/91	6/25/92	484	5.0224	5.03	19542	329.16	329.11	-0.05
BZ20	TBD	139	2/7/91			5.0552	5.14	19670	328.12		

TEST No.	pH	F.45 LPE	+F.45 SOLN	+DIW+HN03	F.45 DF	F50 LPE	+F50 SOLN	+DIW+HN03	F50 DF
BZ1	10.5	24.3	26.61	10.07	5.3593				
BZ2	10.62	24.46	26.83	10.01	5.2236				
BZ3	10.73	24.29	26.43	10.06	5.7009				
BZ4	10.48	24.35	26.56	10.06	5.552				
BZ5	10.98	24.28	26.51	10.04	5.5022				
BZ6	11.01	24.27	26.56	10.17	5.4411				
BZ7	11.02	24.13	25.94	10.03	6.5414				
BZ8	11.1	24.25	25.76	10.1	7.6887				
BZ9	11.24	24.52	25.97	15.06	11.386				
BZ10	11.33	24.22	25.66	15.05	11.451				
BZ11	11.26	24.56	26.19	15.21	10.331				
BZ12	11.43	24.23	25.89	15.13	10.114				
BZ13	10.92	8.05	8.85	10.09	13.612	8.01	9.35	5.14	4.8358
BZ14	11.29	8.03	8.99	10.16	11.583	7.99	9	5.13	6.0792
BZ15	11.06	11.06	12.16	10	10.091	10.98	11.95	5.09	6.2474
BZ16	11.02	11.02	12.08	10.04	10.472	11.06	12.02	5.06	6.2708
BZ17	11.96	10.83	11.56	15.08	21.658	10.9	12.21	15.25	12.641
BZ18	11.97	10.93	11.65	15.23	22.153	10.99	12.32	15.32	12.519
BZ19	11.86	10.82	11.61	15.4	20.494	10.96	12.36	15.15	11.821
BZ20									

TEST No.	TARGET TIME	VESSEL No.	DATE IN	DATE OUT	RXN TIME, d	SA, mm2	EJ-13, g	SA/V /m	VESSEL IN	VESSEL OUT	CHANGE, g
PX1	7	337	8/28/90	9/4/90	7	172	17.19	10	340.57	340.5	-0.07
PX2	7	338	8/28/90	9/4/90	7	172	17.1	10	340.75	340.72	-0.03
PX3	28	339	8/28/90	9/25/90	28	168	16.78	10	341.15	340.81	-0.34
PX4	28	340	8/28/90	9/25/90	28	168	16.78	10	339.7	339.67	-0.03
PX5	91	341	8/28/90	11/30/90	94	168	16.79	10	340.25	340.24	-0.02
PX6	91	342	8/28/90	11/30/90	94	168	16.79	10	342.25	342.13	-0.12
PX7	238	343	8/28/90	4/23/91	238	165	16.51	9.99	341.81	338.89	-2.97
PX8	238	344	8/28/90	4/23/91	238	172	17.17	10	343.06	343.04	-0.02
PX9	600	345	8/28/90	4/20/92	601	185	18.51	9.99	345.24	343.76	-1.48
PX10	600	346	8/28/90	4/20/92	601	171	17.11	9.99	340.85	339.34	-1.51
PX11	1400	347	8/28/90	6/21/94	1404	166	16.61	9.99	341.74	337.93	-3.06
PX12	1400	348	8/28/90	6/21/94	1404	169	16.91	9.99	340.99	338.77	-2.22
PX13	2800	349	8/28/90	2/26/98		169	16.93	9.98	341.02		
PX14	2800	350	8/28/90	2/26/98		166	16.63	9.98	340.58		
PX15	TBD	91	2/7/91			173	17.3	10	339		
PX16	TBD	92	2/7/91			167	16.8	9.94	338.62		

TEST No.	pH	UF LPE	+ UF SOLN	+ DIW+HNO3	UF DF	F50 LPE	+F50 SOLN	+DIW+HNO3	F50 DF
PX1	8.18	24.23	39.21	10.08	1.6729				
PX2	8.24	24.19	39.15	10.13	1.6771				
PX3	8.3	24.24	38.66	10.08	1.699				
PX4	8.24	24.32	38.95	10.16	1.6945				
PX5	8.44	24.36	38.96	9.45	1.6473				
PX6	8.31	24.51	38.96	10.06	1.6962				
PX7	8.58	7.68	15.19	6.99	1.9308	7.71	9.5	5.1	3.8492
PX8	9.27	7.71	19.7	6.99	1.583	7.76	9.67	5.08	3.6597
PX9	9.09	10.79	22.15	5.64	1.4965	10.92	13.64	10.2	4.75
PX10	9.34	10.73	21.62	5.14	1.472	10.81	15	11.07	3.642
PX11	8.96	11.02	18.46	7.51	2.0094	11.01	14.63	7.45	3.058
PX12	9.32	11.11	20.57	7.49	1.7917	11.1	14.81	7.46	3.0108
PX13									
PX14									
PX15									
PX16									

TEST No.	TARGET TIME	VESSEL No.	DATE IN	DATE OUT	RXN TIME	SA, mm2	EJ-13, g	SA/V /m	VESSEL IN	VESSEL OUT	CHANGE, g
PW1	14		not run			671					
PW2	14		not run			666					
PW3	28		not run			673					
PW4	28		not run			669					
PW5	56		not run			669					
PW6	56		not run			671					
PW7	91	41	7/1/91	9/30/91	91	676	2.06	328	290.16	289.85	-0.31
PW8	91	54	7/1/91	9/30/91	91	672	2.05	328	291.95	291.82	-0.13
PW9	TBD	269	7/1/91			669	2.05	326	325.06		
PW10	TBD	173	7/1/91			665	2.05	324	292.88		
PW11	360	299	7/1/91	6/25/92	360	672	2.05	328	297.82	297.42	-0.4
PW12	360	178	7/1/91	6/25/92	361	671	2.05	327	293.91	293.78	-0.13
PW13	540	344	7/1/91	12/22/92	540	675	2.05	329	294.93	294.44	-0.49
PW14	540	309	7/1/91	12/22/92	540	668	2.05	326	292.88	292.78	-0.1
PW15	720	358	7/1/91	6/20/93	715	676	2.05	330	298.91	298.47	-0.44
PW16	720	320	7/1/91	6/20/93	715	668	2.05	326	326.44	326.38	-0.06
PW17	180		not run			671					
PW18	180		not run			681					

TEST No.	pH	UF LPE	+UF SOLN	+DIW+HNO3	UF DF
PW1					
PW2					
PW3					
PW4					
PW5					
PW6					
PW7	9.83	11.01	12.28	10.12	8.9685
PW8	4.3	10.9	11.96	10.29	10.708
PW9					
PW10					
PW11	9.51	10.87	11.42	15.07	28.4
PW12	6.54	11	12.2	15.09	13.575
PW13	7.56	11.02	11.66	5.06	8.9063
PW14	5.15	10.84	12.16	5.05	4.8258
PW15	7.93	10.9	11.85	9.68	11.189
PW16	8.31	11.07	11.98	9.74	11.703
PW17					
PW18					

TEST No.	TARGET TIME	VESSEL No.	DATE IN	DATE OUT	RXN TIME, d	SAMPLE IN	EJ-13, g	SA/V, /m	VESSEL IN	VESSEL OUT	CHANGE, g
PZ1	3	359	8/28/90	8/31/90	3	5.0469	5.05	19988	334.48	334.46	-0.02
PZ2	3	380	8/28/90	8/31/90	3	5.0583	5.04	20073	328.77	328.76	-0.01
PZ3	7	381	8/28/90	9/4/90	7	5.023	5.02	20012	328.97	328.94	-0.03
PZ4	7	382	8/28/90	9/4/90	7	5.0365	5.05	19947	328.14	328.11	-0.03
PZ5	14	383	8/28/90	9/11/90	14	4.9886	5	19954	330.56	330.52	-0.04
PZ6	14	384	8/28/90	9/11/90	14	5.0328	5.09	19775	328.89	328.82	-0.07
PZ7	28	145	10/8/90	9/27/90	28	4.9982	5.01	19953	326.38	326.32	-0.06
PZ8	28	156	10/8/90	9/27/90	28	4.9922	4.99	20009	327.67	327.65	-0.02
PZ9	56	353	10/8/90	12/3/90	56	5.0169	5.03	19948	328.64	328.13	-0.51
PZ10	56	354	10/8/90	12/3/90	56	5.0138	5.03	19936	328.82	328.17	-0.65
PZ11	98	383	10/8/90	1/14/91	98	4.998	4.99	20032	329.25	329.2	-0.05
PZ12	98	384	10/8/90	1/14/91	98	5.0046	4.99	20059	328.4	328.29	-0.11
PZ13	182	366	8/15/90	2/13/91	182	4.9839	4.98	20016	330.14	329.91	-0.23
PZ14	182	367	8/15/90	2/13/91	182	4.9914	5.01	19926	328.32	328.27	-0.05
PZ15	364	368	8/15/90	8/14/91	364	4.9962	5.02	19905	298.9	298.38	-0.52
PZ16	364	369	8/15/90	8/14/91	364	5.0273	5.02	20029	329.54	329.29	-0.25
PZ17	728	370	8/15/90	8/12/92	728	5.0247	5.05	19900	331.55	330.86	-0.69
PZ18	728	371	8/15/90	8/12/92	728	5.0703	5.09	19923	337.17	336.87	-0.3
PZ19	TBD	297	2/7/91	8/12/92	552	5.0131	4.99	20093	328.49	328.41	-0.08
PZ20	TBD	244	2/7/91	2/11/94	1100	4.9938	5	19975	331.44	331.38	-0.06

TEST No.	pH	TEST NO.	F.45 LPE	+F.45 SOLN	+DIW+HNO3	F.45 DF	F50 LPE	+F50 SOLN	+DIW+HNO3	F50 DF
PZ1	11.94	PZ1	24.47	26.6	14.59	7.8498				
PZ2	11.86	PZ2	24.19	26.34	14.98	7.9674				
PZ3	11.99	PZ3	24.28	26.28	15.12	8.56				
PZ4	11.98	PZ4	24.36	26.26	15.19	8.9947				
PZ5	12.05	PZ5	24.47	26.36	15.64	9.2751				
PZ6	12.04	PZ6	24.08	25.99	15.06	8.8848				
PZ7	12.12	PZ7	24.18	25.89	14.96	9.7485				
PZ8	12.11	PZ8	24.15	25.86	14.94	9.7368				
PZ9	12.12	PZ9	24.44	25.47	15.09	15.651				
PZ10	12.14	PZ10	24.24	25.36	15.06	14.446				
PZ11	12.14	PZ11	24.41	26.48	15.08	8.285				
PZ12	12.12	PZ12	24.43	26.46	15.02	8.399				
PZ13	12.18	PZ13	7.73	8.11	4.68	13.316	7.74	8.57	4.79	6.7711
PZ14	12.19	PZ14	7.76	8.45	4.68	7.7826	7.74	8.48	4.8	7.4865
PZ15	12.2	PZ15	11.02	11.56	10.08	19.667	11.04	12.17	10.03	9.8761
PZ16	12.17	PZ16	11.03	11.56	10.08	20.019	11.07	11.89	10.04	13.244
PZ17	12.02	PZ17	10.83	11.41	15.05	26.948	10.96	11.38	15.05	36.833
PZ18	12.05	PZ18	10.88	11.61	15.08	21.658	11.1	12.12	15.16	15.863
PZ19	12.04	PZ19	10.97	11.93	14.97	16.594	10.97	11.92	15.15	16.947
PZ20	12.18	PZ20	10.99	12.41	14.15	10.965	11.04	11.31	11.71	44.37

TEST No.	TARGET TIME	VESSEL No.	DATE IN	DATE OUT	RXN TIME, d	SA, mm2	EJ-13, g	SA/V, /m	VESSEL IN	VESSEL OUT	CHANGE, g
TW1	14	1	6/26/91	7/10/91	14	703	2.05	343	36.77	36.78	0.01
TW2	14	2	6/26/91	7/10/91	14	710	2.03	350	36.44	36.51	0.07
TW3	28	3	6/26/91	7/24/91	28	704	2.04	345	36.95	37	0.05
TW4	28	4	6/26/91	7/24/91	28	699	2.05	341	36.5	36.48	-0.02
TW5	56	5	6/26/91	8/21/91	56	705	2.05	344	36.48	36.44	-0.04
TW6	56	6	6/26/91	8/21/91	56	699	2.06	339	36.14	36.13	-0.01
TW7	91	7	6/26/91	9/25/91	91	707	2.06	343	37.03	37	-0.03
TW8	91	8	6/26/91	9/25/91	91	708	2.06	344	37.05	37.05	0
TW9	180	9	6/26/91	12/23/91	181	708	2.05	345	36.69	36.7	0.01
TW10	180	10	6/26/91	12/23/91	181	693	2.05	338	36.11	36.14	0.03
TW11	360	11	6/26/91	6/20/92	360	706	2.05	344	37.11	37.16	0.05
TW12	360	12	6/26/91	6/20/92	360	701	2.05	342	36.67	36.67	0
TW13	540	13	6/29/91	12/18/92	540	700	2.05	341	36.59	36.6	-0.01
TW14	540	14	6/26/91	12/18/92	540	710	2.06	345	36.47	36.54	0.07
TW15	720	15	6/26/91	6/15/93	720	698	2.05	340	36.93	36.94	0.01
TW16	720	19	6/26/91	6/15/93	720	698	2.05	340	36.35	36.32	0.03
TW17	TBD	136	7/16/92			713	2.02	353	36.17		
TW18	TBD	137	7/16/92			699	2.02	346	36.16		

TEST No.	pH	UF LPE	+UF SOLN	+DIW+HN03	UF DF	F50 LPE	+F50 SOLN	+DIW+HN03	F50 DF
TW1	8.95	11	12.22	5.23	5.2869				
TW2	9.31	11.08	11.71	5.14	9.1587	11.08	11.52	5.11	12.614
TW3	9.24	11.03	11.48	10.06	23.356	11.04	11.67	10.08	17
TW4	9.28	11.04	11.58	10.06	19.63	11.08	11.89	10.08	13.444
TW5	9.5	11	12.44	10.04	7.9722				
TW6	9.44	11.08	12.93	10.04	7.9241				
TW7	9.36	11.15	11.52	10	28.027	7.8	8.53	10.09	14.822
TW8	9.41	8.02	9.55	10	7.5359				
TW9	9.4	10.88	11.4	14.98	29.808	10.87	11.7	14.93	18.988
TW10	9.58	11.04	11.58	15.09	28.944	11.06	11.93	14.99	18.23
TW11	9.63	10.87	12.28	15.28	11.837				
TW12	9.59	10.87	12	15.36	14.593				
TW13	10.48	10.93	12.33	5.04	4.6				
TW14	9.58	10.86	12.35	5.09	4.4161				
TW15	9.31	11.08	12.74	9.62	6.7952				
TW16	9.75	11.04	12.67	9.77	6.9939				
TW17									
TW18									

TEST No.	TARGET TIME	VESSEL No.	DATE IN	DATE OUT	RXN TIME, d	SA, mm2	EJ-13, g	SA/V, /m	VESSEL IN	VESSEL OUT	CHANGE, g
TW31	14	21	6/26/91	7/10/91	14	701	2.02	347	36.17	36.21	0.07
TW32	14	22	6/26/91	7/10/91	14	704	2.07	340	36.99	37.07	0.08
TW33	28	23	6/26/91	7/24/91	28	706	2.07	341	36.64	36.75	0.11
TW34	28	26	6/26/91	7/24/91	28	704	2.04	345	36.19	36.08	-0.11
TW35	56	27	6/26/91	8/21/91	56	706	2.05	344	36.53	36.56	-0.03
TW36	56	28	6/26/91	8/21/91	56	701	2.03	345	36.14	36.3	0.16
TW37	91	29	6/26/91	9/25/91	91	718	2.06	349	36.23	36.22	-0.01
TW38	91	30	6/26/91	9/25/91	91	697	2.05	340	36.07	36.06	-0.01
TW39	180	33	6/26/91	12/22/91	181	703	2.04	345	36.51	36.09	-0.42
TW40	180	34	6/26/91	12/22/91	181	706	2.05	344	36.56	36.59	0.03
TW41	360	35	6/26/91	6/20/92	360	706	2.03	348	36.51	36.56	0.05
TW42	360	36	6/26/91	6/20/92	360	704	2.03	347	37.01	37.04	0.03
TW43	540	37	6/29/91	12/18/92	540	696	2.05	340	36.61	36.74	0.13
TW44	540	38	6/26/91	12/18/92	540	709	2.03	349	37.05	37.2	-0.15
TW45	720	39	6/26/91	6/15/93	720	715	2.05	349	36.48	36.5	0.02
TW46	720	40	6/26/91	6/15/93	720	705	2.03	347	36.67	36.68	0.01
TW47	TBD	138	7/16/92			698	2.02	346	36.13		
TW48	TBD	139	7/16/92			699	2.04	343	36.23		

TEST No.	pH	UF LPE	+UF SOLN	+DIW+HN03	UF DF	F50 LPE	+F50 SOLN	+DIW+HN03	F50 DF
TW31	9.38	11.05	12.35	5.14	4.9538				
TW32	9.23	11.11	12.05	5.12	6.4468	11.07	11.5	5.1	12.86
TW33	9.44	10.99	11.59	10.09	17.817	11.05	11.67	10.07	17.242
TW34	9.45	11.07	11.5	10.06	24.395	11	11.82	10.01	13.207
TW35	9.66	10.95	12.28	10.04	8.5489				
TW36	9.71	11.1	12.62	10.04	7.6053				
TW37	9.74	11.08	11.63	10	19.182	7.8	8.65	10.04	12.812
TW38	9.63	8.01	9.43	9.98	8.0282				
TW39	9.88	11.02	11.14	14.99	125.92	10.99	11.57	15.01	26.879
TW40	9.69	10.71	11.11	14.96	38.4	10.99	11.66	14.91	23.254
TW41	9.79	10.86	12.44	15.35	10.715				
TW42	9.79	10.93	12.45	15.29	11.059				
TW43	9.69	10.77	12.46	5.11	4.0237				
TW44	9.78	10.81	12.52	5.12	3.9942				
TW45	9.24	10.94	12.56	9.75	7.0185				
TW46	9.42	10.98	12.57	9.71	7.1069				
TW47									
TW48									

TEST No.	TARGET TIME	VESSEL NO.	DATE IN	DATE OUT	RXN TIME	SAMPLE IN	MASS DIW, g	SA/V, /m	VESSEL IN	VESSEL OUT	CHANGE, g
TY1	3	112	7/8/91	7/11/91	3	0.9492	10.09	1882	44.32	44.31	-0.01
TY2	3	113	7/8/91	7/11/91	3	0.9842	10.03	1963	44.12	44.15	0.03
TY3	7	128	3/25/92	4/1/92	7	0.949	10.05	1889	44.31	44.31	0
TY4	7	129	3/25/92	4/1/92	7	0.9577	10.04	1908	44.57	44.57	0
TY5	14	109	3/18/92	4/1/92	14	0.9675	10.05	1925	44.7	44.65	-0.05
TY6	14	124	3/18/92	4/1/92	14	0.9774	10.01	1953	44.19	44.19	0
TY7	30	119	3/2/62	4/1/92	28	0.9885	10.01	1975	44.09	44.19	0.1
TY8	30	114	3/2/92	4/1/92	28	0.9786	10.01	1955	44.12	42.27	0.15
TY9	70	115	1/22/92	4/1/92	56	0.9752	10.01	1949	44.6	44.58	-0.02
TY10	70	117	1/22/92	4/1/92	56	0.9743	10.02	1945	44.17	44.23	0.05
TY11	140	101	6/28/91	11/15/91	140	1.0016	9.99	2005	44.22	44.03	-0.19
TY12	140	102	6/28/91	11/15/91	140	0.9913	10.04	1975	44.25	43.84	0.41
TY13	280	103	6/28/91	4/4/92	280	1.0105	10.04	2013	44.2	44.12	0.08
TY14	280	104	6/28/91	4/4/92	280	1.0136	10.03	2021	44.26	44.17	-0.09
TY15	560	105	6/28/91	1/8/93	560	1.0133	10.05	2017	44.23	44.25	0.02
TY16	560	106	6/28/91	1/8/93	560	1.007	10.07	2000	44.25	44.23	-0.02
TY17	980	107	6/28/91	2/24/94	980	1.011	10.07	2008	44.37	44.13	-0.24
TY18	980	108	6/28/91	2/24/94	980	0.9797	10.07	1946	44.26	44.27	0.01
TY19	TBD	140	7/16/92			1.0078	10.07	2002	44.24		
TY20	TBD	141	7/16/92			1.0106	10.03	2015	44.32		
TY21(EJ-13)	TBD	142	7/16/92			1.0024	10.02	2001	44.32		
TY22(EJ-13)	TBD	143	7/16/92			0.9992	10.07	1989	44.17		

TEST No.	pH	F.45 LPE	+F.45 SOLN	+DIW+HN03	F.45 DF	F50 LPE	+F50 SOLN	+DIW+HN03	F50 DF
TY1	9.96	10.92	15.97	5.12	2.0139	11.01	14.93	5.1	2.301
TY2	9.93	10.96	14.7	5.12	2.369	11.12	14.88	5.1	2.3564
TY3	9.99	11.01	18.03	10.03	2.4288	10.85	12.32	13.89	10.449
TY4	10.07	10.8	17.8	8.08	2.1543	10.75	12.15	15.05	11.75
TY5	10.02	11	18	8.66	2.2371	11.03	12.27	14.78	12.919
TY6	9.97	10.97	17.44	9.66	2.493	10.74	12.21	15.06	11.245
TY7	10.24	10.92	17.94	10.05	2.4316	10.93	12.4	15.04	11.231
TY8	10.12	10.75	17.92	8.22	2.1464	10.97	12.42	14.97	11.324
TY9	10.32	10.89	12.43	14.48	10.403	10.86	12.32	15.02	11.288
TY10	10.33	10.86	12.71	14.3	8.7297	11.03	12.44	15.01	11.645
TY11	10.17	11.02	18.83	10	2.2804	10.97	11.62	14.96	24.015
TY12	10.28	11.04	18.57	9.95	2.3214	10.91	11.83	14.95	17.25
TY13	10.39	10.94	12.6	15.01	10.042	10.79	12.26	15.01	11.211
TY14	10.31	10.79	12.63	15.09	9.2011	10.78	12.35	14.93	10.51
TY15	10.11	10.95	16.54	5.01	1.8962	11.01	14.35	4.97	2.488
TY16	10.29	10.96	14.36	4.99	2.4676	11.02	13.7	4.91	2.8321
TY17	9.95	11.02	16.84	10.03	2.7234	11.07	13.39	10.07	5.3405
TY18	9.84	11.06	16.85	10.05	2.7358	10.9	13.21	10.03	5.342
TY19									
TY20									
TY21									
TY22									

TEST No.	TARGET TIME	VESSEL No.	DATE IN	DATE OUT	RXN TIME, d	SAMPLE IN	DIW, g	SA/V, /m	VESSEL IN	VESSEL OUT	CHANGE, g
TZ1	3	41	7/8/91	7/11/91	3	4.9913	5.03	19846	43.73	43.73	0
TZ2	3	42	7/8/91	7/11/91	3	4.9969	5.01	19948	43.65	43.69	0.04
TZ3	7	43	6/26/91	7/3/91	7	5.0338	5.03	20015	43.62	43.58	-0.04
TZ4	7	44	6/26/91	7/3/91	7	4.9978	4.98	20071	43.62	43.61	-0.01
TZ5	14	45	6/26/91	7/10/91	14	5.0089	4.99	20076	44.1	44.28	0.18
TZ6	14	46	6/26/91	7/10/91	14	4.9973	4.99	20029	43.45	43.56	0.11
TZ7	28	47	6/26/91	7/24/91	28	5.0202	4.99	20121	44.29	44.37	0.18
TZ8	28	48	6/26/91	7/24/91	28	4.9914	4.98	20046	43.16	43.25	0.09
TZ9	56	49	6/26/91	8/21/91	56	5.013	4.99	20092	43.59	43.81	0.22
TZ10	56	50	6/26/91	8/21/91	56	5.0052	4.99	20061	43.77	43.79	0.02
TZ11	91	51	6/26/91	9/25/91	91	5.0083	4.97	20154	43.02	43.09	0.07
TZ12	91	52	6/26/91	9/25/91	91	4.9979	4.99	20032	44.24	44.35	0.11
TZ13	182	53	6/26/91	12/23/91	181	5.0157	4.98	20143	44.1	44.22	0.12
TZ14	182	54	6/26/91	12/23/91	181	5.0057	4.98	20103	43.93	44.08	0.15
TZ15	364	55	6/26/91	6/24/92	364	4.9949	4.99	20020	44.07	44.08	0.01
TZ16	364	56	6/26/91	6/24/92	364	4.9962	5	19985	43.7	43.87	0.17
TZ17	TBD	132	7/16/92			4.9915	5.02	19886	43.52		
TZ18	TBD	133	7/16/92			5.0145	5.03	19938	43.56		

TEST No.	pH	F.45 LPE	+F.45 SOLN	+DIW+HN03	F.45 DF	F50 LPE	+F50 SOLN	+DIW+HN03	F50 DF
TZ1	11.09	11.05	12.96	10.12	6.2984	11.1	12.02	5.11	6.5543
TZ2	11.14	10.95	12.91	10.14	6.1735	10.96	11.92	5.09	6.3021
TZ3	10.93	10.98	14.01	10.01	4.3036				
TZ4	11.21	10.98	12.81	10.06	6.4973	10.95	12.03	5.02	5.6481
TZ5	11.34	11.05	13.82	10.13	4.657				
TZ6	11.33	10.07	13.56	10.12	3.8997	10.95	11.38	5.12	12.907
TZ7	11.49	11.06	12.52	10.03	7.8699	10.92	11.99	10.05	10.393
TZ8	11.46	10.96	11.6	10.07	16.734	11.13	13.21	10.04	5.8269
TZ9	11.46	11.13	14.26	10.04	4.2077				
TZ10	11.5	10.92	13.66	10.02	4.6569				
TZ11	11.43	11.04	12.15	10	10.009	7.7	9.16	10	7.8493
TZ12	11.5	11.09	12.27	10	9.4746	8.03	9.47	10.02	7.9583
TZ13	11.62	10.77	12.64	14.9	8.9679	11.05	11.98	14.96	17.086
TZ14	11.69	10.91	12.84	15.03	8.7876	10.87	11.82	14.95	16.737
TZ15	11.72	11	11.43	15.31	36.605	10.97	12.72	15.22	9.6971
TZ16	11.76	10.88	11.65	15.24	20.792	10.88	12.41	15.32	11.013
TZ17									
TZ18									

TEST No.	TARGET TIME	VESSEL No.	DATE IN	DATE OUT	RXN TIME, d	SAMPLE IN	EJ-13, g	SA/V, /m	VESSEL IN	VESSEL OUT	CHANGE, g
TZ31	3	57	7/8/91	7/11/91	3	5.0103	5.06	19804	44.08	44.07	-0.01
TZ32	3	58	7/8/91	7/11/91	3	5.0162	5.02	19985	43.62	43.63	0.01
TZ33	7	59	6/26/91	7/3/91	7	5.0151	5.04	19901	43.67	43.7	0.03
TZ34	7	60	6/26/91	7/3/91	7	5.0156	5.11	19631	44.14	44.2	0.06
TZ35	14	61	6/26/91	7/10/91	14	5.0136	5.01	20014	43.45	43.52	0.07
TZ36	14	62	6/26/91	7/10/91	14	5.0014	4.99	20046	44.04	44.14	0.1
TZ37	28	63	6/26/91	7/24/91	28	5.001	5.01	19964	43.72	43.86	0.14
TZ38	28	64	6/26/91	7/24/91	28	4.9993	5	19997	43.79	43.94	0.15
TZ39	56	65	6/26/91	8/21/91	56	5.001	5.04	19845	43.58	43.59	0.01
TZ40	56	66	6/26/91	8/21/91	56	4.9999	5.05	19802	43.23	43.22	-0.01
TZ41	91	67	6/26/91	9/25/91	91	5.015	5.05	19861	43.91	43.96	0.05
TZ42	91	68	6/26/91	9/25/91	91	5.0058	5.04	19864	44.29	44.38	0.09
TZ43	182	69	6/26/91	12/23/91	181	4.9975	5.06	19753	43.8	43.91	0.11
TZ44	182	70	6/26/91	12/23/91	181	5.0035	5.06	19777	43.6	43.76	0.16
TZ45	364	71	6/26/91	6/24/92	364	5.0038	5.07	19739	43.22	43.23	0.01
TZ46	364	72	6/26/91	6/24/92	364	5.0069	5.06	19790	44.01	44.08	0.07
TZ47	TBD	134	7/16/92		230	5.006	5.01	19984	43.13		
TZ48	TBD	135	7/16/92		230	4.9911	4.99	20004	43.18		

TEST No.	pH	F.45 LPE	+F.45 SOLN	+DIW+HNO3	F.45 DF	F50 LPE	+F50 SOLN	+DIW+HNO3	F50 DF
TZ31	11.02	11.02	12.94	10.13	6.276	11.11	11.98	5.08	6.8391
TZ32	11.04	11.04	12.86	10.06	6.5275	10.96	11.92	5.09	6.3021
TZ33	11.03	10.92	12.71	10.03	6.6034	10.89	11.99	5.02	5.5636
TZ34	11.01	10.98	14.01	10.05	4.3168				
TZ35	11.23	10.98	13.52	10.09	4.9724				
TZ36	11.28	10.87	13.32	10.14	5.1388	11.09	11.51	5.11	13.167
TZ37	11.37	11.02	12.04	10.21	11.01	11.13	12.91	10.05	6.6461
TZ38	11.38	10.98	11.94	10.11	11.531	11.09	12.83	10.09	6.7989
TZ39	11.37	10.94	13.84	10.02	4.4552				
TZ40	11.41	11.01	13.96	10.04	4.4034				
TZ41	11.4	10.97	12.19	9.97	9.1721	7.75	9.18	10	7.993
TZ42	11.38	11.07	12.4	9.96	8.4887	8.05	9.5	9.97	7.8759
TZ43	11.63	11.03	12.99	15	8.6531	11.04	12.01	14.95	16.412
TZ44	11.66	10.93	12.89	14.98	8.6429	10.94	11.86	14.94	17.239
TZ45	11.09	10.86				10.88	12.41	15.32	11.013
TZ46	11.75	10.97	11.52	15.18	28.6	10.88	12.58	15.35	10.029
TZ47									
TZ48									

Table D.3. Cation Results

<u>Column Heading</u>	<u>Comment</u>
TEST NUMBER	Test number in test matrix (Table D.1). Corresponds to unfiltered leachate in tests with monolithic samples and to leachates filtered through 0.45 μm filters in tests with crushed glass samples.
-UF	Unfiltered solution.
-F25	Solution filtered using Amicon model CF25A filter.
-F50	Solution filtered using Amicon model CF50A filter.
RXN TIME	Actual reaction time, in days.
pH	Leachate pH measured at room temperature.
DIL FACTOR	Dilution factor for solutions.
SA/V	Surface area-to-leachant volume ratio, in inverse meters.
LEACHANT	Mass of EJ-13 solution or deionized water added as leachant, in grams.
ACL NUMBER	Solution identification number.
i, ACL	Analytical result for species i in submitted solution, in parts per million.
i, ppm	Concentration of species i corrected for dilution, in parts per million.
NL(i), g/m ²	Normalized mass loss based on element i, in grams per square meter.
f(i)	Elemental weight fraction of species i in the glass.
BKGND	Average concentration of species measured in blank tests, in parts per million.

TEST NUMBER	RXN TIME	pH	DIL FACTOR	SA/V	LEACHANT VO	ACL NUMBER	Al, ACL	Al, ppm	NL(Al), g/m	B, ACL	B, ppm
BX1	7	8.21	1.694	10.01	16.62	90-0561-01	0.54	0.91476	1.7458	0.11	0.18634
BX2	7	8.2	1.689	10	16.66	90-0561-02	0.52	0.87828	1.5679	0.1	0.1689
BX3	28	8.28	1.6762	9.99	17.42	90-0561-03	0.49	0.82134	1.2887	0.12	0.20114
BX4	28	8.34	1.707	10	16.9	90-0561-04	0.42	0.71694	0.7731	0.11	0.18777
BX5	94	8.26	1.6726	10	17.41	91-0093-05	0.2	0.33452	-1.1107	0.65	1.0872
BX6	94	8.59	1.716	10	16.48	91-0093-06	0.3	0.5148	-0.22266	0.13	0.22308
BX7	238	8.43	1.5657	9.99	164	91-0346-01	0.35	0.548	-0.059197	0.16	0.25051
BX7-F50			2.805			91-0346-03	0.21	0.58905		0.13	0.36465
BX8	238	9.04	1.5219	10.02	15.99	91-0346-02	0.37	0.5631	0.015255	1.69	2.572
BX8-F50			2.6755			91-0346-04	0.22	0.58861		1.06	2.836
BX9	600	8.57	1.4838	10.01	16.67	92-0353-01	0.33	0.48965	-0.34619	2.49	3.6947
BX9-F50			5.7874			92-0353-02	-0.1	-0.57874		0.62	3.5882
BX10	600	8.58	2.0096	10.01	16.93	92-0353-03	0.26	0.5225	-0.18456	1.44	2.8938
BX10-F50			5.1468			92-0353-04	-0.1	-0.51468		0.55	2.8307
BKGND			1				0.56	0.56		0.15	0.15
f(i)			1				0.0203	0.0203		0.0248	0.0248

TEST NO.	NL(B), g/m2	Ba, ACL	Ba, ppm	NL(Ba), g/m	Ca, ACL	Ca, ppm	NL(Ca), g/m	Cr, ACL	Cr, ppm	NL(Cr), g/m	Cu, ACL
BX1	0.14639	-0.02	-0.03388	-1.6923	3.16	5.353	9.096	-0.02	-0.03388	-6.7692	0.06
BX2	0.07621	-0.02	-0.03378	-1.689	3.17	5.3541	9.1178	-0.02	-0.03378	-6.756	0.2
BX3	0.20643	-0.02	-0.033524	-1.6779	3.24	5.4309	10.02	-0.02	-0.033524	-6.7115	0.25
BX4	0.1523	-0.02	-0.03414	-1.707	3.1	5.2917	8.3919	-0.02	-0.03414	-6.828	0.06
BX5	3.779	-0.02	-0.033452	-1.6726	2.65	4.4324	-1.6001	-0.02	-0.033452	-6.6904	4.82
BX6	0.29468	-0.02	-0.03432	-1.716	2.77	4.7533	2.1316	-0.02	-0.03432	-6.864	0.26
BX7	0.4057	-0.02	-0.031314	-1.5673	2.29	3.5855	-11.46	-0.02	-0.031314	-6.2691	0.18
BX7-F50		-0.02	-0.0561	0	1.1	3.0855		-0.02	-0.0561	0	-0.02
BX8	9.7467	-0.02	-0.030438	-1.5189	2.09	3.1808	-16.122	-0.02	-0.030438	-6.0754	0.34
BX8-F50		-0.02	-0.05351	0	1.16	3.1036		-0.02	-0.05351	0	-0.02
BX9	14.279	0.02	0.029676	1.4823	1.76	2.6115	-22.751	-0.02	-0.029676	-5.9293	0.25
BX9-F50		-0.02	-0.11575		0.37	2.1413		-0.02	-0.11575		-0.02
BX10	11.053	-0.02	-0.040192	-2.0076	2.05	4.1197	-5.2311	-0.02	-0.040192	-8.0304	0.28
BX10-F50		-0.02	-0.10294		0.62	3.191		-0.02	-0.10294		-0.02
BKGND		0	0		4.57	4.57		0	0		0
f(i)		0.002	0.002		0.0086	0.0086		0.0005	0.0005		0.0032

TEST NO.	Cu, ppm	NL(Cu), g/m	Fe, ACL	Fe, ppm	NL(Fe), g/m	K, ACL	K, ppm	NL(K), g/m2	Li, ACL	Li, ppm	NL(Li), g/m
BX1	0.10164	3.1731	0.03	0.05082	0.063621	4.38	7.4197	1.2276	0.08	0.13552	0.43368
BX2	0.3378	10.556	0.03	0.05067	0.063496	4.46	7.5329	1.5953	0.08	0.13512	0.43208
BX3	0.41905	13.108	0.03	0.050286	0.063078	4.48	7.5094	1.5205	0.12	0.20114	0.768
BX4	0.10242	3.2006	0.07	0.11949	0.14974	4.42	7.5449	1.6341	0.11	0.18777	0.69934
BX5	8.0619	251.94	0.21	0.35125	0.44016	5.11	8.547	4.877	0.52	0.86975	4.1612
BX6	0.44616	13.942	0.1	0.1716	0.21504	4.32	7.4131	1.2075	0.15	0.2574	1.0528
BX7	0.28183	8.8159	0.02	0.031314	0.03928	4.9	7.6719	2.0471	0.2	0.31314	1.3371
BX7-F50	-0.0561	0	-0.02	-0.0561							
BX8	0.51745	16.138	0.15	0.22829	0.2855	6.78	10.318	10.589	1.32	2.0089	9.9238
BX8-F50	-0.05351	0	0.04	0.10702							
BX9	0.37095	11.581	0.18	0.26708	0.33436	7.82	11.603	14.753	1.88	2.7895	13.892
BX9-F50	-0.11575		-0.02	-0.11575		2.05	11.864		0.45	2.6043	
BX10	0.56269	17.566	0.23	0.46221	0.57863	5.15	10.349	10.699	1.05	2.1101	10.447
BX10-F50	-0.10294		-0.02	-0.10294		2.2	11.323		0.4	2.0587	
BKGND	0		0	0		7.04	7.04		0.05	0.05	
f(i)	0.0032		0.0798	0.0798		0.0309	0.0309		0.0197	0.0197	

TEST NO.	Mg, ACL	Mg, ppm	NL(Mg), g/m	Mn, ACL	Mn, ppm	NL(Mn), g/m	Na, ACL	Na, ppm	NL(Na), g/m	Ni, ACL	Ni, ppm
BX1	0.24	0.40656	3.8282	0.09	0.15246	1.0879	33.2	56.241	7.437	-0.03	-0.05082
BX2	0.24	0.40536	3.817	0.06	0.10134	0.72386	33.3	56.244	7.4489	-0.03	-0.05067
BX3	0.23	0.38553	3.5726	0.09	0.15086	1.0786	34.3	57.494	9.3492	-0.03	-0.050286
BX4	0.1	0.1707	0.88375	0.05	0.08535	0.60964	33.5	57.185	8.8722	-0.03	-0.05121
BX5	0.069	0.11541	0.19262	0.095	0.1589	1.135	33.8	56.534	7.8879	0.05	0.08363
BX6	0.065	0.11154	0.14425	0.038	0.065208	0.46577	31.9	54.74	5.1746	0.03	0.05148
BX7	0.12	0.18788	1.0996	0.06	0.093942	0.67169	36.1	56.522	7.8774	-0.03	-0.046971
BX7-F50	0.11	0.30855		0.02	0.0561		22.3	62.551		-0.03	-0.08415
BX8	0.01	0.015219	-1.0576	0.04	0.060876	0.43396	40.7	61.941	16.037	-0.03	-0.045657
BX8-F50	0.12	0.32106		-0.01	-0.026755		26	69.563		-0.03	-0.080265
BX9	0.02	0.029676	-0.87817	0.08	0.1187	0.84704	45	66.771	23.352	0.03	0.044514
BX9-F50	0.03	0.17362		-0.01	-0.057874		11.6	67.134		-0.03	-0.17362
BX10	0.02	0.040192	-0.74685	0.06	0.12058	0.8604	32	64.307	19.628	-0.03	-0.060288
BX10-F50	0.03	0.1544		-0.01	-0.051468		12.5	64.335		-0.03	-0.1544

BKGND	0.1	0.1		0	0		51.32	51.32		0	0
f(i)	0.008	0.008		0.014	0.014		0.0661	0.0661		0.0065	0.0065

TEST NO.	NL(Ni), g/m	Si, ACL	Si, ppm	NL(Si), g/m	Th, ACL	Th, ppm	NL(Th), g/m	Ti, ACL	Ti, ppm	NL(Ti), g/m	U, ACL
BX1	-0.78107	26.7	45.23	-8.6649e-05	-0.06	-0.10164	-4.4147				0.00434
BX2	-0.77954	27	45.603	0.1631	-0.06	-0.10134	-4.4061				0.00379
BX3	-0.77441	27.1	45.425	0.08536	-0.06	-0.10057	-4.3771				0.00355
BX4	-0.78785	26.5	45.236	0.0024052	-0.06	-0.10242	-4.453				0.00417
BX5	1.2866	31.2	52.185	3.0412	-0.1	-0.16726	-7.2722	-0.01	-0.016726	0	0.0135
BX6	0.792	26.8	45.989	0.33179	-0.1	-0.1716	-7.4609	-0.01	-0.01716	0	0.0172
BX7	-0.72335	27.8	43.526	-0.74563	-0.05	-0.078285	-3.4071	-0.02	-0.031314	0	0.011
BX7-F50		15.4	43.197		-0.05	-0.14025	0	-0.02	-0.0561	0	0.00496
BX8	-0.70101	37.3	56.767	5.0345	-0.05	-0.076095	-3.3019	-0.02	-0.030438	0	0.12
BX8-F50		21.3	56.988		-0.05	-0.13377	0	-0.02	-0.05351	0	0.0362
BX9	0.68415	42.4	62.913	7.7243	0.29	0.4303	18.69	-0.02	-0.029676	0	0.0206
BX9-F50		10.8	62.504		-0.05	-0.28937		-0.02	-0.11575	0	
BX10	-0.92658	30.8	61.896	7.2799	-0.05	-0.10048	-4.3643	-0.02	-0.040192	0	0.024
BX10-F50		11.5	59.188		-0.05	-0.25734		-0.02	-0.10294	0	

BKGND		45.23	45.23		0	0		0	0		0
f(i)		0.2287	0.2287		0.0023	0.0023		0.0054	0.0054		0.0164

TEST NO.	U, ppm	NL(U), g/m2	Zn, ACL	Zn, ppm	NL(Zn), g/m	Zr, ACL	Zr, ppm	NL(Zr), g/m
BX1	0.007352	0.044784				-0.02	0	0
BX2	0.0064013	0.039032				-0.02	0	0
BX3	0.0059505	0.03632				-0.02	0	0
BX4	0.0071182	0.043404				-0.02	0	0
BX5	0.02258	0.13768	0.014	0.0234	11.7	-0.02	0	0
BX6	0.029515	0.17997	0.03	0.0515	15.75	-0.02	0	0
BX7	0.017223	0.10512	0.05	0.078285	39.182	-0.05	0	0
BX7-F50	0.013913		0.03	0.08415		-0.05		
BX8	0.18263	1.1114	0.09	0.13697	68.349	-0.05	0	0
BX8-F50	0.096853		0.04	0.10702		-0.05		
BX9	0.030566	0.18619	-0.02	-0.029676	-14.823	-0.05		
BX9-F50			0.04	0.2315		-0.05		
BX10	0.04823	0.29379	0.07	0.14067	70.266	-0.05		
BX10-F50			0.05	0.25734		-0.05		

BKGND	0		0	0		0		
f(i)	0.0164		0.0002	0.0002		0.0007		

TEST No.	RXN TIME	pH	DIL FACTOR	SA/V	LEACHANT	ACL NUMBER	Al, ACL	Al, ppm	NL(Al),g/m2	B, ACL	B, ppm
BY1	3	9.27	2.3364	1986	10	91-0061-01	0.67	1.5654	0.024938	2.02	4.7195
BY2	3	9.3	2.3441	2049	9.96	91-0061-02	0.69	1.6174	0.025422	1.83	4.2897
BY3	7	9.48	2.3289	1947	10	90-0513-06	1.03	2.3988	0.046523	3.62	8.4306
BY4	7	9.48	2.3757	2046	9.99	90-0513-07	1.03	2.447	0.045432	3.96	9.4077
BY5	14	9.77	2.3396	2004	9.95	90-0487-18	1.15	2.6905	0.052372	5.8	13.57
BY6	14	9.74	2.3568	1967	10.05	90-0487-19	1.19	2.8046	0.056213	5.36	12.632
BY7	30	9.94	2.3527	2041	10.01	90-0487-10	1.35	3.1761	0.063143	8.36	19.669
BY8	30	9.87	2.3518	1983	10.09	90-0487-11	1.36	3.1984	0.065544	8.62	20.273
BY9	70	10.09	2.2805	1993	10.29	91-0205-01	0.86	1.9612	0.034634	11.1	25.314
BY10	70	10.11	2.3765	1981	10.02	91-0205-02	1.2	2.8518	0.05699	9.45	22.458
BY11-UF			9.6541			91-0310-01	0.49	4.7305		3.39	32.727
BY11	140	10.02	6.6709	1997	9.96	91-0310-02	0.56	3.7357	0.078337	4.37	29.152
BY11-F50			3.3648			91-0310-03	0.66	2.2208		8.67	29.173
BY12-UF			9.6752			91-0310-04	0.48	4.6441		3.44	33.283
BY12	140	10.11	6.475	2004	10.22	91-0310-05	0.56	3.626	0.075367	4.36	28.231
BY12-F50			3.3057			91-0310-06	0.72	2.3801		8.92	29.487
BY13	280	10.47	2.6038	1959	10.03	91-0346-05	1.41	3.6714	0.078238	13	33.849
BY14	280	10.45	2.5802	1996	10	91-0346-06	1.35	3.4833	0.072146	12.8	33.027
BY15-UF			11.12			92-0134-01	0.41	4.5592		4.14	46.037
BY15	560	10.33	4.159	2017	9.99	92-0134-02	0.93	3.8679	0.080788	9.89	41.133
BY15-F50			11.12			92-0134-03	0.16	1.7792		3.55	39.476
BY16-UF			10.838			92-0134-04	0.46	4.9855		4.61	49.963
BY16	560	10.42	4.3245	1999	10.02	92-0134-05	0.89	3.8488	0.081046	9.49	41.04
BY16-F50			10.838			92-0134-06	0.16	1.7341		3.69	39.992
BY17	980	10.52	4.337	1893	10.01	PACKET.1	0.337	1.462	0.023473	10.9	47.27
BY17-F50			4.907			PACKET.1	0.457	2.242		8.37	41.07
BY18	980	10.65	4.2876	1990	10	PACKET.1	0.338	1.449	0.022007	10.5	45.02
BY18-F50			4.877			PACKET.1	0.286	1.395		8.81	42.97
BKGND			1				0.56	0.56		0.15	0.15
f(i)			1				0.0203	0.0203		0.0248	0.0248

TEST NO.	NL(B), g/m2	Ba, ACL	Ba, ppm	NL(Ba), g/m	Ca, ACL	Ca, ppm	NL(Ca), g/m	Cr, ACL	Cr, ppm	NL(Cr), g/m	Cu, ACL
BY1	0.092777	-0.02	-0.046728	-0.011764	0.32	0.74765	-0.2238	-0.02	-0.046728	-0.047057	0.09
BY2	0.081466	-0.02	-0.046882	-0.01144	0.38	0.89076	-0.20879	-0.02	-0.046882	-0.045761	0.08
BY3	0.17149	-0.01	-0.023289	-0.0059807	0.16	0.37262	-0.25068	-0.02	-0.046578	-0.047846	0.1
BY4	0.18245	-0.01	-0.023757	-0.0058057	0.15	0.35635	-0.23947	-0.02	-0.047514	-0.046445	0.12
BY5	0.27002	-0.01	-0.023396	-0.0058373	0.14	0.32754	-0.24616	0.02	0.046792	0.046699	0.14
BY6	0.25588	-0.01	-0.023568	-0.0059908	0.13	0.30638	-0.25204	0.024	0.056563	0.057512	0.23
BY7	0.38561	0.011	0.02588	0.00634	0.13	0.30585	-0.24294	0.045	0.10587	0.10374	0.21
BY8	0.40917	0.012	0.028222	0.0071159	0.12	0.28222	-0.25143	0.11	0.2587	0.26092	0.14
BY9	0.50911	-0.02	-0.04561	-0.011443	0.07	0.15963	-0.25732	0.11	0.25085	0.25174	0.05
BY10	0.45407	-0.02	-0.04753	-0.011996	0.13	0.30894	-0.25011	0.07	0.16635	0.16795	0.19
BY11-UF		-0.02	-0.19308		-0.02	-0.19308		0.09	0.86887		0.1
BY11	0.58559	-0.02	-0.13342	-0.033405	0.05	0.33354	-0.24668	0.12	0.80051	0.80171	0.09
BY11-F50		-0.02	-0.067296		0.44	1.4805		0.21	0.70661		0.04
BY12-UF		-0.02	-0.1935		-0.02	-0.1935		0.08	0.77402		0.08
BY12	0.56502	-0.02	-0.1295	-0.03231	0.06	0.3885	-0.24263	0.1	0.6475	0.64621	0.09
BY12-F50		-0.02	-0.066114		0.37	1.2231		0.18	0.59503		0.03
BY13	0.69364	-0.02	-0.052076	-0.013291	0.13	0.33849	-0.25117	0.22	0.57284	0.58482	0.22
BY14	0.66416	-0.02	-0.051604	-0.012927	0.14	0.36123	-0.24519	0.41	1.0579	1.06	0.19
BY15-UF		-0.02	-0.2224		0.11	1.2232		0.35	3.892		0.12
BY15	0.8193	-0.02	-0.08318	-0.02062	0.34	1.4141	-0.18194	0.82	3.4104	3.3816	0.19
BY15-F50		-0.02	-0.2224		0.37	4.1144		0.29	3.2248		-0.02
BY16-UF		-0.02	-0.21676		0.1	1.0838		0.26	2.8179		0.09
BY16	0.8248	-0.02	-0.08649	-0.021633	0.26	1.1244	-0.20043	0.53	2.292	2.2931	0.17
BY16-F50		-0.02	-0.21676		0.73	7.9117		0.2	2.1676		-0.02
BY17	1.0037	0.009	0.039	0.010301	0.884	3.834	-0.045209	0.421	1.826	1.9292	0.005
BY17-F50		0.004	0.02		0.88	4.318		0.353	1.732		0.015
BY18	0.90918	0.01	0.043	0.010804	0.929	3.984	-0.034241	0.789	3.383	3.4	0.006
BY18-F50		0.003	0.015		0.883	4.306		0.663	3.233		0.008
BKGND		0	0		4.57	4.57		0	0		0
F(i)		0.002	0.002		0.0086	0.0086		0.0005	0.0005		0.0032

TEST NO.	Cu, ppm	NL(Cu), g/m	Fe, ACL	Fe, ppm	NL(Fe), g/m	K, ACL	K, ppm	NL(K), g/m2	Li, ACL	Li, ppm	NL(Li), g/m
BY1	0.21028	0.033087	0.18	0.42055	0.0026536	5.66	13.224	0.10077	3.6	8.411	0.21371
BY2	0.18753	0.028601	0.13	0.30473	0.0018637	5.33	12.494	0.086143	3.44	8.0637	0.19853
BY3	0.23289	0.03738	0.51	1.1877	0.0076446	6.81	15.86	0.1466	5.62	13.088	0.33993
BY4	0.28508	0.043543	0.59	1.4017	0.0085848	6.87	16.321	0.1468	5.83	13.85	0.34238
BY5	0.32754	0.051077	0.88	2.0588	0.012874	8.57	20.05	0.2101	7.69	17.992	0.45446
BY6	0.54206	0.086118	0.73	1.7205	0.010961	8.06	18.996	0.19671	7.36	17.346	0.44635
BY7	0.49407	0.075647	1.35	3.1761	0.019501	10.1	23.762	0.26515	9.83	23.127	0.57395
BY8	0.32925	0.051887	1.46	3.4336	0.021698	10.1	23.753	0.27276	9.85	23.165	0.59171
BY9	0.11402	0.017879	0.33	0.75257	0.0047319	12	27.366	0.33005	13.1	29.875	0.75963
BY10	0.45153	0.071229	1.61	3.8262	0.024203	10.7	25.429	0.3004	12.1	28.756	0.73556
BY11-UF	0.96541		0.78	7.5302		3.6	34.755		3.69	35.624	
BY11	0.60038	0.09395	0.88	5.8704	0.036837	4.45	29.686	0.36698	4.56	30.419	0.77195
BY11-F50	0.13459		0.05	0.16824		9.43	31.73		8.77	29.509	
BY12-UF	0.77402		0.65	6.2889		3.59	34.734		3.65	35.314	
BY12	0.58275	0.090873	0.7	4.5325	0.028342	4.59	29.72	0.36626	4.73	30.627	0.77451
BY12-F50	0.099171		0.03	0.099171		10.3	34.049		9.13	30.181	
BY13	0.57284	0.091379	2.06	5.3638	0.034311	13.4	34.891	0.46009	14.5	37.755	0.97701
BY14	0.49024	0.076753	2.09	5.3926	0.033856	13.6	35.091	0.4548	14.3	36.897	0.93707
BY15-UF	1.3344		0.88	9.7856		4.43	49.262		5.14	57.157	
BY15	0.79021	0.12243	1.97	8.1932	0.050903	10.03	41.715	0.55635	11.5	47.828	1.2024
BY15-F50	-0.2224		-0.02	-0.2224		3.46	38.475		4	44.48	
BY16-UF	0.97542		0.94	10.188		4.79	51.914		5.56	60.259	
BY16	0.73516	0.11493	1.84	7.9571	0.049881	9.96	43.072	0.58333	10.9	47.137	1.1957
BY16-F50	-0.21676		-0.02	-0.21676		3.61	39.125		4.08	44.219	
BY17	0.022	0.0036318	0.235	1.019	0.0067456	9.09	39.42	0.55356	11.7	50.74	1.3593
BY17-F50	0.074		0.009	0.044		8.48	41.61		9.87	48.43	
BY18	0.026	0.0040829	0.137	0.587	0.0036964	9.89	42.41	0.57521	12	51.46	1.3114
BY18-F50	0.039		-0.003	-0.015		7.83	38.19		9.76	47.6	
BKGND	0		0	0		7.04	7.04		0.05	0.05	
f(i)	0.0032		0.0798	0.0798		0.0309	0.0309		0.0197	0.0197	

TEST NO.	Mg, ACL	Mg, ppm	NL(Mg), g/m	Mn, ACL	Mn, ppm	NL(Mn), g/m	Na, ACL	Na, ppm	NL(Na), g/m	Ni, ACL	Ni, ppm
BY1	0.038	0.088783	-0.00070599	0.041	0.095792	0.0034453	32.9	76.868	0.19461	-0.03	-0.070092
BY2	0.032	0.075011	-0.0015245	0.02	0.046882	0.0016343	32.5	76.183	0.18358	-0.03	-0.070323
BY3	0.071	0.16535	0.0041957	0.054	0.12576	0.0046137	37.7	87.8	0.28345	-0.03	-0.069867
BY4	0.086	0.20431	0.0063727	0.067	0.15917	0.0055569	38	90.276	0.28805	-0.03	-0.07127
BY5	0.11	0.25736	0.0098151	0.11	0.25736	0.0091729	43.1	100.84	0.37381	0.038	0.088905
BY6	0.091	0.21447	0.0072743	0.092	0.21683	0.0078737	42.1	99.221	0.36842	0.071	0.16733
BY7	0.14	0.32938	0.014048	0.18	0.42349	0.014821	50.6	119.05	0.50201	0.045	0.10587
BY8	0.18	0.42332	0.020381	0.2	0.47036	0.016943	49.9	117.35	0.50379	0.053	0.12465
BY9	0.02	0.04561	-0.0034113	0.06	0.13683	0.0049039	59	134.55	0.63178	0.03	0.068415
BY10	0.19	0.45153	0.022182	0.3	0.71295	0.025707	55.5	131.9	0.61534	0.06	0.14259
BY11-UF	0.07	0.67579		0.13	1.255		18.9	182.46		0.03	0.28962
BY11	0.08	0.53367	0.027145	0.14	0.93393	0.033405	23.9	159.43	0.81904	0.04	0.26684
BY11-F50	0.07	0.23554		-0.01	-0.033648					-0.03	-0.10094
BY12-UF	0.06	0.58051		0.11	1.0643		18.7	180.93		0.03	0.29026
BY12	0.07	0.45325	0.022034	0.12	0.777	0.027695	23.5	152.16	0.76128	0.04	0.259
BY12-F50	0.04	0.13223		-0.01	-0.033057					-0.03	-0.099171
BY13	0.21	0.5468	0.028509	0.49	1.2759	0.04652	66	171.85	0.93081	0.07	0.18227
BY14	0.2	0.51604	0.026055	0.35	0.90307	0.032317	65.1	167.97	0.88415	0.08	0.20642
BY15-UF	0.09	1.0008		0.16	1.7792		20.4	226.85		-0.03	-0.3336
BY15	0.19	0.79021	0.042775	0.35	1.4556	0.051549	48.8	202.96	1.1374	0.07	0.29113
BY15-F50	0.01	0.1112		-0.01	-0.1112		16.8	186.82		-0.03	-0.3336
BY16-UF	0.09	0.97542		0.17	1.8425		22	238.44		0.03	0.32514
BY16	0.19	0.82166	0.045126	0.32	1.3838	0.049448	46.3	200.22	1.1269	0.05	0.21623
BY16-F50	0.02	0.21676		-0.01	-0.10838		17.3	187.5		-0.03	-0.32514
BY17	0.013	0.056	-0.0029054	0.01	0.043	0.0016225	47.7	206.9	1.2434	0.004	0.017
BY17-F50	0.067	0.329		0.005	0.025		42.5	208.5		0.007	0.034
BY18	0.014	0.06	-0.0025126	0.01	0.043	0.0015434	49.8	213.5	1.2329	0.028	0.12
BY18-F50	0.051	0.249		0.001	0.007		41.4	201.9		-0.001	-0.005
BKGND	0.1	0.1		0	0		51.32	51.32		0	0
F(i)	0.008	0.008		0.014	0.014		0.0661	0.0661		0.0065	0.0065

TEST NO.	NL(Ni), g/m	Si, ACL	Si, ppm	NL(Si), g/m	Th, ACL	Th, ppm	NL(Th), g/m	Ti, ACL	Ti, ppm	NL(Ti), g/m	U, ACL
BY1	-0.0054297	26.1	60.98	0.034677	-0.1	-0.23364	-0.051149	-0.01	-0.023364	-0.0021786	0.636
BY2	-0.0052801	24.8	58.134	0.027536	-0.1	-0.23441	-0.04974	-0.01	-0.023441	-0.0021186	0.564
BY3	-0.0055207	34.6	80.58	0.079388	-0.2	-0.46578	-0.10401	0.027	0.06288	0.0059807	1.428
BY4	-0.0053591	36	85.524	0.086114	-0.2	-0.47514	-0.10097	0.035	0.083149	0.0075259	1.5
BY5	0.0068252	43.3	101.3	0.12235	-0.2	-0.46792	-0.10152	0.055	0.12868	0.011891	1.492
BY6	0.013088	40.6	95.686	0.11216	-0.2	-0.47136	-0.10419	0.046	0.10841	0.010207	1.532
BY7	0.0079804	52.3	123.05	0.16671	-0.2	-0.47054	-0.10024	0.079	0.18586	0.016864	1.08
BY8	0.0096703	52.4	123.23	0.172	-0.2	-0.47036	-0.10313	0.091	0.21401	0.019986	1.066
BY9	0.0052812	64.8	147.78	0.22498	0.07	0.15963	0.034825	0.03	0.068415	0.006357	1.047
BY10	0.011074	62.4	148.29	0.22749	0.1	0.23765	0.052159	0.1	0.23765	0.022216	0.92
BY11-UF		18.6	179.57		-0.05	-0.48271		0.04	0.38616		0.392
BY11	0.020557	22.8	152.1	0.23399	-0.05	-0.33354	-0.072619	0.05	0.33354	0.03093	0.462
BY11-F50		42.7	143.68		-0.05	-0.16824		-0.02	-0.067296		0.747
BY12-UF		18	174.15		-0.05	-0.48376		0.04	0.38701		0.395
BY12	0.019883	23	148.93	0.22625	-0.05	-0.32375	-0.07024	0.05	0.32375	0.029917	0.481
BY12-F50		45	148.76		-0.05	-0.16529		-0.02	-0.066114		0.824
BY13	0.014314	69.4	180.7	0.30238	0.11	0.28642	0.063568	0.13	0.33849	0.031998	1.4795
BY14	0.01591	69.5	179.32	0.29375	0.11	0.28382	0.061824	0.13	0.33543	0.03112	1.413
BY15-UF		22.2	246.86		-0.05	-0.556		0.05	0.556		0.597
BY15	0.022206	52.6	218.76	0.37619	0.07	0.29113	0.062756	0.12	0.49908	0.045822	1.374
BY15-F50		18.2	202.38		-0.05	-0.556		-0.02	-0.2224		0.393
BY16-UF		25.6	277.45		-0.05	-0.5419		0.05	0.5419		0.64
BY16	0.016641	51.8	224.01	0.39105	0.07	0.30272	0.065841	0.1	0.43245	0.040062	1.237
BY16-F50		19.2	208.09		-0.05	-0.5419		-0.02	-0.21676		0.379
BY17	0.0013816	52.9	229.4	0.4254	0.044	0.191	0.043869	0.02	0.087	0.0085109	1.11
BY17-F50		44.8	219.8		0.011	0.054		0.006	0.029		0.906
BY18	0.0092772	53.1	227.7	0.40093	0.043	0.187	0.040856	0.02	0.086	0.008003	1.223
BY18-F50		43.6	212.6		0.009	0.44		0.005	0.024		1.024
BKGND		45.23	45.23		0	0		0	0		0
F(i)		0.2287	0.2287		0.0023	0.0023		0.0054	0.0054		0.0164

TEST NO.	U, ppm	NL(U), g/m2	Zn, ACL	Zn, ppm	NL(Zn), g/m	Zr, ACL	Zr, ppm	NL(Zr), g/m
BY1	1.486	0.045623	-0.01	-0.023364	-0.058822	-0.02	-0.046728	-0.033612
BY2	1.3221	0.039343	-0.01	-0.023441	-0.057201	-0.02	-0.046882	-0.032686
BY3	3.3257	0.10415				-0.02	-0.046578	-0.034176
BY4	3.5635	0.1062				-0.02	-0.047514	-0.033175
BY5	3.4907	0.10621				-0.02	-0.046792	-0.033356
BY6	3.6106	0.11193				-0.02	-0.047136	-0.034233
BY7	2.5409	0.075911				-0.02	-0.047054	-0.032935
BY8	2.507	0.077089				0.02	0.047036	0.033885
BY9	2.3877	0.073051	-0.02	-0.04561	-0.11443	-0.05	-0.11402	-0.081732
BY10	2.1864	0.067297	0.02	0.04753	0.11996	-0.05	-0.11882	-0.085689
BY11-UF	3.7844		-0.02	-0.19308		-0.05	-0.48271	
BY11	3.082	0.094103	-0.02	-0.13342	-0.33405	-0.05	-0.33354	-0.2386
BY11-F50	2.5135		0.05	0.16824		-0.05	-0.16824	
BY12-UF	3.8217		-0.02	-0.1935		-0.05	-0.48376	
BY12	3.1145	0.094764	-0.02	-0.1295	-0.3231	-0.05	-0.32375	-0.23079
BY12-F50	2.7239		-0.02	-0.066114		-0.05	-0.16529	
BY13	3.8523	0.11991	0.02	0.052076	0.13291	-0.05	-0.13019	-0.094939
BY14	3.6458	0.11138	-0.02	-0.051604	-0.12927	-0.05	-0.12901	-0.092335
BY15-UF	6.6386		-0.02	-0.2224		-0.05	-0.556	
BY15	5.7145	0.17275	0.04	0.16636	0.41239	-0.05	-0.20795	-0.14728
BY15-F50	4.3702		0.03	0.3336		-0.05	-0.556	
BY16-UF	6.9363		-0.02	-0.21676		-0.05	-0.5419	
BY16	5.3494	0.16317	0.02	0.08649	0.21633	-0.05	-0.21623	-0.15452
BY16-F50	4.1076		0.09	0.97542		-0.05	-0.5419	
BY17	4.814	0.15506	0.019	0.082	0.21659	0.082	0.006	0.004528
BY17-F50	4.446		0.07	0.343		0.343	0.001	
BY18	5.244	0.16068	0.039	0.167	0.4196	0.167	0.006	0.0043073
BY18-F50	4.994		0.03	0.146		0.146	0.001	
BKGND	0		0	0		0	0	
f(i)	0.0164		0.0002	0.0002		0.0007	0.0007	

TEST NUMBER	RXN TIME	pH	DIL FACTOR	SA/V	LEACHANT VO	ACL NUMBER	Al, ACL	Al, ppm	NL(Al), g/m	B, ACL	B, ppm
BZ1	3	10.5	7.5238	20089	4.99	91-0061-03	0.72	5.4171	0.01191	8.18	61.545
BZ2	3	10.62	7.3333	19607	5.03	91-0061-04	0.75	5.5	0.012411	8.65	63.433
BZ3	7	10.73	5.7009	20072	5	90-0513-08	1.51	8.6084	0.019752	14.9	84.943
BZ4	7	10.48	5.552	19820	5.01	90-0513-09	1.5	8.328	0.019307	15	83.28
BZ5	14	10.98	7.7444	19902	4.97	90-0487-20	1.5	11.617	0.027367	12.6	97.579
BZ6	14	11.01	7.6245	19997	4.98	90-0487-21	1.48	11.284	0.026418	13.1	99.881
BZ7	28	11.02	9.3039	20297	4.95	90-0487-12	0.25	2.326	0.004286	13.5	125.6
BZ8	28	11.1	11	20047	5.02	90-0487-13	0.59	6.49	0.014572	11	121
BZ9	56	11.24	11.386	19957	4.99	91-0134-10	0.16	1.8218	0.0031145	13.6	154.85
BZ10	56	11.33	11.451	20034	5.01	91-0134-09	0.2	2.2903	0.0042545	13.5	154.59
BZ11	98	11.26	10.331	20132	4.97	91-0205-05	0.14	1.4464	0.0021689	18.4	190.1
BZ12	98	11.43	10.114	20015	4.98	91-0205-06	0.14	1.416	0.0021067	17.5	177
BZ13	182	10.92	19.862	19963	4.98	91-0346-07	-0.1	-1.9862	-0.0062831	13.1	260.19
BZ13-F50			4.8358			91-0346-09	0.22	1.0639		57.9	279.99
BZ14	182	11.29	16.792	20168	5	91-0346-08	-0.1	-1.6792	-0.0054693	17.1	287.14
BZ14-F50			6.0792			91-0346-10	0.2	1.2158		50.5	307
BZ15	364	12.03	14.636	20008	5	91-0481-01	0.11	1.61	0.0025851	220	3219.9
BZ15-F50			11.402			91-0481-03	-0.1	-1.1402		273	3112.7
BZ16	364	12.02	15.89	20088	5	91-0481-02	-0.1	-1.589	-0.0052699	164	2606
BZ16-F50			11.479			91-0481-04	0.12	1.3775		217	2490.9
BZ17	728	11.96	21.658	20000	5.02	92-0388-05	-0.1	-2.1658	-0.0067138	392	8489.9
BZ17-F50			12.641			92-0388-06	-0.1	-1.2641		664	8393.6
BZ18	728	11.97	22.153	20000	5.02	92-0388-07	-0.1	-2.2153	-0.0068357	395	8750.4
BZ18-F50			10.263			92-0388-08	-0.1	-1.0263		703	7214.9
BZ19	504	11.86	20.494	20000	5.03	92-0388-09	-0.1	-2.0494	-0.0064271	285	5840.8
BZ19-F50			11.821			92-0388-10	-0.1	-1.1821		491	5804.1
BKGND			1				0.56	0.56		0.15	0.15
f(1)			1				0.0203	0.0203		0.0248	0.0248

TEST NO.	NL(B), g/m2	Ba, ACL	Ba, ppm	NL(Ba), g/m	Ca, ACL	Ca, ppm	NL(Ca), g/m	Cr, ACL	Cr, ppm	NL(Cr), g/m	Cu, ACL
BZ1	0.12323	-0.02	-0.15048	-0.0037452	0.04	0.30095	-0.02471	0.02	0.15048	0.014981	0.55
BZ2	0.13014	-0.02	-0.14667	-0.0037401	0.05	0.36667	-0.024928	0.02	0.14667	0.014961	0.6
BZ3	0.17034	0.02	0.11402	0.0028402	0.11	0.6271	-0.022842	0.054	0.30785	0.030674	1
BZ4	0.16912	0.02	0.11104	0.0028012	0.1	0.5552	-0.023554	0.05	0.2776	0.028012	1.01
BZ5	0.1974	-0.02	-0.15489	-0.0038913	0.11	0.85188	-0.021723	0.054	0.4182	0.042026	1.01
BZ6	0.2011	0.021	0.16011	0.0040035	0.096	0.73195	-0.022318	0.046	0.35073	0.035078	1.01
BZ7	0.24923	-0.01	-0.093039	-0.0022919	0.015	0.13956	-0.025381	0.036	0.33494	0.033004	0.058
BZ8	0.24308	-0.01	-0.11	-0.0027436	0.048	0.528	-0.023445	0.04	0.44	0.043897	0.34
BZ9	0.31257	-0.02	-0.22772	-0.0057053	0.01	0.11386	-0.025964	0.06	0.68316	0.068463	0.02
BZ10	0.31085	-0.02	-0.22903	-0.005716	0.02	0.22903	-0.025195	0.06	0.68708	0.068592	0.06
BZ11	0.38044	-0.02	-0.20663	-0.0051318	-0.01	-0.10331	-0.026992	0.4	4.1325	0.41054	-0.02
BZ12	0.35628	-0.02	-0.20228	-0.0050532	-0.01	-0.10114	-0.027137	0.03	0.30342	0.030319	-0.02
BZ13	0.52525	-0.02	-0.39724	-0.0099494	0.03	0.59586	-0.023148	0.29	5.76	0.57707	-0.02
BZ13-F50		-0.02	-0.096716		0.43	2.0794		1.22	5.8997		-0.02
BZ14	0.5738	-0.02	-0.33584	-0.0083261	0.06	1.0075	-0.02054	0.09	1.5113	0.14987	-0.02
BZ14-F50		-0.02	-0.12158		0.37	2.2493		0.27	1.6414		-0.02
BZ15	6.4889	0.03	0.43908	0.010973	0.04	0.58544	-0.023157	-0.02	-0.29272	-0.02926	-0.02
BZ15-F50		0.05	0.5701		0.29	3.3066		-0.02	-0.22804		0.08
BZ16	5.2306	0.02	0.3178	0.0079102	0.04	0.6356	-0.022774	-0.02	-0.3178	-0.031641	-0.02
BZ16-F50		0.03	0.34437		0.17	1.9514		-0.02	-0.22958		-0.02
BZ17	17.117	0.05	1.0829	0.027073	0.11	2.3824	-0.012719	-0.02	-0.43316	-0.043316	0.18
BZ17-F50		0.07	0.88487		0.26	3.2867		-0.02	-0.25282		0.27
BZ18	17.642	0.05	1.1077	0.027691	0.12	2.6584	-0.011114	-0.02	-0.44306	-0.044306	0.14
BZ18-F50		0.08	0.82104		0.26	2.6684		-0.02	-0.20526		0.22
BZ19	11.775	0.03	0.61482	0.01537	0.07	1.4346	-0.018229	-0.02	-0.40988	-0.040988	-0.02
BZ19-F50		0.05	0.59105		0.24	2.837		-0.02	-0.23642		-0.02
BKGND		0	0		4.57	4.57		0	0		0
f(i)		0.002	0.002		0.0086	0.0086		0.0005	0.0005		0.0032

TEST NO.	Cu, ppm	NL(Cu), g/m	Fe, ACL	Fe, ppm	NL(Fe), g/m	K, ACL	K, ppm	NL(K), g/m2	Li, ACL	Li, ppm	NL(Li), g/m
B21	4.1381	0.064371	1.67	12.565	0.0078378	6.13	46.121	0.062958	8.67	65.231	0.1647
B22	4.4	0.070128	1.73	12.687	0.0081083	6.42	47.08	0.066088	8.91	65.34	0.16903
B23	5.7009	0.088757	4.23	24.115	0.015055	9.93	56.61	0.079923	14.2	80.953	0.2046
B24	5.6075	0.088413	3.86	21.431	0.01355	10.1	56.075	0.080066	14.5	80.504	0.20605
B25	7.8218	0.12282	4.35	33.688	0.021212	8.9	68.925	0.10063	12.4	96.031	0.24481
B26	7.7007	0.12034	4.24	32.328	0.020259	10.1	77.007	0.11323	12.6	96.069	0.24374
B27	0.53963	0.0083083	0.16	1.4886	0.00091907	9.29	86.433	0.12659	11.6	107.93	0.26979
B28	3.74	0.0583	1.58	17.38	0.010864	7.72	84.92	0.12572	9.81	107.91	0.27311
B29	0.22772	0.0035658	0.04	0.45544	0.00028598	9.12	103.84	0.15697	10.2	116.14	0.29527
B210	0.68708	0.010717	0.18	2.0613	0.0012893	9.33	106.84	0.16122	10.1	115.66	0.29293
B211	-0.20663	-0.0032074	-0.02	-0.20663	-0.00012862	12.6	130.17	0.19794	13.9	143.61	0.36196
B212	-0.20228	-0.0031583	-0.02	-0.20228	-0.00012665	12.2	123.39	0.18813	13.7	138.56	0.35129
B213	-0.39724	-0.0062184	0.04	0.79448	0.00049872	9.29	184.52	0.28771	7.22	143.4	0.36452
B213-F50	-0.096716		0.11	0.53194							
B214	-0.33584	-0.0052038	0.03	0.50376	0.00031301	12.3	206.54	0.32013	8.79	147.6	0.37138
B214-F50	-0.12158		0.07	0.42554							
B215	-0.29272	-0.0045719	-0.02	-0.29272	-0.00018334	107	1566.1	2.5217	41.6	608.86	1.5446
B215-F50	0.91216		0.03	0.34206		130	1482.3		51.4	586.06	
B216	-0.3178	-0.0049439	-0.02	-0.3178	-0.00019825	83.8	1331.6	2.1339	35.1	557.74	1.4093
B216-F50	-0.22958		-0.02	-0.22958		110	1262.7		48.5	556.73	
B217	3.8984	0.060913	0.09	1.9492	0.0012213	149	3227	5.2104	43.8	948.62	2.4075
B217-F50	3.4131		-0.02	-0.25282		241	3046.5		75.3	951.87	
B218	3.1014	0.04846	0.03	0.66459	0.00041641	144	3190	5.1505	45.1	999.1	2.5357
B218-F50	2.2579		-0.02	-0.20526		249	2555.5		81.8	839.51	
B219	-0.40988	-0.0064044	-0.02	-0.40988	-0.00025682	118	2418.3	3.9017	31.4	643.51	1.6332
B219-F50	-0.23642		-0.02	-0.23642		201	2376		53.7	634.79	
BKGND	0		0	0		7.04	7.04		0.05	0.05	
f(i)	0.0032		0.0798	0.0798		0.0309	0.0309		0.0197	0.0197	

TEST NO.	Mg, ACL	Mg, ppm	NL(Mg), g/m	Mn, ACL	Mn, ppm	NL(Mn), g/m	Na, ACL	Na, ppm	NL(Na), g/m	Ni, ACL	Ni, ppm
BZ1	0.262	1.9712	0.011643	0.457	3.4384	0.012226	31.2	234.74	0.13813	0.14	1.0533
BZ2	0.282	2.068	0.012546	0.474	3.476	0.012663	32	234.67	0.14147	0.14	1.0267
BZ3	0.66	3.7626	0.022809	1.21	6.8981	0.024548	49.2	280.48	0.17272	0.31	1.7673
BZ4	0.61	3.3867	0.020729	1.13	6.2738	0.02261	50.5	280.38	0.17484	0.3	1.6656
BZ5	0.63	4.879	0.030016	1.23	9.5256	0.034188	43.4	336.11	0.21648	0.29	2.2459
BZ6	0.63	4.8034	0.029401	1.24	9.4544	0.033771	44.1	336.24	0.21555	0.29	2.2111
BZ7	0.021	0.19538	0.00058741	0.038	0.35355	0.0012442	44.2	411.23	0.26826	-0.03	-0.27912
BZ8	0.21	2.31	0.01378	0.41	4.51	0.016069	36.9	405.9	0.26759	0.12	1.32
BZ9	-0.01	-0.11386	-0.0013395	0.01	0.11386	0.00040752	44.8	510.09	0.34778	-0.03	-0.34158
BZ10	0.02	0.22903	0.00080506	0.04	0.45806	0.0016331	44.1	505.01	0.3426	-0.03	-0.34354
BZ11	-0.01	-0.10331	-0.0012624	-0.01	-0.10331	-0.00036656	57.4	593.02	0.40707	-0.03	-0.30994
BZ12	-0.01	-0.10114	-0.0012562	-0.01	-0.10114	-0.00036094	55.5	561.33	0.3855	-0.03	-0.30342
BZ13	0.02	0.39724	0.0018612	-0.01	-0.19862	-0.00071067	37.6	746.81	0.52707	-0.03	-0.59586
BZ13-F50	0.16	0.77373		-0.01	-0.048358		160	773.73		-0.03	-0.14507
BZ14	0.04	0.67168	0.0035432	-0.01	-0.16792	-0.00059472	49.6	832.88	0.58627	-0.03	-0.50376
BZ14-F50	0.14	0.85109		-0.01	-0.060792		138	838.93		-0.03	-0.18238
BZ15	-0.01	-0.14636	-0.0015391	-0.01	-0.14636	-0.00052251	464	6791.1	5.0961	-0.03	-0.43908
BZ15-F50	0.04	0.45608		-0.01	-0.11402		605	6898.2		-0.03	-0.34206
BZ16	-0.01	-0.1589	-0.001611	-0.01	-0.1589	-0.00056501	352	5593.3	4.1737	-0.03	-0.4767
BZ16-F50	0.02	0.22958		-0.01	-0.11479		469	5383.7		-0.03	-0.34437
BZ17	-0.01	-0.21658	-0.0019786	-0.01	-0.21658	-0.0007735	742	16070	12.117	-0.03	-0.64974
BZ17-F50	0.01	0.12641		-0.01	-0.12641		1214	15346		-0.03	-0.37923
BZ18	-0.01	-0.22153	-0.0020096	-0.01	-0.22153	-0.00079118	758	16792	12.663	-0.03	-0.66459
BZ18-F50	0.01	0.10263		-0.01	-0.10263		1299	13332		-0.03	-0.30789
BZ19	-0.01	-0.20494	-0.0019059	-0.01	-0.20494	-0.00073193	557	11415	8.5959	-0.03	-0.61482
BZ19-F50	0.02	0.23642		-0.01	-0.11821		923	10911		-0.03	-0.35463
BKGND	0.1	0.1		0	0		51.32	51.32		0	0
f(i)	0.008	0.008		0.014	0.014		0.0661	0.0661		0.0065	0.0065

TEST NO.	NL(Ni), g/m	Si, ACL	Si, ppm	NL(Si), g/m	Th, ACL	Th, ppm	NL(Th), g/m	Ti, ACL	Ti, ppm	NL(Ti), g/m	U, ACL
BZ1	0.0080667	28.2	212.17	0.036336	-0.1	-0.75238	-0.016284	0.1	0.75238	0.0069356	1.173
BZ2	0.0080557	29.8	218.53	0.038648	-0.1	-0.73333	-0.016261	0.1	0.73333	0.0069262	1.236
BZ3	0.013546	49.7	283.33	0.051869	-0.2	-1.1402	-0.024698	0.25	1.4252	0.013149	2.318
BZ4	0.012929	49.7	275.93	0.050896	-0.2	-1.1104	-0.024358	0.22	1.2214	0.011412	2.3
BZ5	0.017361	42	325.26	0.061525	-0.2	-1.5489	-0.033837	0.25	1.9361	0.018015	2.027
BZ6	0.017011	43.4	330.9	0.062465	-0.2	-1.5249	-0.033155	0.25	1.9061	0.017652	1.995
BZ7	-0.0021156	38.6	359.13	0.067623	-0.2	-1.8608	-0.03986	0.011	0.10234	0.00093375	0.978
BZ8	0.01013	32.8	360.8	0.06883	-0.2	-2.2	-0.047714	0.094	1.034	0.0095516	1.088
BZ9	-0.0026332	34.1	388.26	0.075158	-0.1	-1.1386	-0.024806	-0.01	-0.11386	-0.0010565	0.493
BZ10	-0.0026381	34.4	393.93	0.076105	0.1	1.1451	0.024852	0.01	0.11451	0.0010585	0.545
BZ11	-0.0023685	50.9	525.86	0.10439	-0.05	-0.51657	-0.011156	-0.02	-0.20663	-0.0019007	0.481
BZ12	-0.0023323	52.4	529.97	0.1059	-0.05	-0.5057	-0.010985	-0.02	-0.20228	-0.0018716	0.409
BZ13	-0.004592	24.5	486.62	0.096678	-0.05	-0.9931	-0.021629	-0.02	-0.39724	-0.003685	0.144
BZ13-F50		105	507.76		-0.05	-0.24179		-0.02	-0.096716		0.582
BZ14	-0.0038428	31.7	532.31	0.1056	-0.05	-0.8396	-0.0181	-0.02	-0.33584	-0.0030837	0.199
BZ14-F50		92.1	559.89		-0.05	-0.30396		-0.02	-0.12158		0.514
BZ15	-0.0033762	95.7	1400.7	0.29622	-0.05	-0.7318	-0.015902	-0.02	-0.29272	-0.0027093	0.0741
BZ15-F50		117	1334		-0.05	-0.5701		-0.02	-0.22804		0.0821
BZ16	-0.0036509	85.1	1352.2	0.2845	-0.05	-0.7945	-0.017196	-0.02	-0.3178	-0.0029297	0.0788
BZ16-F50		112	1285.6		-0.05	-0.57395		-0.02	-0.22958		0.0897
BZ17	-0.004998	77.8	1685	0.3585	-0.05	-1.0829	-0.023541	-0.02	-0.43316	-0.0040107	0.0311
BZ17-F50		130	1643.3		-0.05	-0.63205		-0.02	-0.25282		0.0473
BZ18	-0.0051122	80.5	1783.3	0.37999	-0.05	-1.1077	-0.024079	-0.02	-0.44306	-0.0041024	0.0336
BZ18-F50		141	1447.1		-0.05	-0.51315		-0.02	-0.20526		0.0567
BZ19	-0.0047294	72.6	1487.9	0.3154	-0.05	-1.0247	-0.022276	-0.02	-0.40988	-0.0037952	0.0209
BZ19-F50		126	1489.4		-0.05	-0.59105		-0.02	-0.23642		0.0324
BKGND		45.23	45.23		0	0		0	0		0
f(i)		0.2287	0.2287		0.0023	0.0023		0.0054	0.0054		0.0164

TEST NO.	U, ppm	NL(U), g/m2	Zn, ACL	Zn, ppm	NL(Zn), g/m	Zr, ACL	Zr, ppm	NL(Zr), g/m
BZ1	8.8254	0.026788				-0.02	-0.15048	-0.010701
BZ2	9.064	0.028188				-0.02	-0.14667	-0.010686
BZ3	13.215	0.040144				0.025	0.14252	0.010144
BZ4	12.77	0.039285				0.023	0.1277	0.009204
BZ5	15.698	0.048095				0.021	0.16263	0.011674
BZ6	15.211	0.046382				0.023	0.17536	0.012528
BZ7	9.0992	0.027336				-0.02	-0.18608	-0.013097
BZ8	11.968	0.036402				-0.02	-0.22	-0.015677
BZ9	5.6133	0.017151	-0.01	-0.11386	-0.028526	-0.02	-0.22772	-0.016301
BZ10	6.241	0.018995	0.01	0.11451	0.02858	-0.02	-0.22903	-0.016331
BZ11	4.9694	0.015051	-0.02	-0.20663	-0.051318	-0.05	-0.51657	-0.036656
BZ12	4.1366	0.012602	-0.02	-0.20228	-0.050532	-0.05	-0.5057	-0.036094
BZ13	2.8601	0.0087361	-0.02	-0.39724	-0.099494	-0.05	-0.9931	-0.071067
BZ13-F50	2.8144		0.05	0.24179		-0.05	-0.24179	
BZ14	3.3416	0.010103	-0.02	-0.33584	-0.083261	-0.05	-0.8396	-0.059472
BZ14-F50	3.1247		0.02	0.12158		-0.05	-0.30396	
BZ15	1.0845	0.0033052	-0.02	-0.29272	-0.073151	-0.05	-0.7318	-0.052251
BZ15-F50	0.9361		0.07	0.79814		-0.05	-0.5701	
BZ16	1.2521	0.0038008	-0.02	-0.3178	-0.079102	-0.05	-0.7945	-0.056501
BZ16-F50	1.0297		0.02	0.22958		-0.05	-0.57395	
BZ17	0.67356	0.0020535	-0.02	-0.43316	-0.10829	-0.05	-1.0829	-0.07735
BZ17-F50	0.59792		-0.02	-0.25282		-0.05	-0.63205	
BZ18	0.74434	0.0022693	-0.02	-0.44306	-0.11076	-0.05	-1.1077	-0.079118
BZ18-F50	0.58191		-0.02	-0.20526		-0.05	-0.51315	
BZ19	0.42832	0.0013059	-0.02	-0.40988	-0.10247	-0.05	-1.0247	-0.073193
BZ19-F50	0.383		-0.02	-0.23642		-0.05	-0.59105	
BKGND	0		0	0		0	0	
F(i)	0.0164		0.0002	0.0002		0.0007	0.0007	

TEST NUMBER	RXN TIME	pH	DIL FACTOR	SA/V	LEACHANT VO	ACL NUMBER	Al, ACL	Al, ppm	NL(Al), g/m	B, ACL	B, ppm
PX1	7	8.18	1.6729	9.99	17.19	90-0651-17	0.42	0.70262	0.82521	0.27	0.45168
PX2	7	8.24	1.6771	9.99	17.1	90-0651-18	0.5	0.83855	1.6117	0.16	0.26834
PX3	28	8.3	1.699	10	16.78	91-0061-09	4.6	7.8154	41.939	0.41	0.69659
PX4	28	8.24	1.6945	10	16.78	91-0061-10	0.44	0.74558	1.0727	0.29	0.4914
PX5	94	8.44	1.6473	10	16.78	91-0134-12	0.33	0.54361	-0.094746	1.04	1.7132
PX6	94	8.31	1.6962	10	16.79	91-0134-07	0.34	0.57671	0.096578	0.49	0.83114
PX7	238	8.58	2.3302	12.2	13.54	91-0381-01	0.2	0.46604	-0.44518	0.82	1.9108
PX7-F50			9.4358			91-0381-02	-0.1	-0.94358		0.22	2.0759
PX8	238	9.27	1.583	10	17.17	91-0381-03	0.38	0.60154	0.24012	6.6	10.448
PX8-F50			8.8953			91-0381-04	-0.1	-0.88953		1.22	10.852
PX9	600	9.09	1.4965	10	18.51	92-0353-05	0.63	0.94279	2.2127	6.98	10.446
PX9-F50			4.75			92-0353-06	0.18	0.855		2.92	13.87
PX10	600	9.34	1.472	10	17.11	92-0353-07	0.83	1.2218	3.8252	10.8	15.898
PX10-F50			3.642			92-0353-08	0.19	0.69198		3.38	12.31

BKGND			1				0.56	0.56		0.15	0.15
F(i)			1				0.0173	0.0173		0.03	0.03

TEST NO.	NL(B), g/m2	Ba, ACL	Ba, ppm	NL(Ba), g/m	Ca, ACL	Ca, ppm	NL(Ca), g/m	Cr, ACL	Cr, ppm	NL(Cr), g/m	Cu, ACL
PX1	1.0066	-0.02	-0.033458	-2.3922	2.97	4.9685	6.0441	-0.02	-0.033458	-3.7213	0.2
PX2	0.39485	-0.02	-0.033542	-2.3983	3.23	5.417	12.847	-0.02	-0.033542	-3.7306	0.06
PX3	1.822	-0.02	-0.03398	-2.4271	2.9	4.9271	5.4106	-0.02	-0.03398	-3.7756	0.19
PX4	1.138	-0.02	-0.03389	-2.4207	2.93	4.9649	5.9831	0.05	0.084725	9.4139	0.31
PX5	5.2106	0.02	0.032946	2.3533	2.34	3.8547	-10.838	6.78	11.169	1241	0.47
PX6	2.2705	-0.02	-0.033924	-2.4231	2.71	4.5967	0.40457	-0.02	-0.033924	-3.7693	0.62
PX7	4.8108	-0.02	-0.046604	-2.7286	1.95	4.5439	-0.32427	-0.02	-0.046604	-4.2444	0.11
PX7-F50		-0.02	-0.18872		0.46	4.3405		-0.02	-0.18872		-0.02
PX8	34.326	-0.02	-0.03166	-2.2614	0.13	0.20579	-66.124	-0.02	-0.03166	-3.5178	0.2
PX8-F50		-0.02	-0.17791		0.2	1.7791		-0.02	-0.17791		0.02
PX9	34.319	-0.02	-0.02993	-2.1379	0.7	1.0475	-53.37	-0.02	-0.02993	-3.3256	0.34
PX9-F50		-0.02	-0.095		0.32	1.52		-0.02	-0.095		-0.02
PX10	52.492	-0.02	-0.02944	-2.1029	0.34	0.50048	-61.659	0.06	0.08832	9.8133	0.4
PX10-F50		-0.02	-0.07284		0.28	1.0198		0.03	0.10926		-0.02
BKGND		0	0		4.57	4.57		0	0		0
F(i)		0.0014	0.0014		0.0066	0.0066		0.0009	0.0009		0.0002

TEST NO.	Cu, ppm	NL(Cu), g/m	Fe, ACL	Fe, ppm	NL(Fe), g/m	K, ACL	K, ppm	NL(K), g/m2	Li, ACL	Li, ppm	NL(Li), g/m
PX1	0.33458	167.46	0.03	0.050187	0.056958	4.68	7.8292	2.4533	0.15	0.25094	1.4367
PX2	0.10063	50.363	0.03	0.050313	0.057101	4.55	7.6308	1.8366	0.1	0.16771	0.84163
PX3	0.32281	161.4	0.09	0.15291	0.17337	4.88	8.2911	3.8855	0.23	0.39077	2.4341
PX4	0.5253	262.65	0.16	0.27112	0.30739	4.65	7.8794	2.6069	0.19	0.32195	1.9425
PX5	0.77423	387.12	13.4	22.074	25.027	5.64	9.2908	6.99	0.54	0.88954	5.9967
PX6	1.0516	525.82	0.04	0.067848	0.076925	4.94	8.3792	4.1591	0.3	0.50886	3.2776
PX7	0.25632	105.05	0.04	0.093208	0.086621	4.58	10.672	9.2463	0.43	1.002	5.5737
PX7-F50	-0.18872		-0.02	-0.18872		1.13	10.662		0.09	0.84922	
PX8	0.3166	158.3	0.2	0.3166	0.35896	10.5	16.622	29.756	3.17	5.0181	35.486
PX8-F50	0.17791		-0.02	-0.17791		1.19	10.585		0.55	4.8924	
PX9	0.50881	254.4	0.97	1.4516	1.6458	11.5	17.21	31.583	2.95	4.4147	31.176
PX9-F50	-0.095		-0.02	-0.095		4.73	22.468		1.19	5.6525	
PX10	0.5888	294.4	0	0	0	13.3	19.578	38.937	4.43	6.521	46.221
PX10-F50	-0.07284		0.63	2.2945		4.21	15.333		1.35	4.9167	
BKGND	0		0	0	0	7.04	7.04		0.05	0.05	
F(i)	0.0002		0.0882	0.0882		0.0322	0.0322		0.014	0.014	

TEST NO.	Mg, ACL	Mg, ppm	NL(Mg), g/m	Mn, ACL	Mn, ppm	NL(Mn), g/m	Na, ACL	Na, ppm	NL(Na), g/m	Ni, ACL	Ni, ppm
PX1	0.26	0.43495	4.2442	0.02	0.033458	0.21748	35.1	58.719	8.3757	-0.03	-0.050187
PX2	0.26	0.43605	4.258	0.1	0.16771	1.0901	33.7	56.518	5.9146	-0.03	-0.050313
PX3	0.282	0.47912	4.799	0.046	0.078154	0.50749	34.3	58.276	7.8723	0.04	0.06796
PX4	0.276	0.46768	4.6542	0.078	0.13217	0.85825	33.4	56.596	5.9959	0.05	0.084725
PX5	0.24	0.39535	3.7386	0.69	1.1366	7.3808	37.4	61.609	11.597	5.41	8.9119
PX6	0.25	0.42405	4.1019	0.06	0.10177	0.66086	35.6	60.385	10.229	0.06	0.10177
PX7	0.09	0.20972	1.1384	0.03	0.069906	0.37208	31.1	72.469	19.452	0.04	0.093208
PX7-F50	0.03	0.28307		-0.01	-0.094358		7.74	73.033		-0.03	-0.28307
PX8	0.22	0.34826	3.1425	0.13	0.20579	1.3363	53.1	84.057	36.679	0.08	0.12664
PX8-F50	0.02	0.17791		-0.01	-0.088953		9.78	86.996		-0.03	-0.26686
PX9	0.18	0.26937	2.1439	0.19	0.28433	1.8463	59.2	88.593	41.746	0.06	0.08979
PX9-F50	0.03	0.1425		-0.01	-0.0475		25.2	119.7		0.04	0.19
PX10	0.66	0.97152	11.032	0.28	0.41216	2.6764	69.1	101.72	56.408	0.29	0.42688
PX10-F50	0.04	0.14568		-0.01	-0.03642		21.6	78.667		-0.03	-0.10926

BKGND	0.1	0.1		0	0		51.23	51.23		0	0
f(i)	0.0079	0.0079		0.0154	0.0154		0.0895	0.0895		0.0098	0.0098

TEST NO.	NL(Ni), g/m	Si, ACL	Si, ppm	NL(Si), g/m	Th, ACL	Th, ppm	NL(Th), g/m	Ti, ACL	Ti, ppm	NL(Ti), g/m	U, ACL
PX1	-0.51262	28.3	47.343	1.0343	-0.06	-0.10037	0				0.0219
PX2	-0.51391	28.4	47.63	1.1746	-0.06	-0.10063	0				0.0113
PX3	0.69347	27.5	46.722	0.72983	-0.1	-0.1699	0	-0.01	-0.01699	0	0.0254
PX4	0.86454	26.8	45.413	0.08929	-0.1	-0.16945	0	-0.01	-0.016945	0	0.0161
PX5	90.938	28.4	46.783	0.75957	-0.1	-0.16473	0	0.01	0.016473	0	0.0634
PX6	1.0385	26.1	44.271	-0.46904	-0.1	-0.16962	0	-0.01	-0.016962	0	0.0556
PX7	0.77959	23.3	54.294	3.6329	-0.05	-0.11651	0	-0.02	-0.046604	0	0.0811
PX7-F50		5.75	54.256		-0.05	-0.47179		-0.02	-0.18872		0.0187
PX8	1.2922	43.4	68.702	11.478	-0.05	-0.07915	0	-0.02	-0.03166	0	0.232
PX8-F50		7.64	67.96		-0.05	-0.44477		-0.02	-0.17791		0.03833
PX9	0.91622	53.6	80.212	17.106	-0.05	-0.074825		-0.02	-0.02993	0	0.519
PX9-F50		21.4	101.65		-0.05	-0.2375		-0.02	-0.095		0.19
PX10	4.3559	66	97.152	25.39	-0.05	-0.0736		-0.02	-0.02944	0	1.2
PX10-F50		19.8	72.112		-0.05	-0.1821		-0.02	-0.07284		0.358

BKGND		45.23	45.23		0	0		0	0		0
f(i)		0.2045	0.2045		1.0000e-04	1.0000e-04		0.0231	0.0231		0.0231

TEST NO.	U, ppm	NL(U), g/m2	Zn, ACL	Zn, ppm	NL(Zn), g/m	Zr, ACL	Zr, ppm	NL(Zr), g/m
PX1	0.036637	0.15876				-0.02	-0.033458	0
PX2	0.018951	0.082122				-0.02	-0.033542	0
PX3	0.043155	0.18682	0.031	0.052669	26.335	-0.02	-0.03398	0
PX4	0.027281	0.1181	0.076	0.12878	64.327	-0.02	-0.03389	0
PX5	0.10444	0.45212	0.12	0.19768	98.838	-0.02	-0.032946	0
PX6	0.094309	0.40826	0.02	0.033924	16.962	-0.02	-0.033924	0
PX7	0.18898	0.67057	-0.02	-0.046604	0	-0.05	-0.11651	0
PX7-F50	0.17645		-0.02	-0.18872		-0.05	-0.47179	
PX8	0.36726	1.5899	-0.02	-0.03166	0	-0.05	-0.07915	0
PX8-F50	0.34096		-0.02	-0.17791		-0.05	-0.44477	
PX9	0.77668	3.3623	-0.02	-0.02993	0	-0.05	-0.074825	0
PX9-F50	0.9025		-0.02	-0.095		-0.05	-0.2375	
PX10	1.7664	7.6468	-0.02	-0.02944	0	-0.05	-0.0736	0
PX10-F50	1.3038		0.02	0.07284		-0.05	-0.1821	

BKGND	0		0	0		0	0	
f(i)	0.0231		0.0002	0.0002		0.0016	0.0016	

TEST NUMBER	RXN TIME	pH	DIL FACTOR	SA/V	LEACHANT VO	ACL NUMBER	Al, ACL	Al, ppm	NL(Al), g/m	B, ACL	B, ppm
PY1	3	9.93	2.3146	1990	10.23	90-0561-05	0.62	1.4351	0.025418	19	43.977
PY2	3	10.07	2.2734	2000	10.56	90-0561-06	0.65	1.4777	0.026523	20	45.468
PY3	7	10.26	2.3377	1996	9.88	90-0561-07	0.85	1.987	0.041327	27.9	65.222
PY4	7	10.25	2.3681	1995	9.85	90-0561-08	0.86	2.0366	0.042782	27.3	64.649
PY5	14	10.45	2.3495	2009	9.94	90-0561-09	0.96	2.2555	0.048784	31.8	74.714
PY6	14	10.45	2.3103	1996	10.04	90-0561-10	0.88	2.0331	0.042659	33.8	78.088
PY7	30	10.47	2.3145	2004	10.14	91-0061-05	0.93	2.1525	0.045934	39.3	90.96
PY8	30	10.52	2.2585	2045	10.04	91-0061-06	0.92	2.0778	0.042902	41.7	94.179
PY9	70	10.61	2.3347	2000	9.92	91-0205-03	1.29	3.0118	0.07086	44.7	104.36
PY10	70	10.66	2.3784	2002	9.9	91-0205-04	1.26	2.9968	0.070357	43.4	103.22
PY11	140	10.72	6.5833	2014	10	91-0310-08	0.44	2.8967	0.067064	20.1	132.32
PY11-F50			3.5315			91-0310-09	0.41	1.4479		0.41	1.4479
PY12	140	10.72	6.8502	2006	10	91-0310-11	0.41	2.8086	0.064794	19	130.15
PY12-F50			3.4323			91-0310-12	0.44	1.5102		0.44	1.5102
PY13	280	11.47	2.3972	1990	10.09	91-0440-01	0.49	1.1746	0.017853	130	311.64
PY14	280	11.48	2.4017	1979	9.95	91-0440-02	0.48	1.1528	0.017315	134	321.83
PY15	560	11.44	10.071	2006	10.09	92-0227-01	-0.1	-1.0071	-0.045156	57.6	580.09
PY15-F50			7.7974			92-0227-02	-0.1	-0.77974		74.9	584.03
PY16	560	11.49	13.048	1996	10.18	92-0227-03	-0.1	-1.3048	-0.054004	43.8	571.5
PY16-F50			10.012			92-0227-04	-0.1	-1.0012		55.6	556.66
PY17	980	11.74	3.974	2004	10.15	PACKET.1	0.184	0.731	0.0049323	185	735.2
PY17-F50			8.584			PACKET.1	0.091	0.781		92.5	794
PY18	980	11.72	4.117	1976	10.39	PACKET.1	0.176	0.725	0.0048267	201	827.5
PY18-F50			8.335			PACKET.1	0.084	0.7		96.6	805.2
BKGND			1				0.56	0.56		0.15	0.15
f(i)			1				0.0173	0.0173		0.03	0.03

TEST NO.	NL(B), g/m2	Ba, ACL	Ba, ppm	NL(Ba), g/m	Ca, ACL	Ca, ppm	NL(Ca), g/m	Cr, ACL	Cr, ppm	NL(Cr), g/m	Cu, ACL
PY1	0.73413	-0.02	-0.046292	-0.016616	0.03	0.069438	-0.34267	-0.02	-0.046292	-0.025847	0.02
PY2	0.7553	-0.02	-0.045468	-0.016239	0.03	0.068202	-0.34105	-0.02	-0.045468	-0.02526	-0.02
PY3	1.0867	-0.02	-0.046754	-0.016731	0.03	0.070131	-0.34158	0.04	0.093508	0.052053	0.08
PY4	1.0777	-0.02	-0.047362	-0.016957	0.03	0.071043	-0.34168	0.03	0.071043	0.039567	0.09
PY5	1.2372	-0.02	-0.04699	-0.016707	0.06	0.14097	-0.33403	0.05	0.11747	0.064972	0.05
PY6	1.3016	-0.02	-0.046206	-0.016535	0.03	0.069309	-0.34164	0.05	0.11552	0.064304	0.1
PY7	1.5105	-0.02	-0.04629	-0.016499	0.03	0.069435	-0.34027	0.11	0.2546	0.14116	0.02
PY8	1.5327	-0.02	-0.04517	-0.015777	0.03	0.067755	-0.33357	0.12	0.27102	0.14725	0.02
PY9	1.7369	-0.02	-0.046694	-0.016676	0.03	0.070041	-0.34091	0.21	0.49029	0.27238	0.05
PY10	1.7162	-0.02	-0.047568	-0.016972	0.03	0.071352	-0.34047	0.14	0.33298	0.1848	0.06
PY11	2.1876	-0.02	-0.13167	-0.046697	-0.02	-0.13167	-0.35371	0.1	0.65833	0.3632	0.02
PY11-F50		-0.02	-0.07063		0.17	0.60036		0.17	0.60036		-0.02
PY12	2.1602	-0.02	-0.137	-0.048784	-0.02	-0.137	-0.35552	0.08	0.54802	0.30354	0.02
PY12-F50		-0.02	-0.068646		0.18	0.61781		0.17	0.58349		-0.02
PY13	5.2175	0.02	0.047944	0.017209	0.05	0.11986	-0.33883	0.7	1.678	0.93693	-0.02
PY14	5.4182	0.02	0.048034	0.017337	0.06	0.1441	-0.33885	-0.02	-0.048034	-0.026969	-0.02
PY15	9.6368	-0.02	-0.20142	-0.071721	0.19	1.9135	-0.20065	0.55	5.5391	3.068	-0.02
PY15-F50		-0.02	-0.15595		0.49	3.8207		0.7	5.4582		-0.02
PY16	9.5416	-0.02	-0.26096	-0.093387	0.12	1.5658	-0.22805	0.28	3.6534	2.0338	-0.02
PY16-F50		-0.02	-0.20024		0.28	2.8033		0.36	3.6042		-0.02
PY17	12.226	0.005	0.02	0.0071286	2.22	8.82	0.32133	1.09	4.332	2.4019	0.012
PY17-F50		0.002	0.017		1.17	10.04		0.509	4.369		0.005
PY18	13.957	0.005	0.021	0.0075911	2.3	9.469	0.37564	0.476	1.96	1.1021	0.007
PY18-F50		0.003	0.025		1.26	10.5		0.233	1.942		0.006
BKGND		0	0		4.57	4.57		0	0		0
f(i)		0.0014	0.0014		0.0066	0.0066		0.0009	0.0009		0.0002

TEST NO.	Cu, ppm	NL(Cu), g/m	Fe, ACL	Fe, ppm	NL(Fe), g/m	K, ACL	K, ppm	NL(K), g/m2	Li, ACL	Li, ppm	NL(Li), g/m
PY1	0.046292	0.11631	0.75	1.7359	0.0098904	14.2	32.867	0.40306	8.97	20.762	0.74343
PY2	-0.045468	-0.11367	0.8	1.8187	0.01031	14.8	33.646	0.41314	9.48	21.552	0.76792
PY3	0.18702	0.46848	1.93	4.5118	0.025628	18.4	43.014	0.55972	12.2	28.52	1.0188
PY4	0.21313	0.53416	2.1	4.973	0.028262	18	42.626	0.55396	11.7	27.707	0.99022
PY5	0.11747	0.29237	3.02	7.0955	0.040044	20.5	48.165	0.63572	13.4	31.483	1.1176
PY6	0.23103	0.57873	2.14	4.944	0.028084	20.9	48.285	0.64174	13.8	31.882	1.1391
PY7	0.04629	0.11549	2.85	6.5963	0.037319	24.8	57.4	0.78042	15.7	36.338	1.2934
PY8	0.04517	0.11044	2.74	6.1883	0.034309	25.9	58.495	0.78141	16.6	37.491	1.3078
PY9	0.11674	0.29184	5.25	12.257	0.069485	28	65.372	0.90577	18.7	43.659	1.5575
PY10	0.1427	0.3564	5.05	12.011	0.068021	26.9	63.979	0.88326	17.7	42.098	1.5002
PY11	0.13167	0.32688	1.47	9.6775	0.054479	11.6	76.366	1.069	6.95	45.754	1.6209
PY11-F50	-0.07063	0.03	0.03	0.10594	0.054479	20.6	72.749		11.9	42.025	
PY12	0.137	0.34149	1.46	10.001	0.056527	10.9	74.667	1.047	6.58	45.074	1.6032
PY12-F50	-0.068646	0.03	0.03	0.10297	0.056527	21.6	74.138		13	44.62	
PY13	-0.047944	-0.12046	0.54	1.2945	0.0073752	62.3	149.35	2.2208	38.4	92.052	3.3023
PY14	-0.048034	-0.12136	0.26	0.62444	0.0035775	66.3	159.23	2.3883	38.8	93.186	3.3616
PY15	-0.20142	-0.50204	-0.02	-0.20142	-0.0011384	28.4	286.02	4.319	10.9	109.77	3.907
PY15-F50	-0.15595	0.05	0.05	0.38987	0.0011384	36.2	282.27		13.9	108.38	
PY16	-0.26096	-0.65371	-0.02	-0.26096	-0.0014823	21.4	279.23	4.235	8.56	111.69	3.9952
PY16-F50	-0.20024	0.05	-0.02	-0.20024	0.0014823	27.5	275.32		10.7	107.13	
	0.048	0.11976	0.055	0.219	0.001239	94.4	375.1	5.7038	24.6	97.76	3.4827
	0.043					52.5	450.7		12.3	105.58	
	0.029	0.073381	0.003	0.012	6.8853e-05	104	428.2	6.6192	26.04	107.21	3.8735
	0.05		0.023	0.192		52.6	438.4		12.6	105.02	
BKGND	0		0	0		7.04	7.04		0.05	0.05	
f(i)	0.0002		0.0882	0.0882		0.0322	0.0322		0.014	0.014	
TEST NO.	Mg, ACL	Mg, ppm	NL(Mg), g/m	Mn, ACL	Mn, ppm	NL(Mn), g/m	Na, ACL	Na, ppm	NL(Na), g/m	Ni, ACL	Ni, ppm
PY1	0.13	0.3009	0.012779	0.1	0.23146	0.0075527	73	168.97	0.66054	0.05	0.11573
PY2	0.16	0.36374	0.016693	0.12	0.27281	0.0088574	76.7	174.37	0.68743	0.05	0.11367
PY3	0.22	0.51429	0.026274	0.29	0.67793	0.022055	94.7	221.38	0.95196	0.09	0.21039
PY4	0.26	0.61571	0.032721	0.34	0.80515	0.026207	92.3	218.58	0.93673	0.11	0.26049
PY5	0.37	0.86931	0.048473	0.53	1.2452	0.040249	107.3	252.1	1.1167	0.18	0.42291
PY6	0.22	0.50827	0.025891	0.33	0.7624	0.024803	108.3	250.21	1.1133	0.07	0.16172
PY7	0.235	0.54391	0.028039	0.467	1.0809	0.035023	120	277.74	1.2624	0.1	0.23145
PY8	0.221	0.49913	0.024705	0.46	1.0389	0.032989	127	286.83	1.2867	0.09	0.20327
PY9	0.53	1.2374	0.071987	1.13	2.6382	0.085656	146	340.87	1.6176	0.25	0.58368
PY10	0.52	1.2368	0.071875	1.1	2.6162	0.084858	140	332.98	1.5719	0.23	0.54703
PY11	0.1	0.65833	0.035092	0.28	1.8433	0.059432	63.5	418.04	2.0345	0.06	0.395
PY11-F50	0.02	0.07063	0.035092	-0.1	-0.35315					-0.03	-0.10594
PY12	0.09	0.61652	0.032593	0.28	1.9181	0.062088	60.6	415.12	2.0263	0.04	0.27401
PY12-F50	0.02	0.068646	0.032593	-0.1	-0.34323					-0.03	-0.10297
PY13	0.05	0.11986	0.0012633	0.1	0.23972	0.0078222	334	800.66	4.2073	-0.03	-0.071916
PY14	0.02	0.048034	-0.0033239	0.04	0.096068	0.0031522	341	818.98	4.3341	-0.03	-0.072051
PY15	-0.01	-0.10071	-0.012665	-0.1	-1.0071	-0.0326	144	1450.2	7.7917	-0.03	-0.30213
PY15-F50	0.03	0.23392	0.012665	-0.1	-0.77974		188	1465.9		-0.03	-0.23392
PY16	-0.01	-0.13048	-0.014617	-0.1	-1.3048	-0.042449	111	1448.3	7.8202	-0.03	-0.39144
PY16-F50	0.04	0.40047	0.014617	-0.1	-1.0012		144	1441.7		-0.03	-0.30035
PY17	0.012	0.048	-0.0032846	0	0.002	6.4805e-05	453	1800	9.7497	0.002	0.008
PY17-F50	0.03	0.258	0.0032846	0.001	0.009		226	1940		0.003	0.026
PY18	0.012	0.049	-0.0032671	0.074	0.305	0.010023	489	2013	11.092	0.001	0.001
PY18-F50	0.035	0.292	0.0032671	0.001	0.008		241	2009		0.001	0.003
BKGND	0.1	0.1		0	0		51.32	51.32		0	0
f(i)	0.0079	0.0079		0.0154	0.0154		0.0895	0.0895		0.0098	0.0098

TEST NO.	NL(Ni), g/m	Si, ACL	Si, ppm	NL(Si), g/m	Th, ACL	Th, ppm	NL(Th), g/m	Ti, ACL	Ti, ppm	NL(Ti), g/m	U, ACL
PY1	0.0059343	55.7	128.92	0.20566	-0.06	-0.13888	0				2.45
PY2	0.0057995	58.2	132.31	0.21291	-0.06	-0.1364	0				2.5
PY3	0.010756	72.5	169.48	0.30441	-0.06	-0.14026	0				3.89
PY4	0.013324	71.5	169.32	0.30416	-0.06	-0.14209	0				3.7
PY5	0.02148	79.3	186.32	0.34341	-0.06	-0.14097	0				3.55
PY6	0.0082676	82.8	191.29	0.35784	-0.06	-0.13862	0				3.65
PY7	0.011785	85.5	197.89	0.37251	-0.01	-0.023145	0	0.05	0.11573	0.014807	2.443
PY8	0.010142	89.2	201.46	0.37357	-0.01	-0.022585	0	0.05	0.11293	0.014159	2.527
PY9	0.029779	112	261.49	0.52874	-0.05	-0.11674	0	0.11	0.25682	0.032925	1.96
PY10	0.027882	111	264	0.53436	-0.05	-0.11892	0	0.11	0.26162	0.033508	1.583
PY11	0.020013	38.6	254.12	0.50717	-0.05	-0.32917	0	0.03	0.1975	0.025144	0.588
PY11-F50		68.3	241.2		-0.05	-0.17658		-0.02	-0.07063		0.956
PY12	0.013938	36.8	252.09	0.50425	-0.05	-0.34251	0	0.03	0.20551	0.026268	0.559
PY12-F50		74	253.99		-0.05	-0.17162		-0.02	-0.068646		1.003
PY13	-0.0036876	173	414.72	0.90793	-0.05	-0.11986	0	-0.02	-0.047944	-0.0061776	1.638
PY14	-0.0037151	177	425.1	0.93864	-0.05	-0.12009	0	-0.02	-0.048034	-0.0062236	1.569
PY15	-0.015369	58.6	590.16	1.3284	-0.05	-0.50355	0	-0.02	-0.20142	-0.025746	0.165
PY15-F50		75.6	589.48		-0.05	-0.38987		-0.02	-0.15595		0.184
PY16	-0.020011	43.3	564.98	1.2733	-0.05	-0.6524	0	-0.02	-0.26096	-0.033523	0.124
PY16-F50		57.7	577.68		-0.05	-0.50059		-0.02	-0.20024		0.139
PY17	0.00040735	131	520.6	1.16	0	0	0	0.002	0.008	0.0010236	0.153
PY17-F50		30.1	515.9		0	0		0.001	0.009		0.06
PY18	5.1640e-05	146	601.1	1.3756	0	0	0	0.002	0.008	0.0010381	0.146
PY18-F50		67	558.4		0	0		0.002	0.017		0.065
BKGND		45.23	45.23		0	0		0	0		0
F(i)		0.2045	0.2045		1.0000e-04	1.0000e-04		0.0039	0.0039		0.0231

TEST NO.	U, ppm	NL(U), g/m2	Zn, ACL	Zn, ppm	NL(Zn), g/m	Zr, ACL	Zr, ppm	NL(Zr), g/m
PY1	5.6708	0.12336				-0.02	-0.046292	-0.014539
PY2	5.6835	0.12302				-0.02	-0.045468	-0.014209
PY3	9.0937	0.19723				-0.02	-0.046754	-0.01464
PY4	8.762	0.19013				-0.02	-0.047362	-0.014838
PY5	8.3407	0.17973				-0.02	-0.04699	-0.014619
PY6	8.4326	0.18289				-0.02	-0.046206	-0.014468
PY7	5.6543	0.12214	0.02	0.04629	0.11549	-0.02	-0.04629	-0.014437
PY8	5.7072	0.12081	0.02	0.04517	0.11044	-0.02	-0.04517	-0.013805
PY9	4.576	0.099048	0.02	0.046694	0.11674	-0.05	-0.11674	-0.03648
PY10	3.765	0.081412	0.06	0.1427	0.3564	-0.05	-0.11892	-0.037125
PY11	3.871	0.083205	-0.02	-0.13167	-0.32688	-0.05	-0.32917	-0.10215
PY11-F50	3.3761		-0.02	-0.07063		-0.05	-0.17658	
PY12	3.8293	0.082637	-0.02	-0.137	-0.34149	-0.05	-0.34251	-0.10671
PY12-F50	3.4426		-0.02	-0.068646		-0.05	-0.17162	
PY13	3.9266	0.085419	-0.02	-0.047944	-0.12046	-0.05	-0.11986	-0.037644
PY14	3.7683	0.08243	-0.02	-0.048034	-0.12136	-0.05	-0.12009	-0.037925
PY15	1.6617	0.03586	-0.02	-0.20142	-0.50204	-0.05	-0.50355	-0.15689
PY15-F50	1.4347		-0.02	-0.15595		-0.05	-0.38987	
PY16	1.618	0.035091	-0.02	-0.26096	-0.65371	-0.05	-0.6524	-0.20428
PY16-F50	1.3916		-0.02	-0.20024		-0.05	-0.50059	
BY17	0.608	0.013134	0.011	0.044	0.10978	0	0	0
BY17-F50	0.515		0.013	0.112		0	0	0
BY18	0.601	0.013167	0.01	0.041	0.10374	0	0	0
BY18-F50	0.542		0.027	0.225		0	0	
BKGND	0		0	0		0	0	
F(i)	0.0231		0.0002	0.0002		0.0016	0.0016	

TEST NUMBER	RXN TIME	pH	DIL FACTOR	SA/V	LEACHANT VO	ACL NUMBER	A1, ACL	A1, ppm	NL(A1), g/m	B, ACL	B, ppm
PZ1	3	11.94	7.8498	19988	5.05	90-0561-11	0.13	1.0205	0.0013316	184	1444.4
PZ2	3	11.86	7.9674	20073	5.04	90-0561-12	0.12	0.95609	0.0011406	195	1553.6
PZ3	7	11.99	8.56	20012	5.02	90-0561-13	0.14	1.1984	0.001844	342	2927.5
PZ4	7	11.98	8.9947	19947	5.05	90-0561-14	0.14	1.2593	0.0020263	322	2896.3
PZ5	14	12.05	9.2751	19954	5	90-0561-15	0.14	1.2985	0.0021394	397	3682.2
PZ6	14	12.04	8.8848	19775	5.09	90-0561-16	0.12	1.0662	0.0014796	401	3562.8
PZ7	28	12.12	9.7485	19953	5.01	91-0061-07	0.14	1.3648	0.0023315	450	4386.8
PZ8	28	12.11	9.7368	20009	4.99	91-0061-08	0.13	1.2658	0.0020389	465	4527.6
PZ9	56	12.12	15.651	19948	5.03	91-0134-11	-0.1	-1.5651	-0.0061578	378	5915.9
PZ10	56	12.14	14.446	19936	5.03	91-0134-08	-0.1	-1.4446	-0.0058124	400	5778.6
PZ11	98	12.14	8.3382	20032	4.99	91-0205-07	0.14	1.1673	0.0017525	665	5544.9
PZ12	98	12.12	8.4729	20059	4.99	91-0205-08	0.14	1.1862	0.0018045	675	5719.2
PZ13	182	12.18	13.226	20016	4.98	91-0239-02	-0.1	-1.3226	-0.0054368	575	7605.2
PZ13-F50			6.8415			91-0239-03	0.16	1.0946		1200	8209.8
PZ14	182	12.19	7.8436	19926	5.01	91-0239-05	0.15	1.1765	0.0017885	820	6431.8
PZ14-F50			7.4865			91-0239-06	0.13	0.97324		747	5592.4
PZ15	364	12.2	28.926	19905	5	91-0596-09	-0.1	-2.8926	-0.010026	448	12959
PZ15-F50			15.592			91-0596-10	-0.1	-1.5592		868	13534
PZ16	364	12.17	29.453	20029	5	91-0596-11	-0.1	-2.9453	-0.010116	405	11928
PZ16-F50			19.341			91-0596-12	-0.1	-1.9341		578	11179
PZ17	728	12.02	26.948	20000	5	92-0485-01	-0.1	-2.6948	-0.0094069	801	21585
PZ17-F50			36.833			92-0485-02	-0.1	-3.6833		533	19632
PZ-18	728	12.05	21.658	19900	5	92-0485-03	-0.1	-2.1658	-0.0079176	883	19124
PZ18-F50			15.863			92-0485-04	-0.1	-1.5863		1260	19987
PZ19	552	12.04	16.594	19923	5	92-0485-05	-0.1	-1.6594	-0.0064392	844	14005
PZ19-F50			16.947			92-0485-06	-0.1	-1.6947		864	14642
PZ20	1100	12.18	10.965	20000	5	PACKET.2	0.649	7.1163	0.018949	1925	21108
PZ20-F25			44.37			PACKET.2	0.125	5.546		688.2	30535
BKGND			1				0.56	0.56		0.15	0.15
f(i)			1				0.0173	0.0173		0.03	0.03

TEST NO.	NL(B), g/m2	Ba, ACL	Ba, ppm	NL(Ba), g/m	Ca, ACL	Ca, ppm	NL(Ca), g/m	Cr, ACL	Cr, ppm	NL(Cr), g/m	Cu, ACL
PZ1	2.4085	-0.02	-0.157	-0.0056104	0.04	0.31399	-0.032262	0.02	0.157	0.0087272	-0.02
PZ2	2.5797	-0.02	-0.15935	-0.0056703	0.01	0.079674	-0.033894	0.04	0.3187	0.017641	-0.02
PZ3	4.876	-0.02	-0.1712	-0.0061106	0.08	0.6848	-0.029416	0.07	0.5992	0.033269	-0.02
PZ4	4.8397	-0.02	-0.17989	-0.0064419	0.06	0.53968	-0.030614	0.03	0.26984	0.015031	-0.02
PZ5	6.1509	-0.02	-0.1855	-0.0066403	0.08	0.74201	-0.029067	0.31	2.8753	0.16011	-0.02
PZ6	6.0053	-0.02	-0.1777	-0.0064185	0.08	0.71078	-0.029569	0.06	0.53309	0.029953	-0.02
PZ7	7.3283	0.04	0.38994	0.013959	0.15	1.4623	-0.023599	0.02	0.19497	0.010857	-0.02
PZ8	7.5424	0.04	0.38947	0.013903	0.09	0.87631	-0.02797	0.23	2.2395	0.12436	-0.02
PZ9	9.8853	0.03	0.46951	0.016812	0.09	1.4085	-0.024013	-0.02	-0.31301	-0.017435	0.01
PZ10	9.6616	0.03	0.43339	0.015528	0.09	1.3002	-0.024851	-0.02	-0.28893	-0.016103	-0.01
PZ11	9.2265	0.04	0.33353	0.011893	0.2	1.6676	-0.021952	0.57	4.7528	0.26362	-0.02
PZ12	9.5037	0.04	0.33892	0.012069	0.18	1.5251	-0.022999	0.57	4.8296	0.26752	-0.02
PZ13	12.665	0.04	0.52906	0.01888	0.21	2.7776	-0.013568	-0.02	-0.26453	-0.014684	-0.02
PZ13-F50		0.07	0.4789		0.43	2.9418		-0.02	-0.13683		-0.02
PZ14	10.759	0.06	0.47062	0.01687	0.31	2.4315	-0.016261	-0.02	-0.15687	-0.0087475	-0.02
PZ14-F50		0.05	0.37433		0.32	2.3957		-0.02	-0.14973		-0.02
PZ15	21.701	0.05	1.4463	0.0519	0.17	4.9174	0.0026445	-0.02	-0.57852	-0.032293	-0.02
PZ15-F50		0.07	1.0914		0.31	4.8335		-0.02	-0.31184		-0.02
PZ16	19.852	0.04	1.1781	0.042015	0.14	4.1234	-0.0033783	-0.02	-0.58906	-0.032678	-0.02
PZ16-F50		0.07	1.3539		0.38	7.3496		-0.02	-0.38682		-0.02
PZ17	35.975	0.12	3.2338	0.11549	0.2	5.3896	0.0062091	-0.02	-0.53896	-0.029942	-0.02
PZ17-F50		0.09	3.315		0.2	7.3666		-0.02	-0.73666		-0.02
PZ-18	32.033	0.12	2.599	0.093286	0.22	4.7648	0.0014829	-0.02	-0.43316	-0.024185	-0.02
PZ18-F50		0.16	2.5381		0.4	6.3452		-0.02	-0.31726		-0.02
PZ19	23.432	0.07	1.1616	0.041645	0.22	3.6507	-0.0069915	-0.02	-0.33188	-0.018509	0.02
PZ19-F50		0.08	1.3558		0.32	5.423		-0.02	-0.33894		-0.02
PZ20	35.179	0.796	8.7281	0.31172	5.541	60.757	0.42566	0	0	0	0.25
PZ20-F25		0.0856	3.7981		1.336	59.278		0.00031	0.013755		0
BKGND		0	0		4.57	4.57		0	0		0
F(i)		0.0014	0.0014		0.0066	0.0066		0.0009	0.0009		0.0002

TEST NO.	Cu, ppm	NL(Cu), g/m	Fe, ACL	Fe, ppm	NL(Fe), g/m	K, ACL	K, ppm	NL(K), g/m2	Li, ACL	Li, ppm	NL(Li), g/m
PZ1	-0.157	-0.039273	0.02	0.157	8.9053e-05	88.7	696.28	1.0709	37.5	294.37	1.0518
PZ2	-0.15935	-0.039692	0.02	0.15935	9.0005e-05	92.7	738.58	1.1318	37.9	301.96	1.0743
PZ3	-0.1712	-0.042774	0.02	0.1712	9.6994e-05	164	1403.8	2.1676	36.5	312.44	1.115
PZ4	-0.17989	-0.045093	0.01	0.089947	5.1126e-05	155	1394.2	2.1597	35.7	321.11	1.1497
PZ5	-0.1855	-0.046482	0.8	7.4201	0.0042161	192	1780.8	2.7607	33.1	307.01	1.0988
PZ6	-0.1777	-0.044929	0.02	0.1777	0.00010188	195	1732.5	2.7098	34.4	305.64	1.1038
PZ7	-0.19497	-0.048857	0.03	0.29245	0.00016618	203	1978.9	3.0692	32	311.95	1.1166
PZ8	-0.19474	-0.048662	0.03	0.2921	0.00016552	209	2035	3.1476	30.7	298.92	1.0669
PZ9	0.15651	0.039228	0.01	0.15651	8.8953e-05	173	2707.5	4.2042	19.9	311.44	1.115
PZ10	-0.14446	-0.036232	0.01	0.14446	8.2159e-05	183	2643.7	4.1073	20.9	301.93	1.0816
PZ11	-0.16676	-0.041624	-0.02	-0.16676	-9.4386e-05	302	2518.1	3.893	30.7	255.98	0.91259
PZ12	-0.16946	-0.04224	-0.02	-0.16946	-9.5782e-05	303	2567.3	3.9638	31	262.66	0.93513
PZ13	-0.26453	-0.06608	0.08	1.0581	0.00059936	271	3584.4	5.5504	24.1	318.76	1.1373
PZ13-F50	-0.13683		0.09	0.61573		556	3803.9		49.2	336.6	
PZ14	-0.15687	-0.039364	0.06	0.47062	0.00026778	380	2980.6	4.6344	34.2	268.25	0.96142
PZ14-F50	-0.14973		0.09	0.67379		350	2620.3		31.7	237.32	
PZ15	-0.57852	-0.14532	0.02	0.57852	0.00032952	190	5495.9	8.5638	14.4	416.53	1.4945
PZ15-F50	-0.31184		-0.02	-0.31184		360	5613.1		13.6	212.05	
PZ16	-0.58906	-0.14705	0.03	0.88359	0.00050018	168	4948.1	7.6614	13.6	400.56	1.4283
PZ16-F50	-0.38682		0.04	0.77364		241	4661.2		19.4	375.22	
PZ17	-0.53896	-0.13474	-0.02	-0.53896	-0.00030553	277	7464.6	11.58	23.7	638.67	2.2808
PZ17-F50	-0.73666		-0.02	-0.73666		180	6629.9		15.8	581.96	
PZ-18	-0.43316	-0.10883	-0.02	-0.43316	-0.00024679	313	6779	10.568	26.9	582.6	2.091
PZ18-F50	-0.31726		0.05	0.79315		449	7122.5		38.4	609.14	
PZ19	0.33188	0.083291	-0.02	-0.33188	-0.00018887	315	5227.1	8.137	24.2	401.57	1.4396
PZ19-F50	-0.33894		0.03	0.50841		323	5473.9		25.2	427.06	
PZ20	2.7413	0.68531	0	0	0	711	7796.1	12.095	71.42	783.12	2.7967
PZ20-F25	0		0.026	1.1536		245.2	10880		30.53	1354.6	
BKGND	0		0	0		7.04	7.04		0.05	0.05	
f(i)	0.0002		0.0882	0.0882		0.0322	0.0322		0.014	0.014	

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TEST NO.	Mg, ACL	Mg, ppm	NL(Mg), g/m	Mn, ACL	Mn, ppm	NL(Mn), g/m	Na, ACL	Na, ppm	NL(Na), g/m	Ni, ACL	Ni, ppm
P21	-0.01	-0.078498	-0.0011304	-0.01	-0.078498	-0.00025502	421	3304.8	1.8187	-0.03	-0.23549
P22	-0.01	-0.079674	-0.001133	-0.01	-0.079674	-0.00025774	451	3593.3	1.9716	-0.03	-0.23002
P23	-0.01	-0.0856	-0.001174	-0.01	-0.0856	-0.0002776	769	6582.6	3.6466	-0.03	-0.22568
P24	-0.01	-0.089947	-0.0012054	-0.01	-0.089947	-0.00028281	726	6330.2	3.6291	-0.03	-0.26984
P25	0.01	0.092751	-4.5986e-05	0.01	0.092751	-0.00030183	891	8264.1	4.5987	0.12	1.113
P26	-0.01	-0.088848	-0.0012088	-0.01	-0.088848	-0.00029175	906	7996.3	4.489	-0.03	-0.26654
P27	0.014	0.13648	0.00023142	-0.01	-0.097485	-0.00031726	944	9202.6	5.1245	-0.03	-0.29245
P28	-0.01	-0.097368	-0.0012486	-0.01	-0.097368	-0.00031599	975	9493.4	5.2725	-0.03	-0.2921
P29	-0.01	-0.15651	-0.0016277	-0.01	-0.15651	-0.00050946	838	13115	7.3172	-0.03	-0.46951
P30	-0.01	-0.14446	-0.0015522	-0.01	-0.14446	-0.00047054	876	12655	7.3172	-0.03	-0.43359
P31	0.05	0.41691	0.0020026	-0.01	-0.083382	-0.00027029	1430	11924	6.622	-0.03	-0.25015
P32	-0.01	-0.084729	-0.0011657	-0.01	-0.084729	-0.00027429	1400	11862	6.5788	-0.03	-0.25419
P33	-0.01	-0.13226	-0.0014689	-0.01	-0.13226	-0.00042909	1250	16533	9.2004	-0.03	-0.39679
P34	-0.01	-0.068415	-0.0011335	-0.01	-0.068415	-0.00025561	2630	18130	7.844	-0.03	-0.20524
P35	-0.01	-0.078436	-0.0024754	-0.01	-0.078436	-0.00025561	1790	14040	7.844	-0.03	-0.23531
P36	-0.01	-0.078436	-0.0024754	-0.01	-0.078436	-0.00025561	1620	12128	14.812	-0.03	-0.22459
P37	-0.01	-0.28926	-0.0024934	-0.01	-0.28926	-0.00094364	914	26438	14.812	-0.03	-0.86778
P38	-0.01	-0.29453	-0.0024934	-0.01	-0.29453	-0.00094364	1758	27411	13.641	-0.03	-0.46776
P39	-0.01	-0.26948	-0.0023385	-0.01	-0.26948	-0.00094364	832	24505	22.93	-0.03	-0.88359
P40	-0.01	-0.21658	-0.0020137	-0.01	-0.21658	-0.00070672	1189	22996	20.473	-0.03	-0.58023
P41	-0.01	-0.31726	-0.0016897	-0.01	-0.31726	-0.00070672	1525	41096	15.178	-0.03	-0.80844
P42	-0.01	-0.16594	-0.001109	-0.01	-0.16594	-0.00054085	1014	37349	23.849	-0.03	-1.105
P43	0.02	0.33894	0.001109	-0.01	-0.16594	-0.00054085	1686	36515	0	-0.03	-0.64974
P44	0.0251	0.27522	0.001109	0.00213	0.023355	7.5829e-05	2411	38246	0	-0.03	-0.47589
P45	0.0394	1.7482	0.001109	0.000674	0.023355	7.5829e-05	1634	27115	0	0.03	0.43782
P46	0.0079	0.0079	0.001109	0	0	0	1691	28657	0	-0.03	-0.50841
P47	0.0079	0.0079	0.001109	0	0	0	3898	42742	0	0	0
P48	0.0079	0.0079	0.001109	0	0	0	1138	50493	0	0	0
P49	0.0079	0.0079	0.001109	0	0	0	51.32	51.32	0.0098	0.0098	0.0098
P50	0.0079	0.0079	0.001109	0.0154	0.0154	0.0154	0.0895	0.0895	0.0895	0.0895	0.0895
P51	0.0079	0.0079	0.001109	0.0154	0.0154	0.0154	0.0895	0.0895	0.0895	0.0895	0.0895
P52	0.0079	0.0079	0.001109	0.0154	0.0154	0.0154	0.0895	0.0895	0.0895	0.0895	0.0895
P53	0.0079	0.0079	0.001109	0.0154	0.0154	0.0154	0.0895	0.0895	0.0895	0.0895	0.0895
P54	0.0079	0.0079	0.001109	0.0154	0.0154	0.0154	0.0895	0.0895	0.0895	0.0895	0.0895
P55	0.0079	0.0079	0.001109	0.0154	0.0154	0.0154	0.0895	0.0895	0.0895	0.0895	0.0895
P56	0.0079	0.0079	0.001109	0.0154	0.0154	0.0154	0.0895	0.0895	0.0895	0.0895	0.0895
P57	0.0079	0.0079	0.001109	0.0154	0.0154	0.0154	0.0895	0.0895	0.0895	0.0895	0.0895
P58	0.0079	0.0079	0.001109	0.0154	0.0154	0.0154	0.0895	0.0895	0.0895	0.0895	0.0895
 

TEST NO.	NL(N ₁), g/m	SL, ACL	SL, ppm	NL(S ₁), g/m	Th, ACL	Th, ppm	NL(Th), g/m	Ti, ACL	Ti, ppm	NL(Ti), g/m	U, ACL
PZ1	-0.0012022	126	989.07	0.23091	-0.06	-0.47099	-0.23564	-0.01	-0.097485	-0.0012528	0.198
PZ2	-0.0012151	127	1011.9	0.23548	-0.06	-0.47804	-0.23815	-0.01	-0.097368	-0.0012477	0.183
PZ3	-0.0013094	137	1172.7	0.2755	-0.06	-0.5136	-0.25665	-0.01	-0.15651	-0.0020117	0.202
PZ4	-0.0013804	132	1187.3	0.27998	-0.06	-0.53968	-0.27056	-0.02	-0.14446	-0.0018858	0.221
PZ5	0.0056917	135	1252.1	0.29577	-0.06	-0.55651	-0.27889	-0.02	-0.16976	-0.0021346	0.233
PZ6	-0.0013754	140	1293.9	0.2964	-0.06	-0.53309	-0.26958	-0.02	-0.16466	-0.0021661	0.171
PZ7	-0.0014956	129	1257.6	0.29711	-0.1	-0.57485	-0.48857	-0.01	-0.26453	-0.0033887	0.282
PZ8	-0.0014897	127	1236.6	0.29115	-0.1	-0.97368	-0.48662	-0.01	-0.13683	-0.0020186	0.322
PZ9	-0.0024017	93.5	1463.3	0.34763	-0.1	-1.5651	-0.78456	-0.01	-0.15687	-0.0020186	0.262
PZ10	-0.0022183	93.5	1350.7	0.32022	-0.1	-1.4446	-0.72464	-0.01	-0.14973	-0.0074523	0.0625
PZ11	-0.0012742	182	1517.6	0.35941	-0.05	-0.41691	-0.20812	-0.02	-0.57852	-0.0075411	0.0574
PZ12	-0.0012931	182	1542.1	0.3649	-0.05	-0.42365	-0.2112	-0.02	-0.58906	-0.0075411	0.115
PZ13	-0.0020228	137	1812	0.43164	-0.05	-0.66132	-0.3304	-0.02	-0.38682	-0.0069097	0.0778
PZ13-F50		286	1956.7	0.3604	-0.05	-0.34207	-0.19682	-0.02	-0.73526	-0.0069097	0.078
PZ14	-0.001205	193	1513.8	0.3604	-0.05	-0.39218	-0.19682	-0.02	-0.43316	-0.0055812	0.124
PZ14-F50		194	1452.4	0.53606	-0.05	-0.37433	-0.7266	-0.02	-0.31726	-0.0042713	0.176
PZ15	-0.0044486	77	2227.3	0.53606	-0.05	-1.4463	-0.7266	-0.02	-0.33888	-0.0042713	0.093
PZ15-F50		142	2214.1	0.53606	-0.05	-1.4776	-0.7266	-0.02	-0.33894	-0.0042713	0.096
PZ16	-0.0045016	71.6	2108.8	0.50382	-0.05	-1.4726	-0.73526	-0.02	0	0	0.067
PZ16-F50		102	1972.8	0.50382	-0.05	-0.96705	-0.73526	-0.02	0	0	0.067
PZ17	-0.0041247	124	3341.6	0.80595	-0.05	-1.3474	-0.6737	-0.02	0	0	0.067
PZ17-F50		83.5	3053.6	0.80595	-0.05	-1.8417	-0.6737	-0.02	0	0	0.067
PZ17-F50		141	3075.8	0.73928	-0.05	-1.9829	-0.54417	-0.02	0	0	0.067
PZ18	-0.0033317	200	3172.6	0.73928	-0.05	-0.79315	-0.54417	-0.02	0	0	0.067
PZ18-F50		149	2472.5	0.59576	-0.05	-0.8297	-0.41645	-0.02	0	0	0.067
PZ19	0.0025497	154	2609.8	0.59576	-0.05	-0.84735	-0.41645	-0.02	0	0	0.067
PZ19-F50		154	2609.8	0.62703	-0.05	-0.84735	-0.41645	-0.02	0	0	0.067
PZ20	0	418	4583	0.62703	0	0	0	0	0	0	0.067
PZ20-F25		365	16195	0.62703	0	0	0	0	0	0	0.067
BKGD		45.23	45.23	0	1.00000-04	1.00000-04	1.00000-04	0.0039	0.0039	0.0039	0.06231
f(L)		0.2045	0.2045	0.2045	1.00000-04	1.00000-04	1.00000-04	0.0039	0.0039	0.0039	0.06231

TEST NO.	U, ppm	NL(U), g/m2	Zn, ACL	Zn, ppm	NL(Zn), g/m	Zr, ACL	Zr, ppm	NL(Zr), g/m
PZ1	1.5543	0.0033662				-0.02	-0.157	-0.0049091
PZ2	1.458	0.0031444				-0.02	-0.15935	-0.0049615
PZ3	1.7291	0.0037404				-0.02	-0.1712	-0.0053468
PZ4	1.9878	0.0043141				-0.02	-0.17989	-0.0056366
PZ5	1.6788	0.0036421				-0.02	-0.1855	-0.0058103
PZ6	1.3683	0.0029953				-0.02	-0.1777	-0.0056162
PZ7	2.4079	0.0052241	0.02	0.19497	0.048857	-0.02	-0.19497	-0.0061072
PZ8	2.22	0.004803	0.01	0.097368	0.024331	-0.02	-0.19474	-0.0060828
PZ9	2.4258	0.0052644	0.01	0.15651	0.039228	-0.02	-0.31301	-0.0098071
PZ10	2.5137	0.0054583	0.01	0.14446	0.036232	-0.02	-0.28893	-0.009058
PZ11	1.9428	0.0041985	-0.02	-0.16676	-0.041624	-0.05	-0.41691	-0.013008
PZ12	2.3131	0.004992	-0.02	-0.16946	-0.04224	-0.05	-0.42365	-0.0132
PZ13	2.2617	0.0048916	0.04	0.52906	0.13216	-0.05	-0.66132	-0.02065
PZ13-F50	1.9293		-0.02	-0.13683		-0.05	-0.34207	
PZ14	2.5256	0.0054871	0.03	0.23531	0.059045	-0.05	-0.39218	-0.012301
PZ14-F50	1.9615		-0.02	-0.14973		-0.05	-0.37433	
PZ15	1.8079	0.0039318	-0.02	-0.57852	-0.14532	-0.05	-1.4463	-0.045413
PZ15-F50	1.7931		0.31	4.8335		-0.05	-0.7796	
PZ16	1.6906	0.003654	0.03	0.88359	0.22058	-0.05	-1.4726	-0.045954
PZ16-F50	1.5047		0.45	8.7034		-0.05	-0.96705	
PZ17	3.126	0.0067662	-0.02	-0.53896	-0.13474	-0.05	-1.3474	-0.042106
PZ17-F50	2.873		0.02	0.73666		-0.05	-1.8417	
PZ-18	2.6856	0.0058422	-0.02	-0.43316	-0.10883	-0.05	-1.0829	-0.034011
PZ18-F50	2.7919		-0.02	-0.31726		-0.05	-0.79315	
PZ19	1.5432	0.0033533	0.12	1.9913	0.49974	-0.05	-0.8297	-0.026028
PZ19-F50	1.6269		-0.02	-0.33894		-0.05	-0.84735	
PZ20	8.9913	0.019462	0	0	0	0.00345	0.037829	0.0011822
PZ20-F25	2.9728		0	0	0	0.000594	0.026356	
BKGND	0		0	0		0	0	
f(i)	0.0231		0.0002	0.0002		0.0016	0.0016	

TEST NUMBER	RXN TIME	pH	DIL FACTOR	SA/V	LEACHANT	ACL NUMBER	Al, ACL	Al, ppm	NL(Al), g/m	B, ACL	B, ppm
TW1	14	8.95	13.48	321	2.05	91-0532-01	0.27	3.641	0.5546	0.69	9.236
TW2	14	9.31	25.03	327	2.03	91-0532-02	0.18	4.506	0.6788	0.37	9.262
TW2-F50	14		35.34			91-0532-03	-0.1	-3.534		0.24	8.482
TW3	28	9.24	34.47	323	2.04	91-0533-03	0.12	4.136	0.6308	0.29	9.995
TW3-F50	28		24.94			91-0533-04	-0.1	-2.494		0.41	10.22
TW4	28	9.28	28.89	319	2.05	91-0533-01	0.2	5.778	0.8922	0.36	10.4
TW4-F25	28		19.59			91-0533-02	-0.1	-1.959		0.46	9.013
TW5	56	9.5	11.44	322	2.05	91-0596-01	0.4	4.578	0.7003	1.05	12.02
TW6	56	9.44	11.37	317	2.06	91-0596-02	0.38	4.321	0.672	1.04	11.83
TW7	91	9.36	41.54	321	2.06	91-0639-01	-0.1	-4.154		0.37	15.37
TW7-F50	91		21.67			91-0639-02	-0.1	-2.167		0.76	16.47
TW8	91	9.41	10.8	321	2.06	91-0639-03	0.44	4.754	0.7295	1.53	16.53
TW9	180	9.4	29.81	323	2.05	92-0134-07	0.17	5.067	0.7728	0.62	18.48
TW9-F50	180		18.99			92-0134-08	-0.1	-1.899		0.99	18.8
TW10	180	9.54	28.94	317	2.05	92-0134-09	0.1	2.894	0.4498	0.61	17.66
TW10-F50	180		18.23			92-0134-10	-0.1	-1.823		0.98	17.87
TW11	360	9.63	11.84	322	2.05	PACKET.1	0.411	4.865	0.7554	1.93	22.84
TW12	360	9.59	14.59	325	2.05	PACKET.1	0.269	3.925	0.6039	1.66	24.22
TW13	540	10.48	4.6	320	2.05	PACKET.1					
TW14	540	9.58	4.416	323	2.06	PACKET.1	0.812	3.586	0.5551	3.9	17.22
TW15	720	9.31	6.795	319	2.05	PACKET.1	0.234	1.59	0.2492	2.97	20.18
TW16	720	9.75	6.994	319	2.05	PACKET.1	0.084	0.5875	0.0921	10.4	72.74
BKGND			1				0	0		0	0
F(i)			1				0.0203	0.0203		0.0248	0.0248

TEST No.	NL(B), g/m2	Ba, ACL	Ba, ppm	NL(Ba), g/m	Ca, ACL	Ca, ppm	NL(Ca), g/m	Cr, ACL	Cr, ppm	NL(Cr), g/m	Cu, ACL
TW1	1.16	-0.02	-0.268	0	-0.02	-0.2677	0	-0.02	-0.2677	0	-0.02
TW2	1.142	-0.02	-0.5006	0	-0.02	-0.5006	0	-0.02	-0.5006	0	-0.02
TW2-F50		-0.02	-0.7068		-0.02	-0.7068		-0.02	-0.7068		-0.02
TW3	1.248	-0.02	-0.6893	0	-0.02	-0.6893	0	-0.02	-0.6893	0	-0.02
TW3-F50		-0.02	-0.4987		-0.02	-0.4987		-0.02	-0.4987		-0.02
TW4	1.315	-0.02	-0.5778	0	-0.02	-0.5778	0	-0.02	-0.5778	0	-0.02
TW4-F25		-0.02	-0.3919		-0.02	-0.3919		-0.02	-0.3919		-0.02
TW5	1.505	-0.02	-0.2289	0	0.03	0.3433	0.124	-0.02	-0.2289	0	0.03
TW6	1.505	-0.02	-0.2276	0	0.04	0.4552	0.167	-0.02	-0.2276	0	0.03
TW7		-0.02	-0.8308		-0.02	-0.8308		-0.02	-0.8308		-0.02
TW7-F50		-0.02	-0.4334		0.04	0.8668	0.314	-0.02	-0.4334	0	-0.02
TW8	2.076	-0.02	-0.2161	0	0.05	0.5402	0.1957	-0.02	-0.2161	0	0.04
TW9	2.307	-0.02	-0.5962	0	0.03	0.8942	0.3219	-0.02	-0.5962	0	-0.02
TW9-F50		-0.02	-0.3798		0.03	0.5696		-0.02	-0.3798		-0.02
TW10	2.246	-0.02	-0.5789	0	-0.02	-0.5789	0	-0.02	-0.5789	0	-0.02
TW10-F50	2.272	-0.02	-0.3646	0	0.03	0.5469	0.2006	-0.02	-0.3646	0	-0.02
TW11	2.838	0.019	0.2249	0.3492	0.238	2.817	0.9721	0.017	0.2012	1.25	0.062
TW12	2.981	0.006	0.0876	0.1347	0.144	2.101	0.7184	0.011	0.1605	0.9878	0.025
TW13											
TW14	2.133	0.023	0.1016	0.1572	0.183	2.133	0.7337	0.038	0.1678	1.039	0.124
TW15	2.531	0.011	0.0747	0.1172	0.427	2.902	1.011	0.021	0.1427	0.8947	0.022
TW16	9.121	0.003	0.021	0.0329	0.582	4.07	1.418	0.046	0.3217	2.017	0.005
BKGND		0	0		0	0		0	0		
F(i)		0.002	0.002		0.0086	0.0086		0.0005	0.0005		

TEST No.	Cu, ppm	NL(Cu), g/m	Fe, ACL	Fe, ppm	NL(Fe), g/m	K, ACL	K, ppm	NL(K), g/m2	Li, ACL	Li, ppm	NL(Li), g/m
TW1	-0.2677	0	0.36	4.819	0.1881	0.89	11.91	1.201	0.64	8.566	1.355
TW2	-0.5006	0	0.26	6.508	0.2494	0.54	13.52	1.338	0.35	8.761	1.36
TW2-F50	-0.7068		-0.02	-0.7068		0.47	16.61		0.22	7.775	
TW3	-0.6893	0	0.19	6.549	0.2541	0.48	16.54	1.658	0.26	8.961	1.408
TW3-F50	-0.4987		-0.02	-0.4987		0.63	15.71		0.35	8.728	
TW4	-0.5778	0	0.31	8.956	0.3518	1.18	34.09	3.458	0.31	8.956	1.425
TW4-F25	-0.3919		-0.02	-0.3919		0.56	10.97		0.35	6.857	
TW5	0.3433	0.3332	0.83	9.499	0.3697	1.1	12.59	1.265	0.95	10.87	1.714
TW6	0.3414	0.3365	0.77	8.762	0.3464	1.1	12.52	1.278	0.96	10.92	1.749
TW7	-0.8308		0.23	9.554		0.41	17.03		0.34	14.12	2.233
TW7-F50	-0.4334	0	-0.02	-0.4334	0	0.76	16.47	1.66	0.64	13.87	
TW8	0.4322	0.4207	0.94	10.16	0.3965	1.6	17.29	1.743	1.37	14.8	2.341
TW9	-0.5962	0	0.32	9.538	0.3701	0.57	16.99	1.702	0.56	16.69	2.623
TW9-F50	-0.3798		-0.02	-0.3798		0.87	16.52		0.87	16.52	
TW10	-0.5789	0	0.17	4.921	0.1945	0.52	15.05	1.537	0.55	15.92	2.549
TW10-F50	-0.365	0	-0.02	-0.3646	0	0.8	14.58		0.87	15.86	
TW11	0.7339	0.7597	0.469	5.552	0.2155	1.51	17.87	1.791	1.64	19.41	3.06
TW12	0.3648	0.3742	0.137	1.999	0.0769	1.2	17.51	1.738	1.39	20.28	3.168
TW13											
TW14	0.5476	0.5651	1.3	5.741	0.2222	3.96	17.49	1.747	3.83	16.91	2.658
TW15	0.1495	0.1562	0.293	1.991	0.078	2.69	18.28	1.848	2.72	18.48	2.941
TW16	0.035	0.0365	0.129	0.9022	0.0354	5.87	41.05	4.152	7.06	49.38	7.857
BKGND	0		0	0		0	0		0	0	
F(i)	0.0032		0.0798	0.0798		0.0309	0.0309		0.0197	0.0197	

TEST No.	Mg, ACL	Mg, ppm	NL(Mg), g/m	Mn, ACL	Mn, ppm	NL(Mn), g/m	Na, ACL	Na, ppm	NL(Na), g/m	Ni, ACL	Ni, ppm
TW1	0.05	0.6693	0.2606	0.06	0.8031	0.1787	2.07	27.71	1.306	-0.03	-0.4015
TW2	0.04	1.001	0.3827	0.05	1.252	0.2734	1.12	28.04	1.297	-0.03	-0.751
TW2-F50	-0.01	-0.3534		-0.01	-0.3534		0.78	27.57		-0.03	-1.06
TW3	0.02	0.6893	0.2668	0.03	1.034	0.2287	0.89	30.68	1.437	-0.03	-1.034
TW3-F50	-0.01	-0.2494		-0.01	-0.2494		1.23	30.67		-0.03	-0.7481
TW4	0.04	1.156	0.4528	0.05	1.444	0.3234	1.06	30.62	1.452	-0.03	-0.8667
TW4-F25	-0.01	-0.1959		-0.01	-0.1959		1.48	29		-0.03	-0.5878
TW5	0.11	1.259	0.4887	0.15	1.717	0.3808	3.1	35.48	1.667	0.03	0.3433
TW6	0.1	1.138	0.4487	0.14	1.593	0.359	3.07	34.93	1.667	-0.03	-0.3414
TW7	0.03	1.246		0.04	1.662		1.12	46.53	2.193	-0.03	-1.246
TW7-F50	0.01	0.2167	0.08439	-0.01	-0.2167	0	2.83	61.33		-0.03	-0.6501
TW8	0.14	1.513	0.589	0.17	1.837	0.4087	4.48	48.4	2.281	-0.03	-0.3241
TW9	0.06	1.788	0.6921	0.06	1.788	0.3955	1.88	56.04	2.625	-0.03	-0.8942
TW9-F50	-0.01	-0.1899		-0.01	-0.1899		3.04	57.72		-0.03	-0.5696
TW10	0.03	0.8683	0.3424	0.03	0.8683	0.1957	1.8	52.1	2.486	-0.03	-0.8683
TW10-F50	-0.01	-0.1823	0	-0.01	-0.1823	0	3.03	55.24		-0.03	-0.5469
TW11	0.127	1.503	0.5836	0.232	2.743	0.6092	5.24	62.03	2.914	0.054	0.6392
TW12	0.045	0.6567	0.2526	0.082	1.197	0.263	4.5	65.67	3.057	0.021	0.3065
TW13											
TW14	0.222	0.9804	0.3794	0.328	1.449	0.3203	12.6	55.64	2.606	0.112	0.4946
TW15	0.04	0.2718	0.1065	0.072	0.4893	0.1096	9.1	61.84	2.933	0.041	0.2786
TW16	0.013	0.0909	0.0356	0.014	0.0979	0.0219	26.7	186.7	8.856	0.027	0.1888
BKGND	0	0		0	0		0	0		0	0
F(i)	0.008	0.008		0.014	0.014		0.0661	0.0661		0.0065	0.0065

TEST No.	NL(Ni), g/m	Si, ACL	Si, ppm	NL(Si), g/m	Th, ACL	Th, ppm	NL(Th), g/m	Ti, ACL	Ti, ppm	NL(Ti), g/m	U, ACL
TW1	0	4.86	65.05	0.8861	-0.05	-0.6693	0	-0.02	-0.2677	0	0.0645
TW2	0	2.86	71.59	0.9573	-0.05	-1.252	0	-0.02	-0.5006	0	0.0335
TW2-F50		1.74	61.49		-0.05	-1.767		-0.02	-0.7068		0.0123
TW3	0	2.17	74.79	1.012	-0.05	-1.723	0	-0.02	-0.6893	0	0.0204
TW3-F50		2.62	65.33		-0.05	-1.247		-0.02	-0.4987		0.0097
TW4	0	2.66	76.84	1.053	-0.05	-1.444	0	-0.02	-0.5778	0	0.0268
TW4-F25		2.92	57.21		-0.05	-0.9796		-0.02	-0.3919		0.0124
TW5	0	7.6	86.98	1.181	-0.05	-0.5722	0	0.03	0.3433	0	0.0742
TW6	0	7.48	85.12	1.174	-0.05	-0.569	0	-0.02	-0.2276	0	0.0767
TW7		2.46	102.2		-0.05	-2.077		-0.02	-0.8308		0.034
TW7-F50	0	4.37	94.7	1.29	-0.05	-1.084	0	-0.02	-0.4334	0	0.0378
TW8	0	9.96	107.6	1.466	-0.05	-0.5402	0	0.04	0.4322	0	0.153
TW9	0	4.36	130	1.759	-0.05	-1.49	0	-0.02	-0.5961	0	0.0834
TW9-F50		6.22	118.1		-0.05	-0.9494		-0.02	-0.3798		0.085
TW10	0	3.84	111.1	1.533	-0.05	-1.447	0	-0.02	-0.5789	0	0.0642
TW10-F50	0	5.74	104.6	1.443	-0.05	-0.9115	0	-0.02	-0.3646	0	0.0691
TW11	0.2836	11.94	141.3	1.917	0.07	0.8286	1.287	0.051	0.6037	0.375	0.179
TW12	0.1347	8.91	130	1.747	0.04	0.5837	0.898	0.025	0.3648	0.2245	0.141
TW13											
TW14	0.2188	25.6	113.1	1.528	0.106	0.4681	0.7246	0.085	0.3754	0.2324	0.301
TW15	0.1248	14.4	97.85	1.339	0.097	0.6591	1.033	0.032	0.2174	0.1363	0.173
TW16	0.0846	23.4	163.7	2.24	0.019	0.1329	0.2083	0.005	0.035	0.0219	0.553
BKGND		0	0		0	0		0	0		0
f(i)		0.2287	0.2287		0.0023	0.0023		0.0054	0.0054		0.0164

TEST No.	U, ppm	NL(U), g/m2	Zn, ACL	Zn, ppm	NL(Zn), g/m	Zr, ACL	Zr, ppm	NL(Zr), g/m
TW1	0.8633	0.164	-0.02	-0.2677	0	-0.05	-0.6693	0
TW2	0.8386	0.1564	-0.02	-0.5006	0	-0.05	-1.252	0
TW2-F50	0.4347		-0.02	-0.7068		-0.05	-1.767	
TW3	0.7031	0.1327	-0.02	-0.6893	0	-0.05	-1.723	0
TW3-F50	0.2419		-0.02	-0.4987		-0.05	-1.247	
TW4	0.7742	0.148	-0.02	-0.5778	0	-0.05	-1.444	0
TW4-F25	0.2429		0.1	-0.1959		-0.05	-0.9796	
TW5	0.8492	0.1608	-0.02	-0.2289	0	-0.05	-0.5722	0
TW6	0.8728	0.1679	-0.02	-0.2276	0	-0.05	-0.569	0
TW7	1.412		-0.02	-0.8308		-0.05	-2.077	
TW7-F50	0.8192	0.1556	-0.02	-0.4334	0	-0.05	-1.084	0
TW8	1.653	0.314	-0.02	-0.2161	0	-0.05	-0.5402	0
TW9	2.486	0.4693	-0.02	-0.5962	0	-0.05	-1.49	0
TW9-F50	1.614		-0.02	-0.3798		-0.05	-0.9494	
TW10	1.858	0.3574	-0.02	-0.5789	0	-0.05	-1.447	0
TW10-F50	1.26	0.2423	-0.02	-0.3646	0	-0.05	-0.9115	0
TW11	2.119	0.4113	0.024	0.2841	4.411	0.011	0.1302	0.5778
TW12	2.058	0.3957	0.004	0.0584	0.898	0.001	0.0146	0.0641
TW13								
TW14	1.329	0.2572	0.108	0.4769	7.383	0.016	0.0707	0.3125
TW15	1.176	0.2303	0.018	0.1223	1.917	0.01	0.068	0.3043
TW16	3.868	0.7578	0.021	0.1469	2.302	0.001	0.007	0.0313
BKGND	0		0	0	0	0	0	
f(i)	0.0164		0.0002	0.0002		0.0007	0.0007	

TEST NUMBER	RXN TIME	pH	DIL FACTOR	SA/V	LEACHANT VOL	ACL NUMBER	Al, ACL	Al, ppm	NL(Al), g/m2	B, ACL
TW31	14	9.38	12.646	325	2.02	91-0532-04	0.11	1.3911	0.05018	0.61
TW32	14	9.23	17.085	318	2.07	91-0532-05	0.11	1.8794	0.12693	0.11
TW32-F50	14		36.116		2.07	91-0532-06	-0.1	-3.6116		0.05
TW33	28	9.44	26.15	319	2.07	91-0533-05	0.1	2.615	0.24013	0.13
TW33-F50	28		25.306		2.07	91-0533-06	-0.1	-2.5306		0.14
TW34	28	9.45	36.023	323	2.04	91-0533-07	-0.1	-3.6023	-0.71106	0.08
TW34-F25	28		19.305		2.04	91-0533-08	-0.1	-1.9305		0.14
TW35	56	9.66	12.308	322	2.05	91-0596-03	0.3	3.6925	0.40273	0.64
TW36	56	9.71	10.895	323	2.03	91-0596-04	0.37	4.031	0.45312	0.8
TW37	91	9.74	28.273	325	2.06	91-0639-04	0.11	3.11	0.31072	0.38
TW37-F50	91		18.694		2.06	91-0639-05	-0.1	-1.8694		0.59
TW38	91	9.63	11.549	318	2.05	91-0639-06	0.44	5.0817	0.623	0.94
TW39	180	9.88	125.92	323	2.04	92-0134-11	-0.1	-12.592	-2.082	0.16
TW39-F50	180		26.879		2.04	92-0134-12	-0.1	-2.6879		0.85
TW40	180	9.69	38.4	322	2.05	92-0134-13	-0.1	-3.84	-0.74963	0.26
TW40-F50	180		23.254		2.05	92-0134-14	-0.1	-2.3254		0.43
TW41	360	9.79	10.715	325	2.03	PACKET.1	0.275	2.948	0.36175	1.3
TW42	360	9.79	11.059	325	2.03	PACKET.1	0.384	4.247	0.5588	1.14
TW43	540	9.69	4.0237	318	2.05	PACKET.1	0.908	3.65	0.47921	4.06
TW44	540	9.78	3.9942	327	2.03	PACKET.1	0.385	1.536	0.1473	4.6
TW45	720	9.24	7.0185	327	2.05	PACKET.1	0.161	1.13	0.085866	1.83
TW46	720	9.42	7.1069	325	2.03	PACKET.1	0.144	1.023	0.070238	2.64
BKGND			1				0.56	0.56		0.15
f(i)			1				0.0203	0.0203		0.0248

B, ppm	NL(B), g/m2	Ba, ACL	Ba, ppm	NL(Ba), g/m2	Ca, ACL	Ca, ppm	NL(Ca), g/m2	Cr, ACL	Cr, ppm	NL(Cr), g/m2
7.7141	0.93847	-0.02	-0.25292	-0.38911	0.02	0.25292	-1.5088	-0.02	-0.25292	-0.15564
1.8794	0.21928	-0.02	-0.3417	-0.53727	0.08	1.3668	-1.1347	-0.02	-0.3417	-0.21491
1.8058		-0.02	-0.72233		0.03	1.0835		-0.02	-0.72233	
3.3995	0.41075	-0.02	-0.523	-0.81975	0.04	1.046	-1.2481	-0.02	-0.523	-0.3279
3.5429		-0.02	-0.50613		0.02	0.50613		-0.02	-0.50613	
2.8819	0.34104	-0.02	-0.72047	-1.1153	0.04	1.4409	-1.0905	-0.02	-0.72047	-0.44611
2.7027		-0.02	-0.3861		-0.02	-0.3861		-0.02	-0.3861	
7.8773	0.96766	-0.02	-0.24617	-0.38225	0.09	1.1077	-1.2142	-0.02	-0.24617	-0.1529
8.7158	1.0693	-0.02	-0.21789	-0.3373	0.11	1.1984	-1.1778	-0.02	-0.21789	-0.13492
10.744	1.3143	-0.02	-0.56545	-0.86993	0.03	0.84818	-1.2958	-0.02	-0.56545	-0.34797
11.03		-0.02	-0.37388		0.03	0.56082		-0.02	-0.37388	
10.856	1.3576	-0.02	-0.23099	-0.36319	0.12	1.3859	-1.1277	-0.02	-0.23099	-0.14527
20.147	2.4963	-0.02	-2.5183	-3.8983	-0.02	-2.5183	-2.5158	-0.02	-2.5183	-1.5593
22.847		-0.02	-0.53759		0.05	1.344		-0.02	-0.53759	
9.984	1.2315	-0.02	-0.768	-1.1925	-0.02	-0.768	-1.8915	-0.02	-0.768	-0.47702
9.9991		-0.02	-0.46507		0.08	1.8603		-0.02	-0.46507	
13.929	1.7096	0.008	0.08572	0.13188	0.202	2.1644	-0.86067	0.009	0.096435	0.059345
12.607	1.5456	0.017	0.188	0.28924	0.245	2.7095	-0.66567	0.011	0.12165	0.074861
16.336	2.0524	0.032	0.12876	0.20245	0.171	0.68805	-1.4195	0.034	0.13681	0.086041
18.373	2.2471	0.022	0.087872	0.13436	0.639	2.5523	-0.71748	0.047	0.18773	0.11482
12.844	1.5653	0.003	0.021055	0.032195	0.246	1.7266	-1.011	0.008	0.056148	0.034341
18.762	2.3092	0.005	0.035535	0.054668	0.322	2.2884	-0.81631	0.011	0.078176	0.048108
0.15		0	0		4.57	4.57		0	0	
0.0248		0.002	0.002		0.0086	0.0086		0.005	0.005	

Cu, ACL	Cu, ppm	NL(Cu), g/m2	Fe, ACL	Fe, ppm	NL(Fe), g/m2	K, ACL	K, ppm	NL(K), g/m2	Li, ACL	Li, ppm
-0.02	-0.25292	0	0.08	1.0117	0.038623	1.3	16.44	0.87825	0.62	7.8405
-0.02	-0.3417	0	0.14	2.3919	0.093863	0.75	12.814	0.52857	0.24	4.1004
-0.02	-0.72233		-0.02	-0.72233		0.47	16.975		0.1	3.6116
-0.02	-0.523	0	0.18	4.707	0.18451	0.56	14.644	0.71258	0.22	5.753
-0.02	-0.50613		-0.02	-0.50613		0.57	14.425		0.22	5.5674
-0.02	-0.72047	0	0.09	3.2421	0.12539	0.43	15.49	0.78852	0.13	4.683
-0.02	-0.3861		-0.02	-0.3861		0.75	14.479		0.21	4.054
0.03	0.36925	0.35836	0.73	8.9851	0.34928	1.2	14.77	0.7186	0.75	9.2312
0.04	0.43579	0.42162	0.88	9.5873	0.37157	1.5	16.342	0.87389	0.9	9.8052
-0.02	-0.56545	0	0.22	6.22	0.23944	0.59	16.681	0.90225	0.41	11.592
-0.02	-0.37388		0.02	0.37388		1.11	20.75		0.61	11.403
0.04	0.46197	0.45398	0.91	10.51	0.41376	1.84	21.251	1.3872	1.05	12.127
-0.02	-2.5183	0	-0.02	-2.5183	0	-0.3	-37.775	0	0.16	20.147
-0.02	-0.53759		-0.02	-0.53759		1.1	29.567		0.79	21.235
-0.02	-0.768	0	0.1	3.84	0.14905	0.4	15.36	0.77791	0.31	11.904
-0.02	-0.46507		-0.02	-0.46507		0.84	19.533		0.5	11.627
0.019	0.20358	0.19575	0.056	0.60004	0.023136	1.81	19.394	1.2302	1.55	16.608
0.074	0.81837	0.78689	0.484	5.3526	0.20638	1.62	17.916	1.083	1.38	15.261
0.143	0.57539	0.56544	1.57	6.3172	0.24894	5.35	21.527	1.4743	4.67	18.791
0.007	0.027959	0.02672	0.276	1.1024	0.042246	5.54	22.128	1.4932	4.96	19.811
0.005	0.035092	0.033536	0.044	0.30881	0.011834	2.58	18.108	1.0953	1.94	13.616
0.006	0.042641	0.041001	0.082	0.58277	0.02247	3.17	22.529	1.5423	2.58	18.336
0	0		0	0		7.04	7.04		0.05	0.05
0.0032	0.0032		0.0798	0.0798		0.0309	0.0309		0.0197	0.0173

NL(Li), g/m2	Mg, ACL	Mg, ppm	NL(Mg), g/m2	Mn, ACL	Mn, ppm	NL(Mn), g/m2	Na, ACL	Na, ppm	NL(Na), g/m2	Ni, ACL
1.2168	-0.01	-0.12646	0	0.01	0.12646	0.02779	6.32	79.923	1.1601	-0.03
0.64656	0.03	0.51255	0.059966	0.03	0.51255	0.11513	3.77	64.411	0.44771	-0.03
	-0.01	-0.36116		-0.01	-0.36116		1.79	64.648		-0.03
0.9075	0.03	0.7845	0.16634	0.03	0.7845	0.17566	2.69	70.343	0.72767	-0.03
	-0.01	-0.25306		-0.01	-0.25306		2.8	70.858		-0.03
0.72811	0.02	0.72047	0.1395	0.02	0.72047	0.15932	2.01	72.407	0.8153	-0.03
	-0.01	-0.19305	0	-0.01	-0.19305		3.59	69.305		-0.03
1.4474	0.1	1.2308	0.33806	0.13	1.6001	0.35494	6.46	79.512	1.1516	-0.03
1.5331	0.12	1.3074	0.36663	0.16	1.7432	0.38548	7.12	77.57	1.0571	0.04
1.8027	0.03	0.84818	0.18776	0.04	1.1309	0.24855	3.08	87.08	1.4933	-0.03
	-0.01	-0.18694		-0.01	-0.18694		4.81	89.919		-0.03
1.9278	0.15	1.7324	0.53946	0.17	1.9634	0.44101	8.06	93.087	1.812	0.03
3.1583	-0.01	-1.2592	0	-0.01	-1.2592	0	0.95	119.62	3.0267	-0.03
	-0.01	-0.26879		-0.01	-0.26879		5.1	137.08		-0.03
1.8687	0.02	0.768	0.15839	0.02	0.768	0.17036	2.25	86.4	1.4753	-0.03
	0.02	0.46507		-0.01	-0.23254		3.99	92.782		-0.03
2.5862	0.031	0.33217	0.12776	0.057	0.61076	0.13423	9.24	99.007	2.2207	0.02
2.3758	0.141	1.5593	0.59974	0.177	1.9574	0.43021	8.52	94.223	1.998	0.068
2.9916	0.318	1.2795	0.50296	0.383	1.5411	0.34615	26.2	105.42	2.5748	0.129
3.0676	0.063	0.25163	0.096191	0.124	0.49528	0.10819	26.5	105.85	2.5236	0.041
2.1059	0.008	0.056148	0.021463	0.013	0.091241	0.01993	12.2	85.626	1.5881	0.006
2.8561	0.005	0.035535	0.013667	0.0169	0.11371	0.024991	14.9	105.89	2.5413	0.009
	0	0		0	0		51.3	51.3		0
	0.008	0.008		0.014	0.014		0.0661	0.0661		0.0065

Ni, ppm	NL(Ni), g/m2	Si, ACL	Si, ppm	NL(Si), g/m2	Th, ACL	Th, ppm	NL(Th), g/m2	Ti, ACL	Ti, ppm	NL(Ti), g/m2
-0.37938	0	5.01	63.356	0.78943	-0.05	-0.6323	0	-0.02	-0.25292	0
-0.51255	0	3.26	55.697	0.7015	-0.05	-0.85426	0	-0.02	-0.3417	0
-1.0835		1.51	54.536		-0.05	-1.8058		-0.02	-0.72233	
-0.7845	0	2.5	65.375	0.83195	-0.05	-1.3075	0	-0.02	-0.523	0
-0.75919		2.41	60.989		-0.05	-1.2653		-0.02	-0.50613	
-1.0807	0	1.82	65.562	0.82418	-0.05	-1.8012	0	-0.02	-0.72047	0
-0.57915		2.82	54.44		-0.05	-0.96524		-0.02	-0.3861	
-0.36925	0	7.68	94.528	1.2201	-0.05	-0.61542	0	0.03	0.36925	0.21236
0.43579	0.20757	9.09	99.033	1.2773	-0.05	-0.54474	0	0.04	0.43579	0.24985
-0.84818	0	3.72	105.17	1.352	-0.05	-1.4136	0	-0.02	-0.56545	0
-0.56082		5.24	97.957		-0.05	-0.9347		-0.02	-0.37388	
0.34648	0.16762	10.1	116.65	1.5396	-0.05	-0.57746	0	0.05	0.57746	0.33628
-3.7775	0	1.01	127.18	1.6583	-0.05	-6.2958	0	-0.02	-2.5183	0
-0.80638		4.94	132.78		-0.05	-1.344		-0.02	-0.53759	
-1.152	0	2.74	105.22	1.3652	-0.05	-1.92	0	-0.02	-0.768	0
-0.69761		4.22	98.131		-0.05	-1.1627		-0.02	-0.46507	
0.2143	0.10144	11	117.86	0.97763	0.025	0.26788	0.35836	0.021	0.22501	0.12821
0.75201	0.35598	11	121.65	1.0285	0.04	0.44236	0.58179	0.056	0.6193	0.35288
0.51906	0.25112	35.4	142.44	1.337	0.12	0.48284	0.66016	0.114	0.4587	0.26712
0.16376	0.077046	35	139.8	1.2649	0.131	0.52324	0.69571	0.059	0.23566	0.13346
0.042111	0.019812	11.3	79.309	0.4561	0.021	0.14739	0.19597	0.01	0.070185	0.039747
0.063962	0.030278	1.58	11.229	-0.45705	0.05	0.35535	0.47538	0.017	0.12082	0.068842
0		45.2	45.2		0	0		0	0	
0.0065		0.2287	0.2287		0.0023	0.0023		0.0054	0.0054	

U, ACL	U, ppm	NL(U), g/m2	Zn, ACL	Zn, ppm	NL(Zn), g/m2	Zr, ACL	Zr, ppm	NL(Zr), g/m2
0.142	1.7957	0.15303	-0.02	-0.25292	-3.8911	-0.05	-0.6323	0
0.0508	0.86792	0.09613	-0.02	-0.3417	-5.3727	-0.05	-0.85426	0
0.0168	0.60675		-0.02	-0.72233	-11.357	-0.05	-1.8058	
0.0591	1.5455	0.10197	-0.02	-0.523	-8.1975	-0.05	-1.3075	0
0.0439	1.111		-0.02	-0.50613	-7.9331	-0.05	-1.2653	
0.0284	1.0231	0.18225	-0.02	-0.72047	-11.153	-0.05	-1.8012	0
0.0332	0.64092		0.05	-0.3861	-5.9767	-0.05	-0.96524	
0.134	1.6493	0.31232	-0.02	-0.24617	-3.8225	-0.05	-0.61542	0
0.197	2.1463	0.40517	-0.02	-0.21789	-3.373	-0.05	-0.54474	0
0.0792	2.2392	0.42011	-0.02	-0.56545	-8.6993	-0.05	-1.4136	0
0.0806	1.5067		-0.02	-0.37388	-5.752	-0.05	-0.9347	
0.249	2.8758	0.55142	-0.02	-0.23099	-3.6319	-0.05	-0.57746	0
0.0125	1.574	0.29713	-0.02	-2.5183	-38.983	-0.05	-6.2958	0
0.0525	1.4112		-0.02	-0.53759	-8.3218	-0.05	-1.344	
0.0483	1.8547	0.35122	-0.02	-0.768	-11.925	-0.05	-1.92	0
0.0489	1.1371		-0.02	-0.46507	-7.2216	-0.05	-1.1627	
0.133	1.4251	0.26737	0.004	0.04286	0.65938	0.003	0.032145	0.1413
0.219	2.4219	0.45439	0.016	0.17694	2.7222	0.009	0.099531	0.4375
0.593	2.3861	0.45752	0.064	0.25752	4.049	0.022	0.088521	0.39767
0.518	2.069	0.38581	0.154	0.61511	9.4053	0.019	0.07589	0.33154
0.117	0.82116	0.15312	0.016	0.1123	1.7171	0.002	0.014037	0.061324
0.217	1.5422	0.28934	0.02	0.14214	2.1867	0.06	0.42641	1.8743
0	0		0	0	0	0	0	
0.0164	0.0164		0.0002	0.0002	0.0002		0.0007	

TEST NUMBER	RXN TIME	pH	DIL FACTOR	SA/V, /m	LEACHANT VO	ACL NUMBER	A1, ACL	A1, ppm	NL(A1),g/m2	B, ACL	B, ppm
TY1	3	9.96	3.004	1882	10.09	91-0532-07	1.64	4.9266	0.12895	4.8	14.419
TY1-F50			3.5765			91-0532-08	1.26	4.5064		3.98	14.234
TY2	3	9.93	3.7059	1963	10.03	91-0532-09	1.32	4.8918	0.12276	3.75	13.897
TY2-F50			3.6862			91-0532-10	1.24	4.5709		3.89	14.339
TY3	7	9.99	2.4288	1889	10.05	92-0353-09	2.01	4.8819	0.12731	7.42	18.022
TY3-F50			10.449			92-0353-10	0.37	3.8661		1.76	18.39
TY4	7	10.07	2.1543	1908	10.04	92-0353-11	2.33	5.0195	0.12959	8.53	18.376
TY4-F50			11.75			92-0353-12	0.34	3.995		1.59	18.683
TY5	14	10.02	2.2371	1925	10.05	92-0353-13	2	4.4742	0.1145	9.93	22.214
TY5-F50			12.019			92-0353-14	0.22	2.8422		1.85	23.9
TY6	14	9.97	2.493	1953	10.01	92-0353-15	1.75	4.3628	0.11004	9.05	22.562
TY6-F50			11.245			92-0353-16	0.26	2.9237		2.04	22.94
TY7	30	10.24	2.4316	1975	10.01	92-0278-01	1.7	4.1337	0.1031	10.2	24.802
TY7-F50			11.231			92-0278-02	0.3	3.3693		2.22	24.933
TY8	30	10.12	2.1464	1955	10.01	92-0278-03	1.91	4.0996	0.1033	11.8	25.328
TY8-F50			11.324			92-0278-04	0.31	3.5104		2.19	24.8
TY9	70	10.32	10.403	1947	10.02	92-0278-05	0.43	4.4733	0.11318	2.79	29.024
TY9-F50			11.288			92-0278-06	0.25	2.822		2.36	26.64
TY10	70	10.33	8.7297	1951	9.99	92-0278-07	0.51	4.4521	0.11241	3.37	29.419
TY10-F50			11.645			92-0278-08	0.22	2.5619		2.34	27.249
TY11	140	10.17	2.2804	2005	9.99	92-0087-01	1.73	3.9451	0.096927	14.3	32.61
TY11-F50			24.015			92-0087-02	-0.1	-2.4015		1.26	30.259
TY12	140	10.28	2.3214	1975	10.04	92-0087-03	1.79	4.1553	0.10364	14.4	33.428
TY12-F50			17.25			92-0087-04	-0.1	-1.725		1.7	29.325
TY13	280	10.39	10.042	2013	10.04	92-0278-09	0.41	4.1172	0.10075	3.7	37.155
TY13-F50			11.211			92-0278-10	0.19	2.1301		3.43	38.454
TY14	280	10.31	9.2011	2021	10.03	92-0278-11	0.47	4.3245	0.10541	4.29	39.473
TY14-F50			10.51			92-0278-12	0.19	1.9969		3.6	37.836
TY15	560	10.11	1.896	2017	10.05	PACKET.1	1.05	1.9908	0.048621	19.4	36.782
TY15-F50			2.488			PACKET.1	0.662	1.6471		14.5	36.076
TY16	560	10.29	1.78	2000	10.07	PACKET.1	1.36	2.4208	0.059626	20.8	37.024
TY16-F50			2.063			PACKET.1	0.6	1.2378		13.2	27.232
TY17	980	9.95	2.723	2008	10.07	PACKET.2	0.687	1.8707	0.045893	17.09	46.536
TY17-F25			5.341			PACKET.2	0.132	0.70501		8.375	44.731
TY18	980	9.84	2.736	1946	10.07	PACKET.2	0.556	1.5212	0.038508	14.57	39.864
TY18-F25			5.342			PACKET.2	0.499	2.6657		9	48.078
f(i)								0.0203		0.0248	
BKGND								0		0	

TEST NO.	NL(B), g/m2	Ba, ACL	Ba, ppm	NL(Ba), g/m	Ca, ACL	Ca, ppm	NL(Ca), g/m	Cr, ACL	Cr, ppm	NL(Cr), g/m	Cu, ACL
TY1	0.30894	-0.02	-0.06008	-0.015962	-0.02	-0.06008	-0.003712	-0.02	-0.06008	-0.063847	0.02
TY1-F50		-0.02	-0.07153	-0.019004	0.13	0.46494		-0.02	-0.07153		-0.02
TY2	0.28547	-0.02	-0.074118	-0.018879	-0.02	-0.074118	-0.0043904	-0.02	-0.074118	-0.075515	-0.02
TY2-F50		-0.02	-0.073724	-0.018778	0.16	0.58979		-0.02	-0.073724		-0.02
TY3	0.38469	-0.02	-0.048576	-0.012858	0.27	0.65578	0.040367	-0.02	-0.048576	-0.05143	0.04
TY3-F50		-0.02	-0.20898		0.25	2.6123		-0.02	-0.20898		-0.02
TY4	0.38835	-0.02	-0.043086	-0.011291	0.05	0.10772	0.0065645	-0.02	-0.043086	-0.045164	0.04
TY4-F50		-0.02	-0.235		0.18	2.115		-0.02	-0.235		-0.02
TY5	0.46532	-0.02	-0.044742	-0.011621	0.19	0.42505	0.025675	0.02	0.044742	0.046485	0.07
TY5-F50		-0.02	-0.25838		0.28	3.6173		-0.02	-0.25838		-0.02
TY6	0.46582	-0.02	-0.04986	-0.012765	0.37	0.92241	0.054919	-0.02	-0.04986	-0.05106	0.05
TY6-F50		-0.02	-0.2249		0.38	4.2731		-0.02	-0.2249		-0.02
TY7	0.50638	-0.02	-0.048632	-0.012312	0.44	1.0699	0.062991	0.03	0.072948	0.073871	0.08
TY7-F50		-0.02	-0.22462		0.51	5.7278		-0.02	-0.22462		-0.02
TY8	0.52239	-0.02	-0.042928	-0.010979	0.21	0.45074	0.026809	0.03	0.064392	0.065874	0.08
TY8-F50		-0.02	-0.22648		0.59	6.6812		-0.02	-0.22648		-0.02
TY9	0.6011	-0.02	-0.20806	-0.053431	0.24	2.4967	0.14911	-0.02	-0.20806	-0.21372	0.03
TY9-F50		-0.02	-0.22576		0.93	10.498		-0.02	-0.22576		0.02
TY10	0.60802	-0.02	-0.17459	-0.044745	0.07	0.61108	0.03642	0.02	0.17459	0.17898	0.03
TY10-F50		-0.02	-0.2329		0.78	9.0831		-0.02	-0.2329		-0.02
TY11	0.65581	-0.02	-0.045608	-0.011374	0.15	0.34206	0.019838	0.04	0.091216	0.090989	0.12
TY11-F50		-0.02	-0.4803	-0.11978	0.02	0.4803		-0.02	-0.4803		-0.02
TY12	0.68249	-0.02	-0.046428	-0.011754	0.15	0.34821	0.020501	0.06	0.13928	0.14105	0.13
TY12-F50		-0.02	-0.345	-0.087342	0.04	0.69		-0.02	-0.345		-0.02
TY13	0.74426	-0.02	-0.20084	-0.049886	0.56	5.6235	0.32484	-0.02	-0.20084	-0.19954	0.04
TY13-F50		-0.02	-0.22422		0.7	7.8477		-0.02	-0.22422		-0.02
TY14	0.78755	-0.02	-0.18402	-0.045527	0.63	5.7967	0.33352	-0.02	-0.18402	-0.18211	0.04
TY14-F50		-0.02	-0.2102		0.57	5.9907		-0.02	-0.2102		-0.02
TY15	0.73533	0.025	0.0474	0.01175	2.127	4.0328	0.23249	0.142	0.26923	0.26696	0.154
TY15-F50		0.005	0.01244		1.63	4.0554		0.089	0.22143		0.021
TY16	0.74645	0.033	0.05874	0.014685	2.12	3.7736	0.2194	0.138	0.24564	0.24564	0.229
TY16-F50		0.006	0.012378		1.5	3.0945		0.077	0.15885		0.038
TY17	0.93449	0.0104	0.028319	0.0070516	0.817	2.2247	0.12883	0.0458	0.12471	0.12422	0.0704
TY17-F25		0.00174	0.0092933		0.624	3.3328		0.0298	0.15916		0.00746
TY18	0.826	0.00923	0.025253	0.0064885	0.762	2.0848	0.12457	0.045	0.12312	0.12654	0.0588
TY18-F25		0.00163	0.0087075		0.584	3.1197		0.0284	0.15171		0.00277
F(i)			0.002			0.0086			0.0005		
BKGND			0			0			0		

TEST NO.	Cu, ppm	NL(Cu), g/m	Fe, ACL	Fe, ppm	NL(Fe), g/m	K, ACL	K, ppm	NL(K), g/m2	Li, ACL	Li, ppm	NL(Li), g/m
TY1	0.06008	0.0099761	0.67	2.0127	0.013401	5.11	15.35	0.26396	4.5	13.518	0.36461
TY1-F50	-0.07153	-0.011877	0.08	0.28612		4.22	15.093		3.74	13.376	
TY2	-0.074118	-0.011799	0.53	1.9641	0.012539	3.98	14.749	0.24316	3.61	13.378	0.34595
TY2-F50	-0.073724	-0.011737	0.08	0.2949		4.07	15.003		3.6	13.27	
TY3	0.097152	0.016072	1.18	2.866	0.019012	6.63	16.103	0.27588	6.01	14.597	0.39225
TY3-F50	-0.20898	0.07	0.07	0.73143		1.41	14.733		1.37	14.315	
TY4	0.086172	0.014114	1.36	2.9298	0.019243	7.55	16.265	0.27588	7.36	15.856	0.42183
TY4-F50	-0.235		0.02	0.235		1.23	14.453		1.23	14.453	
TY5	0.1566	0.025422	1.98	4.4295	0.028835	8.66	19.373	0.3257	8.5	19.015	0.50143
TY5-F50	-0.25838		-0.02	-0.25838		1.53	19.766		1.44	18.603	
TY6	0.12465	0.019945	1.81	4.5123	0.028953	7.72	19.246	0.31892	7.67	19.121	0.49699
TY6-F50	-0.2249		-0.02	-0.2249		1.8	20.241		1.56	17.542	
TY7	0.19453	0.03078	2.18	5.3009	0.033634	9.26	22.517	0.36896	9.33	22.687	0.5831
TY7-F50	-0.22462		-0.02	-0.22462		1.87	21.002		1.88	21.114	
TY8	0.17171	0.027448	2.4	5.1514	0.03302	10.5	22.537	0.37307	10.4	22.323	0.5796
TY8-F50	-0.22648		0.02	0.22648		1.81	20.496		1.92	21.742	
TY9	0.31209	0.050091	0.66	6.866	0.044191	2.32	24.135	0.40116	2.41	25.071	0.65365
TY9-F50	0.22576		0.02	0.22576		1.96	22.124		2.02	22.802	
TY10	0.26189	0.041948	0.82	7.1584	0.045978	2.78	24.269	0.40256	2.96	25.84	0.67231
TY10-F50	-0.2329		-0.02	-0.2329		1.91	22.242		1.98	23.057	
TY11	0.27365	0.042651	3.97	9.0532	0.056583	13.5	30.785	0.4969	14.1	32.154	0.81405
TY11-F50	-0.4803	-0.07486	-0.02	-0.4803		1.25	30.019		1.19	28.578	
TY12	0.30178	0.04775	3.99	9.2624	0.05877	13.1	30.41	0.49831	13.8	32.035	0.82337
TY12-F50	-0.345	-0.054589	-0.02	-0.345		1.65	28.462		1.64	28.29	
TY13	0.40168	0.062357	1.02	10.243	0.063764	3.16	31.733	0.51016	3.48	34.946	0.88123
TY13-F50	-0.22422		0.02	0.22422		2.67	29.933		3.09	34.642	
TY14	0.36804	0.056909	1.18	10.857	0.067321	3.58	32.94	0.52747	4.19	38.553	0.96833
TY14-F50	-0.2102		-0.02	-0.2102		2.86	30.059		3.28	34.473	
TY15	0.29198	0.045238	1.37	2.5975	0.016138	16.2	30.715	0.49282	20.4	38.678	0.97341
TY15-F50	0.052248		0	0		11.7	29.11		15.3	38.066	
TY16	0.40762	0.063691	2.28	4.0584	0.025429	17.8	31.684	0.51269	22.1	39.338	0.99843
TY16-F50	0.078394		0	0		10.8	22.28		13.6	28.057	
TY17	0.1917	0.029834	1.973	5.3725	0.033528	13.47	36.679	0.59114	18.4	50.103	1.2666
TY17-F25	0.039844		0.00697	0.037227		6.898	36.842		8.432	45.035	
TY18	0.16088	0.025835	1.345	3.6799	0.023697	10.87	29.74	0.49459	14.49	39.645	1.0341
TY18-F25	0.014797		0	0		7.182	38.366		8.924	47.672	
F(1)	0.0032			0.0798			0.0309			0.0197	
BKGND	0			0			0			0	

TEST NO.	Mg, ACL	Mg, ppm	NL(Mg), g/m	Mn, ACL	Mn, ppm	NL(Mn), g/m	Na, ACL	Na, ppm	NL(Na), g/m	Ni, ACL	Ni, ppm
TY1	0.1	0.3004	0.019952	0.07	0.21028	0.0079809	14.9	44.76	0.3598	-0.03	-0.09012
TY1-F50	0.03	0.10729		-0.01	-0.035765		13.2	47.21		-0.03	-0.10729
TY2	0.08	0.29647	0.018879	0.06	0.22235	0.0080909	11.9	44.1	0.33987	-0.03	-0.11118
TY2-F50	0.04	0.14745		-0.01	-0.036862		13.1	48.289		-0.03	-0.11059
TY3	0.15	0.36432	0.024108	0.14	0.34003	0.012858	21.2	51.491	0.41238	-0.03	-0.072864
TY3-F50	0.01	0.10449		-0.01	-0.10449		4.92	51.409		-0.03	-0.31347
TY4	0.17	0.36623	0.023993	0.16	0.34469	0.012904	25.4	54.719	0.43387	0.04	0.086172
TY4-F50	-0.01	-0.1175		-0.01	-0.1175		4.42	51.935		-0.03	-0.3525
TY5	0.22	0.49216	0.031959	0.26	0.58165	0.021582	29.6	66.218	0.52041	0.05	0.11185
TY5-F50	-0.01	-0.12919		-0.01	-0.12919		5.4	69.763		-0.03	-0.38757
TY6	0.2	0.4986	0.031912	0.23	0.57339	0.020971	26.9	67.062	0.51948	-0.03	-0.07479
TY6-F50	0.03	0.33735		-0.01	-0.11245		6.16	69.269		-0.03	-0.33735
TY7	0.2	0.48632	0.03078	0.29	0.70516	0.025503	32.8	79.756	0.61094	0.06	0.1459
TY7-F50	-0.01	-0.11231		-0.01	-0.11231		6.98	78.392		-0.03	-0.33693
TY8	0.22	0.47221	0.030192	0.32	0.68685	0.025095	36.8	78.988	0.61124	0.06	0.12878
TY8-F50	-0.01	-0.11324		-0.01	-0.11324		6.82	77.23		-0.03	-0.33972
TY9	0.06	0.62418	0.040073	0.09	0.93627	0.034348	8.3	86.345	0.67092	0.04	0.41612
TY9-F50	0.02	0.22576		-0.01	-0.11288		7.46	84.208		-0.03	-0.33864
TY10	0.07	0.61108	0.039152	0.11	0.96027	0.035157	10.3	89.916	0.69723	0.03	0.26189
TY10-F50	0.01	0.11645		-0.01	-0.11645		7.36	85.707		-0.03	-0.34935
TY11	0.35	0.79814	0.049759	0.59	1.3454	0.047931	50	114.02	0.86033	0.06	0.13682
TY11-F50	-0.01	-0.24015		-0.01	-0.24015		4.39	105.43		-0.03	-0.72045
TY12	0.35	0.81249	0.051423	0.59	1.3696	0.049534	48.8	113.28	0.86776	0.1	0.23214
TY12-F50	-0.01	-0.1725		-0.01	-0.1725		6.05	104.36		-0.03	-0.5175
TY13	0.09	0.90378	0.056121	0.15	1.5063	0.053449	11.9	119.5	0.89809	0.04	0.40168
TY13-F50	0.02	0.22422		-0.01	-0.11211		11.1	124.44		-0.03	-0.33633
TY14	0.1	0.92011	0.056909	0.17	1.5642	0.055283	13.7	126.06	0.94361	0.04	0.36804
TY14-F50	-0.01	-0.1051		-0.01	-0.1051		11.8	124.02		-0.03	-0.3153
TY15	0.221	0.41902	0.025968	0.508	0.96317	0.034109	70.6	133.86	1.004	0.119	0.22562
TY15-F50	0.081	0.20153		0.006	0.014928		53.8	133.85		0	0
TY16	0.309	0.55002	0.034376	0.783	1.3937	0.049776	77	137.06	1.0368	0.163	0.29014
TY16-F50	0.13	0.26819		0.006	0.012378		49	101.09		0	0
TY17	0.154	0.41934	0.026104	0.344	0.93671	0.033321	60.15	163.79	1.234	0.0815	0.22192
TY17-F25	0.0735	0.39256		0.0026	0.013887		30.21	161.35		0.00556	0.035037
TY18	0.162	0.44323	0.028471	0.242	0.66211	0.024303	48.53	132.78	1.0322	0.0629	0.17209
TY18-F25	0.0222	0.11859		0.00085	0.0045407		13.38	71.476		0.00114	0.0060899
F(1)		0.008			0.014			0.0661			0.0065
BKGND		0			0			0			0

TEST NO.	NL(Ni), g/m	Si, ACL	Si, ppm	NL(Si), g/m	Th, ACL	Th, ppm	NL(Th), g/m	Ti, ACL	Ti, ppm	NL(Ti), g/m	U, ACL
TY1	-0.007367	31.2	93.725	0.21776	-0.05	-0.1502	-0.034699	0.02	0.06008	0.0059118	0.758
TY1-F50		25.7	91.916		-0.05	-0.17883		-0.02	-0.07153		0.616
TY2	-0.0087133	24.6	91.165	0.20307	-0.05	-0.1853	-0.041041	-0.02	-0.074118	-0.0069921	0.623
TY2-F50		24.9	91.786		-0.05	-0.18431		-0.02	-0.073724		0.601
TY3	-0.0059343	46.5	112.94	0.26142	-0.05	-0.12144	-0.027951	0.04	0.097152	0.0095241	0.648
TY3-F50		9.68	101.15		-0.05	-0.52245		-0.02	-0.20898		0.126
TY4	0.0069482	51.5	110.95	0.25425	-0.05	-0.10772	-0.024545	0.04	0.086172	0.0083636	0.0752
TY4-F50		9	105.75		-0.05	-0.5875		-0.02	-0.235		0.13
TY5	0.0089395	57.4	128.41	0.29168	-0.05	-0.11185	-0.025264	0.06	0.13423	0.012913	0.477
TY5-F50		10.2	131.77		-0.05	-0.64595		-0.02	-0.25838		0.0667
TY6	-0.0058915	53.4	133.13	0.29805	-0.05	-0.12465	-0.02775	0.05	0.12465	0.011819	0.435
TY6-F50		11.1	124.82		-0.05	-0.56225		-0.02	-0.2249		0.0744
TY7	0.011365	54.2	131.79	0.29178	-0.05	-0.12158	-0.026765	0.07	0.17021	0.01596	0.421
TY7-F50		11.7	131.4		-0.05	-0.56155		-0.02	-0.22462		0.0706
TY8	0.010134	64.7	138.87	0.3106	-0.05	-0.10732	-0.023867	0.08	0.17171	0.016265	0.48
TY8-F50		12	135.89		-0.05	-0.5662		-0.02	-0.22648		0.0691
TY9	0.032881	14.9	155	0.34811	-0.05	-0.52015	-0.11615	0.02	0.20806	0.019789	0.119
TY9-F50		12.1	136.58		-0.05	-0.5644		-0.02	-0.22576		0.0722
TY10	0.020651	18.3	159.75	0.35804	-0.05	-0.43649	-0.097271	0.03	0.26189	0.024858	0.147
TY10-F50		12	139.74		-0.05	-0.58225		-0.02	-0.2329		0.0729
TY11	0.010499	77.4	176.5	0.38492	0.05	0.11402	0.024725	0.13	0.29645	0.027381	0.759
TY11-F50		6.31	151.53		-0.05	-1.2007		-0.02	-0.4803		0.0531
TY12	0.018083	76	176.43	0.3906	0.05	0.11607	0.025552	0.14	0.325	0.030473	0.768
TY12-F50		8.52	146.97		-0.05	-0.8625		-0.02	-0.345		0.0751
TY13	0.030699	18.5	185.78	0.40354	-0.05	-0.5021	-0.10845	0.04	0.40168	0.036952	0.226
TY13-F50		16.3	182.74		-0.05	-0.56055		-0.02	-0.22422		0.157
TY14	0.028017	22	202.42	0.43796	-0.05	-0.46006	-0.098973	0.04	0.36804	0.033724	0.26
TY14-F50		17.5	183.93		-0.05	-0.5255		-0.02	-0.2102		0.159
TY15	0.017209	95.7	181.45	0.39335	0.047	0.089112	0.019209	0.139	0.26354	0.024197	1.59
TY15-F50		70.4	175.16		0.013	0.032344		0.007	0.017416		1.09
TY16	0.022318	103	183.34	0.40083	0.05	0.089	0.019348	0.192	0.34176	0.031644	1.72
TY16-F50		62.3	128.52		0.012	0.024756		0.008	0.016504		1.19
TY17	0.017003	74.9	203.95	0.44412	0.0279	0.075972	0.01645	0.147	0.40028	0.036915	1.493
TY17-F25		36.9	197.08		0.000648	0.003461		0.00394	0.021044		0.656
TY18	0.013605	72.7	198.91	0.44693	0.0252	0.068947	0.015404	0.109	0.29822	0.02838	1.348
TY18-F25		36.5	194.98		0.000829	0.0044285		0.00188	0.010043		0.642
F(i)			0.2287			0.0023			0.0054		
BKGND			0			0			0		

TEST NO.	U, ppm	NL(U), g/m2	Zn, ACL	Zn, ppm	NL(Zn), g/m	Zr, ACL	Zr, ppm	NL(Zr), g/m
TY1	2.277	0.073774	-0.02	-0.06008	-0.15962	-0.05	-0.1502	-0.11401
TY1-F50	2.2031		-0.02	-0.07153		-0.05	-0.17883	
TY2	2.3088	0.071716	-0.02	-0.074118	-0.18879	-0.05	-0.1853	-0.13485
TY2-F50	2.2154		-0.02	-0.073724		-0.05	-0.18431	
TY3	1.5739	0.050803	0.05	0.12144	0.32144	-0.05	-0.12144	-0.09184
TY3-F50	1.3166		0.05	0.52245		-0.05	-0.52245	
TY4	0.162	0.0051773	-0.02	-0.043086	-0.11291	-0.05	-0.10772	-0.080649
TY4-F50	1.5275		0.03	0.3525		-0.05	-0.5875	
TY5	1.0671	0.033801	0.04	0.089484	0.23243	-0.05	-0.11185	-0.083009
TY5-F50	0.8617		0.05	0.64595		-0.05	-0.64595	
TY6	1.0845	0.033858	0.06	0.14958	0.38295	-0.05	-0.12465	-0.091178
TY6-F50	0.83663		0.07	0.78715		-0.05	-0.56225	
TY7	1.0237	0.031606	0.08	0.19453	0.49248	-0.05	-0.12158	-0.087942
TY7-F50	0.79291		0.09	1.0108		-0.05	-0.56155	
TY8	1.0303	0.032134	0.03	0.064392	0.16469	-0.05	-0.10732	-0.078422
TY8-F50	0.78249		0.1	1.1324		-0.05	-0.5662	
TY9	1.238	0.03877	0.05	0.52015	1.3358	-0.05	-0.52015	-0.38165
TY9-F50	0.81499		0.16	1.8061		-0.05	-0.5644	
TY10	1.2833	0.040107	-0.02	-0.17459	-0.44745	-0.05	-0.43649	-0.31961
TY10-F50	0.84892		0.14	1.6303		-0.05	-0.58225	
TY11	1.7308	0.052637	0.03	0.068412	0.1706	-0.05	-0.11402	-0.08124
TY11-F50	1.2752		-0.03	-0.72045		-0.05	-1.2007	
TY12	1.7828	0.055043	0.03	0.069642	0.17631	-0.05	-0.11607	-0.083957
TY12-F50	1.2955		-0.02	-0.345		-0.05	-0.8625	
TY13	2.2695	0.068745	0.1	1.0042	2.4943	-0.05	-0.5021	-0.35633
TY13-F50	1.7601		0.12	1.3453		-0.05	-0.56055	
TY14	2.3923	0.072178	0.11	1.0121	2.504	-0.05	-0.46006	-0.3252
TY14-F50	1.6711		0.09	0.9459		-0.05	-0.5255	
TY15	3.0146	0.091135	0.034	0.064464	0.1598	0.011	0.020856	0.014772
TY15-F50	2.7119		0.042	0.1045		0.001	0.002488	
TY16	3.0616	0.093341	0.044	0.07832	0.1958	0.013	0.02314	0.016529
TY16-F50	2.455		0.1	0.2063		0.002	0.004126	
TY17	4.0654	0.12345	0.0169	0.046019	0.11459	0.00677	0.018435	0.013115
TY17-F25	3.5037		0.0136	0.072638		0.000311	0.0016611	
TY18	3.6881	0.11556	0.0284	0.077702	0.19965	0.00471	0.012887	0.0094601
TY18-F25	3.4296		0.0207	0.11058		0.000146	0.00077993	
F(i)	0.0164			0.0002			0.0007	
BKGND	0			0			0	

TEST NUMBER	RXN TIME	pH	DIL FACTOR	SA/V	LEACHANT VO	ACL NUMBER	Al, ACL	Al, ppm	NL(Al), g/m	B, ACL	B, ppm
TZ1	3	11.09	6.2984	19846	5.03	91-0532-11	1.16	7.3061	0.018135	12.3	77.47
TZ1-F50	3		11.989		5.03	91-0532-12	0.2	2.3978		6.23	74.692
TZ2	3	11.14	6.1735	19948	5.01	91-0532-13	1.25	7.7169	0.019057	12.4	76.551
TZ2-F50	3		11.51		5.01	91-0532-14	0.21	2.4172		6.06	69.753
TZ3	7	10.93	4.3036	20015	5.03	91-0532-15	2.64	11.362	0.027963	22.4	96.401
TZ4	7	11.21	6.4973	20015	5.03	91-0532-16	1.85	12.02	0.029584	14.9	96.81
TZ4-F50	7		10.278		4.98	91-0532-17	0.22	2.2611		9.41	96.714
TZ5-UF	14		38		4.98	91-0532-18	0.33	12.54		2.79	106.02
TZ5	14	11.34	4.657	20076	4.99	91-0532-19	3.39	15.787	0.038738	24.4	113.63
TZ6	14	11.33	3.8997	20029	4.99	91-0532-20	3.02	11.777	0.028966	22.6	88.133
TZ6-F50	14		24.535		4.99	91-0532-21	-0.1	-2.4535		4.47	109.67
TZ7	28	11.49	11.295	20121	4.99	91-0533-09	0.46	5.1955	0.01272	11.8	133.28
TZ7-F50	28		15.065		4.99	91-0533-10	0.14	2.1091		9.19	138.45
TZ8	28	11.46	24.547	20046	4.98	91-0533-11	0.13	3.1911	0.0078418	5.1	125.19
TZ8-F50	28		8.2356		4.98	91-0533-12	0.24	1.9765		16.7	137.53
TZ9	56	11.46	5.8051	20092	4.99	91-0569-05	0.28	1.6254	0.0039852	26.8	155.58
TZ10	56	11.5	6.4818	20061	4.99	91-0569-06	0.24	1.5556	0.00382	25.7	166.58
TZ11	91	11.43	14.514	20154	4.97	91-0639-07	-0.1	-1.4514	0	13.6	197.39
TZ11-F50	91		11.274		4.97	91-0639-08	0.1	1.1274		17.9	201.8
TZ12	91	11.5	13.712	20032	4.99	91-0639-09	-0.1	-0.13712	0	13.9	190.6
TZ12-F50	91		11.431		4.99	91-0639-10	-0.1	-1.1431		17.2	196.61
TZ13	182	11.62	8.9679	20143	4.98	92-0134-15	0.13	1.1658	0.0028511	31.9	286.08
TZ13-F50	182		17.086		4.98	92-0134-16	-0.1	-1.7086		15.1	258
TZ14	182	11.69	8.7876	20103	4.98	92-0134-17	0.13	1.1424	0.0027993	33	289.99
TZ14-F50	182		16.737		4.98	92-0134-18	-0.1	-1.6737		15.9	266.12
TZ15	364	11.72	36.605	20020	4.99	92-0388-01	-0.1	-3.6605	0	113	4136.4
TZ15-F50	364		9.6971			92-0388-02	0.1	0.96971		361	3500.7
TZ16	364	11.76	20.792	19985	5	92-0388-03	-0.1	-2.079	0	177	3679.8
TZ16-F50	364		9.1111			92-0388-04	-0.1	-0.9111		364	3316
F(i)							0.0203	0.0203		0.0248	0.0248
BKGND							0	0		0	0

TEST NO.	NL(B), g/m2	Ba, ACL	Ba, ppm	NL(Ba), g/m	Ca, ACL	Ca, ppm	NL(Ca), g/m	Cr, ACL	Cr, ppm	NL(Cr), g/m	Cu, ACL
TZ1	0.1574	0.02	0.12597	0.0031736	0.08	0.50387	0.0029522	0.03	0.18895	0.019042	0.25
TZ1-F50		-0.02	-0.23978		0.06	0.71935		-0.02	-0.23978		-0.02
TZ2	0.15474	0.02	0.12347	0.0030948	0.09	0.55562	0.0032387	0.04	0.24694	0.024758	0.29
TZ2-F50		-0.02	-0.23021		0.09	1.0359		-0.02	-0.23021		-0.02
TZ3	0.19421	0.06	0.25822	0.0064506	0.24	1.0329	0.0060005	0.1	0.43036	0.043004	0.7
TZ4	0.19503	0.04	0.25989	0.0064924	0.2	1.2995	0.0075493	0.06	0.38984	0.038955	0.62
TZ4-F50		-0.02	-0.20556		0.1	1.0278		0.02	0.20556		0.08
TZ5-UF		-0.02	-0.76		0.03	1.14		-0.02	-0.76		0.08
TZ5	0.22823	0.06	0.27942	0.0069591	0.31	1.4437	0.0083617	0.09	0.41913	0.041754	0.87
TZ6	0.17743	0.06	0.23398	0.0058411	0.3	1.1699	0.006792	0.08	0.31198	0.031152	0.78
TZ6-F50		-0.02	-0.4907		0.04	0.9814		-0.02	-0.4907		-0.02
TZ7	0.26708	-0.02	-0.22589	0	0.03	0.33884	0.0019581	0.02	0.22589	0.022453	0.08
TZ7-F50		-0.02	-0.3013		0.03	0.45195		-0.02	-0.3013		-0.02
TZ8	0.25182	-0.02	-0.49094	0	-0.02	-0.49094	0	-0.02	-0.49094	0	-0.02
TZ8-F50		-0.02	-0.16471		0.09	0.7412		0.02	0.16471		-0.02
TZ9	0.31223	-0.02	-0.1161	0	0.03	0.17415	0.0010079	0.03	0.17415	0.017336	-0.02
TZ10	0.33483	-0.02	-0.12964	0	-0.02	-0.12964	0	0.03	0.19445	0.019386	-0.02
TZ11	0.39492	-0.02	-0.29028	0	-0.02	-0.29028	0	-0.02	-0.29028	0	-0.02
TZ11-F50		-0.02	-0.22548		0.07	0.78918		-0.02	-0.22548		-0.02
TZ12	0.38365	-0.02	-0.27424	0	-0.02	-0.2742	0	-0.02	-0.2742	0	-0.02
TZ12-F50		-0.02	-0.22861		0.09	1.0288		-0.02	-0.22861		-0.02
TZ13	0.57267	-0.02	-0.17936	0	0.04	0.35872	0.0020708	0.03	0.26904	0.026713	-0.02
TZ13-F50		-0.02	-0.34172		0.09	1.5377		-0.02	-0.34172		0.03
TZ14	0.58166	-0.02	-0.17575	0	0.04	0.3515	0.0020332	0.03	0.26363	0.026228	-0.02
TZ14-F50		-0.02	-0.33474		0.07	1.1716		-0.02	-0.33474		-0.02
TZ15	8.3311	-0.02	-0.7321	0	-0.02	-0.7321	0	0.1	3.6605	0.36568	-0.02
TZ15-F50		-0.02	-0.19394		0.2	1.9394		0.32	3.1031		-0.02
TZ16	7.4246	-0.02	-0.4158	0	0.04	0.8316	0.0048385	0.12	2.4948	0.24967	-0.02
TZ16-F50		0.03	0.2733		0.26	2.369		0.29	2.6422		-0.02
F(i)		0.002	0.002		0.0086	0.0086		0.0005	0.0005		0.0032
BKGND		0	0		0	0		0	0		0

TEST NO.	Cu, ppm	NL(Cu), g/m	Fe, ACL	Fe, ppm	NL(Fe), g/m	K, ACL	K, ppm	NL(K), g/m2	Li, ACL	Li, ppm	NL(Li), g/m
TZ1	1.5746	0.024794	4.66	29.351	0.018533	7.84	49.379	0.080522	10.6	66.763	0.17076
TZ1-F50	-0.23978		0.04	0.47956		3.95	47.357		5.43	65.101	
TZ2	1.7903	0.028047	5.19	32.04	0.020128	8.05	49.697	0.080625	10.9	67.291	0.17123
TZ2-F50	-0.23021		0.04	0.46042		3.87	44.545		5.38	61.926	
TZ3	3.0125	0.047035	12	51.643	0.032334	14.7	63.263	0.10229	19	81.768	0.20738
TZ4	4.0283	0.062895	8.19	53.213	0.033316	9.96	64.713	0.10464	12.5	81.216	0.20598
TZ4-F50	0.82222		0.17	1.7472		5.99	61.564		7.58	77.906	
TZ5-UF	3.04		1.49	56.62		2.16	82.08		2.28	86.64	
TZ5	4.0516	0.063066	15.3	71.252	0.044475	16.8	78.238	0.12612	20.3	94.537	0.23903
TZ6	3.0418	0.047459	13.8	53.816	0.03367	15.8	61.615	0.099557	18.6	72.534	0.18383
TZ6-F50	-0.4907		0.03	0.73605		3.01	73.85		3.29	80.72	
TZ7	0.90356	0.014033	1.33	15.022	0.0093555	8.05	90.921	0.14624	8.71	98.375	0.24818
TZ7-F50	-0.3013		-0.02	-0.3013		6.24	94.006		6.76	101.84	
TZ8	-0.49094	0	0.19	4.6639	0.0029156	3.55	87.142	0.14068	3.63	89.106	0.22564
TZ8-F50	-0.16471		-0.02	-0.16471		11.1	91.415		12	98.827	
TZ9	-0.1161	0	0.1	0.58051	0.00036206	18.7	108.56	0.17485	17	98.687	0.24933
TZ10	-0.12964	0	0.05	0.32409	0.00020245	17.9	116.02	0.18717	15.9	103.06	0.26078
TZ11	-0.29028	0	0.05	0.7257	0.00045122	10	145.14	0.23306	8.11	117.71	0.29647
TZ11-F50	-0.22548		-0.02	-0.22548		13.1	147.69		10.4	117.25	
TZ12	-0.2742	0	0.02	0.2742	0.00017	10.3	141.23	0.22817	8.22	112.71	0.28561
TZ12-F50	-0.22861		-0.02	-0.22861		12.6	144.03		9.97	113.96	
TZ13	-0.17936	0	0.03	0.26904	0.00016737	22.6	202.67	0.32562	16.1	144.38	0.36385
TZ13-F50	0.51258		-0.02	-0.34172		10.9	186.24		7.7	131.56	
TZ14	-0.17575	0	0.04	0.3515	0.00021911	23.2	203.87	0.3282	16.4	144.12	0.3639
TZ14-F50	-0.33474		-0.02	-0.33474		11.3	189.13		8.13	136.07	
TZ15	-0.7321	0	0.05	1.8303	0.0011456	46.5	1702.1	2.7515	10.8	395.33	1.0024
TZ15-F50	-0.19394		0.09	0.87274		160	1551.5		37.6	364.61	
TZ16	-0.4158	0	-0.02	-0.4158	0	71.9	1494.8	2.4206	16.7	347.19	0.88186
TZ16-F50	-0.1822		-0.02	-0.1822		162	1476		38	346.22	
FC(1)	0.0032		0.0798	0.0798		0.0309	0.0309		0.0173	0.0197	
BKGND	0		0	0		0	0		0	0	

TEST NO.	Mg, ACL	Mg, ppm	NL(Mg), g/m	Mn, ACL	Mn, ppm	NL(Mn), g/m	Na, ACL	Na, ppm	NL(Na), g/m	Ni, ACL	Ni, ppm
TZ1	0.69	4.3459	0.027373	1.03	6.4874	0.023349	37.2	234.3	0.17861	0.32	2.0155
TZ1-F50	0.01	0.11989		-0.01	-0.11989		18.1	217		-0.03	-0.35967
TZ2	0.77	4.7536	0.029787	1.16	7.1613	0.025643	37.6	232.12	0.17604	0.36	2.2225
TZ2-F50	0.02	0.23021		-0.01	-0.1151		18.3	210.64		-0.03	-0.34531
TZ3	1.54	6.6275	0.041391	2.59	11.146	0.039778	68.6	295.23	0.22315	0.76	3.2707
TZ4	1.06	6.8871	0.043012	1.74	11.305	0.040346	45.7	296.93	0.22444	0.54	3.5085
TZ4-F50	0.02	0.20556		0.01	0.10278		27.3	280.58		0.06	0.61667
TZ5-UF	0.18	6.84		0.31	11.78		8.19	311.22		0.08	3.04
TZ5	1.88	8.7552	0.054513	3.22	14.996	0.053353	74.6	347.41	0.2618	0.88	4.0982
TZ6	1.68	6.5515	0.040888	2.86	11.153	0.039775	69.3	270.25	0.20413	0.78	3.0418
TZ6-F50	-0.01	-0.24535		-0.01	-0.24535		12.7	311.59		-0.03	-0.73605
TZ7	0.16	1.8071	0.011227	0.27	3.0495	0.010826	37.4	422.41	0.31761	0.09	1.0165
TZ7-F50	-0.01	-0.15065		-0.01	-0.15065		27.2	409.77		-0.03	-0.45195
TZ8	0.02	0.49094	0.0030613	0.04	0.98188	0.0034987	15	368.21	0.27788	-0.03	-0.73641
TZ8-F50	-0.01	-0.082356		-0.01	-0.082356		51.9	427.43		-0.03	-0.24707
TZ9	0.01	0.058051	0.00036116	0.02	0.1161	0.00041275	70.3	408.1	0.30728	-0.03	-0.17415
TZ10	-0.01	-0.064818	0	-0.01	-0.064818	0	67.3	436.23	0.32897	-0.03	-0.19445
TZ11	-0.01	-0.14514	0	-0.01	-0.14514	0	38.2	554.43	0.41619	-0.03	-0.43542
TZ11-F50	0.01	0.11274		-0.01	-0.11274		49.7	560.32		-0.03	-0.33822
TZ12	-0.01	-0.13712	0	-0.01	-0.13712	0	39.3	538.88	0.407	-0.03	-0.41136
TZ12-F50	0.01	0.11431		-0.01	-0.11431		48.2	550.95		-0.03	-0.34292
TZ13	-0.01	-0.089679	0	-0.01	-0.089679	0	86.7	777.52	0.58396	-0.03	-0.26904
TZ13-F50	0.01	0.17086		-0.01	-0.17086		42.1	719.32		-0.03	-0.51258
TZ14	-0.01	-0.087876	0	-0.01	-0.087876	0	88.6	778.58	0.58592	-0.03	-0.26363
TZ14-F50	-0.01	-0.16737		-0.01	-0.16737		43.5	728.05		-0.03	-0.5021
TZ15	-0.01	-0.36605	0	-0.01	-0.36605	0	197	7211.2	5.4493	-0.03	-1.0982
TZ15-F50	0.02	0.19394		-0.01	-0.096971		721	6991.6		0.04	0.38788
TZ16	-0.01	-0.2079	0	-0.01	-0.2079	0	309	6424.1	4.863	-0.03	-0.6237
TZ16-F50	0.02	0.1822		-0.01	-0.09111		706	6432.4		-0.03	-0.2733
F(i)	0.008	0.008		0.014	0.014		0.0661	0.0661		0.0065	0.0065
BKGND	0	0		0	0		0	0		0	0

TEST NO.	NL(Ni), g/m	Si, ACL	Si, ppm	NL(Si), g/m	Th, ACL	Th, ppm	NL(Th), g/m	Ti, ACL	Ti, ppm	NL(Ti), g/m	U, ACL
TZ1	0.015624	45.2	284.69	0.062723	0.09	0.56686	0.012419	0.25	1.5746	0.014693	1.261
TZ1-F50		20.3	243.38		-0.05	-0.59946		-0.02	-0.23978		0.335
TZ2	0.01714	46.3	285.83	0.062654	0.1	0.61735	0.013456	0.28	1.7286	0.016047	1.329
TZ2-F50		19.9	229.06		-0.05	-0.57552		-0.02	-0.23021		0.334
TZ3	0.025141	86	370.11	0.080855	0.19	0.81768	0.017762	0.69	2.9695	0.027475	2.583
TZ4	0.026969	55.9	363.2	0.079346	0.12	0.77968	0.016937	0.47	3.0537	0.028254	1.815
TZ4-F50		29.8	306.28		-0.05	-0.51389		-0.02	-0.20556		0.368
TZ5-UF		10.8	410.4		-0.05	-1.9		0.09	3.42		3.053
TZ5	0.031405	96.8	450.8	0.098183	0.22	1.0245	0.022188	0.89	4.1447	0.038232	
TZ6	0.023364	83.1	324.07	0.070747	0.19	0.74094	0.016084	0.8	3.1198	0.028845	2.874
TZ6-F50		13.5	331.22		-0.05	-1.2267		-0.02	-0.4907		0.119
TZ7	0.0077722	35.5	400.95	0.087132	-0.05	-0.56473	0	0.08	0.90356	0.008316	0.662
TZ7-F50		26.5	399.22		-0.05	-0.75325		-0.02	-0.3013		0.292
TZ8	0	14.7	360.84	0.078709	-0.05	-1.2274	0	-0.02	-0.49094	0	0.23
TZ8-F50		47.6	392.01		-0.05	-0.41178		-0.02	-0.16471		0.6
TZ9	0	69.5	403.45	0.087802	-0.05	-0.29026	0	-0.02	-0.1161	0	0.757
TZ10	0	66	427.8	0.093244	-0.05	-0.32409	0	-0.02	-0.12964	0	0.723
TZ11	0	32.1	465.9	0.10108	-0.05	-0.7257	0	-0.02	-0.29028	0	0.267
TZ11-F50		42.1	474.64		-0.05	-0.5637		-0.02	-0.22548		0.307
TZ12	0	32.7	448.38	0.09787	-0.05	-0.6856	0	-0.02	-0.2742	0	0.278
TZ12-F50		40.5	462.94		-0.05	-0.57153		-0.02	-0.22861		0.305
TZ13	0	65.4	586.5	0.12731	-0.05	-0.4484	0	-0.02	-0.17936	0	0.337
TZ13-F50		31.7	541.63		-0.05	-0.8543		-0.02	-0.34172		0.141
TZ14	0	69.1	607.22	0.13208	-0.05	-0.43938	0	-0.02	-0.17575	0	0.32
TZ14-F50		33.9	567.38		-0.05	-0.83684		-0.02	-0.33474		0.138
TZ15	0	29.6	1083.5	0.23665	-0.05	-1.8303	0	-0.02	-0.7321	0	0.0269
TZ15-F50		100	969.71		-0.05	-0.48485		-0.02	-0.19394		0.0739
TZ16	0	46	956.34	0.20924	-0.05	-1.0395	0	-0.02	-0.4158	0	0.0248
TZ16-F50		104	947.55		-0.05	-0.4556		-0.02	-0.1822		0.0507
f(i)		0.2287	0.2287		0.0023	0.0023		0.0054	0.0054		0.0164
BKGND		0	0		0	0		0	0		0

TEST NO.	U, ppm	NL(U), g/m2	Zn, ACL	Zn, ppm	NL(Zn), g/m	Zr, ACL	Zr, ppm	NL(Zr), g/m
TZ1	7.9423	0.024402	0.02	0.12597	0.031736	-0.05	-0.31492	0
TZ1-F50	4.0163		-0.02	-0.23978		-0.05	-0.59946	
TZ2	8.2046	0.025079	0.02	0.12347	0.030948	-0.05	-0.30868	0
TZ2-F50	3.8445		-0.02	-0.23021		-0.05	-0.57552	
TZ3	11.116	0.033865	0.08	0.34429	0.086007	0.05	0.21518	0.0015358
TZ4	11.793	0.035926	0.16	1.0396	0.2597	-0.05	-0.32487	0
TZ4-F50	3.7822		0.08	0.82222		-0.05	-0.51389	
TZ5-UF	116.01		-0.02	-0.76		-0.05	-1.9	
TZ5			0.07	0.32599	0.081189	0.07	0.32599	0.0023197
TZ6	11.208	0.03412	0.06	0.23398	0.058411	-0.05	-0.19499	0
TZ6-F50	2.9197		-0.02	-0.4907		-0.05	-1.2267	
TZ7	7.477	0.022659	-0.02	-0.22589	0	-0.05	-0.56473	0
TZ7-F50	4.399		-0.02	-0.3013		-0.05	-0.75325	
TZ8	5.6458	0.017173	-0.02	-0.49094	0	-0.05	-1.2274	0
TZ8-F50	4.9414		-0.02	-0.16471		-0.05	-0.41178	
TZ9	4.3945	0.013336	-0.02	-0.1161	0	-0.05	-0.29026	0
TZ10	4.6863	0.014244	-0.02	-0.12964	0	-0.05	-0.32409	0
TZ11	3.8752	0.011724	-0.02	-0.29028	0	-0.05	-0.7257	0
TZ11-F50	3.4611		-0.02	-0.22548		-0.05	-0.5637	
TZ12	3.812	0.0116	-0.02	-0.2742	0	-0.05	-0.6856	0
TZ12-F50	3.4863		-0.02	-0.22861		-0.05	-0.57153	
TZ13	3.0222	0.0091486	-0.02	-0.17936	0	-0.05	-0.4484	0
TZ13-F50	2.4091		0.04	0.68344		-0.05	-0.8543	
TZ14	2.812	0.0085293	-0.02	-0.17575	0	-0.05	-0.43938	0
TZ14-F50	2.3097		0.02	0.33474		-0.05	-0.83684	
TZ15	0.98467	0.0029991	-0.02	-0.7321	0	-0.05	-1.8303	0
TZ15-F50	0.71662		0.02	0.19394		-0.05	-0.48485	
TZ16	0.51559	0.0015731	-0.02	-0.4158	0	-0.05	-1.0395	0
TZ16-F50	0.4619		-0.02	-0.1822		-0.05	-0.4556	
F(i)	0.0164		0.0002	0.0002		0.007	0.007	
BKGND	0		0	0		0	0	

TEST No.	TARGET TIME	pH	DIL FACTOR	SA/V	LEACHANT	ACL NUMBER	Al, ACL	Al, ppm	NL(Al), g/m	B, ACL	B, ppm
TZ31	3	11.02	6.276	1.980e+04	5.06	91-0532-22	0.84	5.272	0.01172	12.1	75.94
TZ31-F50			12.59			91-0532-23	0.1	1.259		5.84	73.5
TZ32	3	11.04	6.528	1.998e+04	5.02	91-0532-24	0.83	5.418	0.01197	11.8	77.02
TZ32-F50			12.55			91-0532-25	0.1	1.255		5.98	75.02
TZ33	7	11.03	6.603	1.990e+04	5.04	91-0532-26	1.39	9.179	0.02133	14.2	93.77
TZ33-F50			10.11			91-0532-27	0.13	1.314		8.5	85.93
TZ34	7	11.01	4.317	1.963e+04	5.11	91-0532-28	1.93	8.331	0.0195	21.5	92.81
TZ35	14	11.23	4.972	2.001e+04	5.01	91-0532-29	2.31	11.49	0.02689	22.3	110.9
TZ35-UF			37.02			91-0532-30	0.28	10.37		2.83	104.8
TZ36	14	11.28	5.139	2.005e+04	4.99	91-0532-31	2.35	12.08	0.0283	21.7	111.5
TZ36-F50			25.07			91-0532-32	-0.1	-2.507		4.28	107.3
TZ37	28	11.37	15.91	1.996e+04	5.01	91-0533-13	0.38	6.046	0.01354	7.46	118.7
TZ37-F50			9.455			91-0533-14	0.19	1.796		13.4	126.7
TZ38	28	11.38	16.74	2.000e+04	5	91-0533-15	0.23	3.85	0.008105	7.22	120.9
TZ38-F50			9.672			91-0533-16	0.2	1.934		13.2	127.7
TZ39	56	11.37	6.179	1.984e+04	5.04	91-0596-07	0.25	1.545	0.002445	23.9	147.7
TZ40	56	11.41	6.098	1.980e+04	5.05	91-0596-08	0.25	1.525	0.0024	24.5	149.4
TZ41	91	11.4	13.27	1.986e+04	5.05	91-0639-11	-0.1	-1.327	-0.00468	13.3	176.5
TZ41-F50			11.49			91-0639-12	-0.1	-1.149		15.8	181.5
TZ42	91	11.38	12.25	1.986e+04	5.04	91-0639-13	0.19	2.327	0.004382	14.7	180
TZ42-F50			11.32			91-0639-14	0.15	1.699		16	181.2
TZ43	182	11.63	8.653	1.975e+04	5.06	92-0134-19	0.13	1.125	0.001409	28.4	245.7
TZ43-F50			16.41			92-0134-20	-0.1	-1.641		14	229.8
TZ44	182	11.66	8.643	1.978e+04	5.06	92-0134-21	0.14	1.21	0.001619	28.2	243.7
TZ44-F50			17.24			92-0134-22	-0.1	-1.724		13.3	229.3
TZ45	364	11.09		1.974e+04	5.07						
TZ45-F50			11.01			92-0485-07	0.1	1.101		310	3414
TZ46	364	11.75	28.6	1.979e+04	5.06	92-0485-08	-0.1	-2.86	-0.008513	122	3489
TZ46-F50			10.03			92-0485-09	0.13	1.304		339	3400
F(1)			1				0.0203	0.0203		0.0248	0.0248
BKGND			1				0.56	0.56		0.15	0.15

TEST NO.	NL(B), g/m2	Ba, ACL	Ba, ppm	NL(Ba), g/m	Ca, ACL	Ca, ppm	NL(Ca), g/m	Cr, ACL	Cr, ppm	NL(Cr), g/m	Cu, ACL
TZ31	0.1543	-0.02	-0.1255	-0.003169	0.09	0.5648	-0.02352	0.03	0.1883	0.01901	0.23
TZ31-F50		-0.02	-0.2517		0.08	1.007		-0.02	-0.2517		-0.02
TZ32	0.1551	-0.02	-0.1305	-0.003266	0.08	0.5222	-0.02355	0.03	0.1958	0.0196	0.24
TZ32-F50		-0.02	-0.2509		0.07	0.8782		-0.02	-0.2509		-0.02
TZ33	0.1897	0.03	0.1981	0.004977	0.16	1.057	-0.02053	0.05	0.3302	0.03318	0.45
TZ33-F50		-0.02	-0.2022		0.18	1.82		-0.02	-0.2022		-0.02
TZ34	0.1903	0.03	0.1295	0.003298	0.23	0.9929	-0.02119	0.06	0.259	0.02639	0.6
TZ35	0.2231	0.04	0.1989	0.004969	0.22	1.094	-0.0202	0.07	0.3481	0.03478	0.66
TZ35-UF		-0.02	-0.7405		-0.02	-0.7405		-0.02	-0.7405		0.07
TZ36	0.224	0.04	0.2056	0.005127	0.21	1.079	-0.02025	0.07	0.3597	0.03589	0.67
TZ36-F50		-0.02	-0.5014		0.06	1.504		-0.02	-0.5014		-0.02
TZ37	0.2394	-0.02	-0.3182	-0.00797	-0.02	-0.3182	-0.02847	-0.02	-0.3182	-0.03188	0.08
TZ37-F50		-0.02	-0.1891		0.09	0.851		0.02	0.1891		-0.02
TZ38	0.2434	-0.02	-0.3348	-0.008371	-0.02	-0.3348	-0.02852	-0.02	-0.3348	-0.03348	0.03
TZ38-F50		-0.02	-0.1934		0.08	0.7738		0.02	0.1934		-0.02
TZ39	0.2998	-0.02	-0.1236	-0.003114	0.02	0.1236	-0.02605	0.02	0.1236	0.01246	-0.02
TZ40	0.3039	-0.02	-0.122	-0.00308	0.02	0.122	-0.02612	0.02	0.122	0.01232	-0.02
TZ41	0.358	-0.02	-0.2654	-0.006682	-0.02	-0.2654	-0.02831	-0.02	-0.2654	-0.02673	-0.02
TZ41-F50		-0.02	-0.2298		0.08	0.9192		-0.02	-0.2298		-0.02
TZ42	0.3652	-0.02	-0.245	-0.006166	-0.02	-0.245	-0.02819	0.02	0.245	0.02466	-0.02
TZ42-F50		-0.02	-0.2265		0.08	0.9059		0.02	0.2265		-0.02
TZ43	0.5013	-0.02	-0.1731	-0.004381	0.04	0.3461	-0.02486	0.03	0.2596	0.02628	-0.02
TZ43-F50		-0.02	-0.3282		0.1	1.641		-0.02	-0.3282		-0.02
TZ44	0.4966	-0.02	-0.1729	-0.00437	0.03	0.2593	-0.02534	0.03	0.2593	0.02622	-0.02
TZ44-F50		-0.02	-0.3448		0.07	1.207		-0.02	-0.3448		-0.02
TZ45											
TZ45-F50		-0.02	-0.2203		0.17	1.872		0.39	4.295		-0.02
TZ46	7.109	-0.02	-0.572	-0.01445	0.02	0.572	-0.02349	0.11	3.146	0.3179	-0.02
TZ46-F50		-0.02	-0.2006		0.19	1.906		0.28	2.808		-0.02
f(1)		0.002	0.002		0.0086	0.0086		0.0005	0.0005		0.0032
BKGND		0	0		4.57	4.57		0	0		0

TEST NO.	Cu, ppm	NL(Cu), g/m	Fe, ACL	Fe, ppm	NL(Fe), g/m	K, ACL	K, ppm	NL(K), g/m2	Li, ACL	Li, ppm	NL(Li), g/m
TZ31	1.4435	0.022778	4.13	25.92	0.016401	8.45	53.032	0.075158	11.3	70.919	0.2069
TZ31-F50	-0.25172		0.03	0.37759		4.11	51.729		5.5	69.224	
TZ32	1.5666	0.024496	4.11	26.828	0.016822	8.22	53.656	0.075487	11.1	72.455	0.20942
TZ32-F50	-0.25091		0.03	0.37636		3.93	49.304		5.48	68.749	
TZ33	2.9715	0.046661	6.69	44.177	0.027817	9.9	65.374	0.094861	12.8	84.524	0.24536
TZ33-F50	-0.20218		0.03	0.30327		6.1	61.666		7.61	76.93	
TZ34	2.5901	0.041231	8.84	38.161	0.02436	15.1	65.184	0.095852	19.4	83.746	0.24644
TZ35	3.2818	0.051242	10.4	51.713	0.032379	15.7	78.067	0.11485	19.6	97.459	0.28133
TZ35-UF	2.5917		1.23	45.539		2.22	82.193		2.44	90.338	
TZ36	3.443	0.053673	10.7	54.985	0.034373	15.2	78.11	0.11474	18.8	96.609	0.27843
TZ36-F50	-0.50143		0.03	0.75214		2.93	73.459		3.5	87.75	
TZ37	1.2729	0.019926	1.16	18.458	0.011586	5.53	87.992	0.13123	6.28	99.926	0.28918
TZ37-F50	-0.1891		0.03	0.28365		9.41	88.972		10.7	101.17	
TZ38	0.50219	0.0078479	0.5	8.3698	0.005245	5.88	98.429	0.1479	5.97	99.935	0.28873
TZ38-F50	-0.19345		-0.02	-0.19345		16.8	162.5		10.6	102.53	
TZ39	-0.12359	-0.0019461	0.08	0.49434	0.00031216	17.4	107.52	0.16386	16.9	104.43	0.30403
TZ40	-0.12197	-0.0019248	0.06	0.3659	0.00023155	17.5	106.72	0.16291	17	103.67	0.30248
TZ41	-0.26541	-0.0041761	0.05	0.66353	0.00041865	10.1	134.03	0.20693	8.68	115.19	0.3351
TZ41-F50	-0.22979		-0.02	-0.22979		12	137.87		10.3	118.34	
TZ42	-0.24496	-0.0038537	0.02	0.24496	0.00015454	10.9	133.5	0.20604	9.47	115.99	0.33738
TZ42-F50	-0.22648		-0.02	-0.22648		12	135.89		10.5	118.9	
TZ43	-0.17306	-0.0027379	0.02	0.17306	0.00010979	20.7	179.12	0.26193	16.6	143.64	0.42019
TZ43-F50	-0.32825		-0.02	-0.32825		10.3	169.05		8.31	136.39	
TZ44	-0.17286	-0.0027314	0.02	0.17286	0.00010953	20.6	178.04	0.27983	16.5	142.61	0.41666
TZ44-F50	-0.34478		-0.02	-0.34478		9.64	166.18		7.91	136.36	
TZ45											
TZ45-F50	-0.22026		0.45	4.9558		129	1420.7		33.6	370.04	
TZ46	-0.572	-0.0090323	0.05	1.43	0.0009055	52.2	1492.9	2.4299	13.4	383.24	1.1192
TZ46-F50	-0.20058		0.06	0.60174		143	1434.1		36.6	367.06	
F(i)	0.0032		0.0798	0.0798		0.0309	0.0309		0.0197	0.0197	
BKGND	0		0	0		7.04	7.04		0.05	0.05	

TEST NO.	Mg, ACL	Mg, ppm	NL(Mg), g/m	Mn, ACL	Mn, ppm	NL(Mn), g/m	Na, ACL	Na, ppm	NL(Na), g/m	Ni, ACL	Ni, ppm
TZ31	0.63	3.954	0.02496	0.96	6.025	0.02173	45.1	283	0.177	0.29	1.82
TZ31-F50	-0.01	-0.1259		-0.01	-0.1259		20.9	263.1		-0.03	-0.3776
TZ32	0.62	4.047	0.02531	0.96	6.266	0.0224	44.3	289.2	0.1801	0.28	1.828
TZ32-F50	0.01	0.1255		-0.01	-0.1255		20.6	258.4		-0.03	-0.3764
TZ33	0.91	6.009	0.03774	1.5	9.905	0.03555	51.1	337.4	0.2175	0.43	2.839
TZ33-F50	0.02	0.2022		-0.01	-0.1011		29.6	299.2		-0.03	-0.3033
TZ34	1.19	5.137	0.03271	1.98	8.547	0.0311	76.4	329.8	0.2146	0.56	2.417
TZ35	1.34	6.663	0.04161	2.32	11.54	0.04117	76.8	381.9	0.2499	0.64	3.182
TZ35-UF	0.16	5.924		0.27	9.996		9.53	352.8		0.07	2.592
TZ36	1.37	7.04	0.0439	2.38	12.23	0.04358	74.4	382.3	0.2498	0.66	3.392
TZ36-F50	-0.01	-0.2507		-0.01	-0.2507		14.2	356		-0.03	-0.7521
TZ37	0.14	2.228	0.01395	0.25	3.978	0.01423	25.3	402.6	0.2662	0.07	1.114
TZ37-F50	0.01	0.09455		-0.01	-0.09455		47.3	447.2		-0.03	-0.2837
TZ38	0.06	1.004	0.006278	0.11	1.841	0.006577	25	418.5	0.2778	0.04	0.6696
TZ38-F50	-0.01	-0.09672		-0.01	-0.09672		46.4	448.8		-0.03	-0.2902
TZ39	-0.01	-0.06179	-0.0003892	0.01	0.06179	0.0002224	72	444.9	0.3001	-0.03	-0.1854
TZ40	-0.01	-0.06098	-0.000385	0.01	0.06098	0.00022	71.5	436	0.2939	-0.03	-0.1829
TZ41	-0.01	-0.1327	-0.0008352	-0.01	-0.1327	-0.0004773	41.7	553.4	0.3824	-0.03	-0.3981
TZ41-F50	0.01	0.1149		-0.01	-0.1149		48.8	560.7		-0.03	-0.3447
TZ42	-0.01	-0.1225	-0.0007707	-0.01	-0.1225	-0.0004404	44.4	543.8	0.3751	-0.03	-0.3674
TZ42-F50	0.01	0.1132		-0.01	-0.1132		49.3	558.3		-0.03	-0.3397
TZ43	-0.01	-0.08653	-0.0005476	-0.01	-0.08653	-0.0003129	84.9	734.6	0.5234	-0.03	-0.2596
TZ43-F50	0.01	0.1641		-0.01	-0.1641		42.8	702.5		-0.03	-0.4924
TZ44	-0.01	-0.08643	-0.0005463	-0.01	-0.08643	-0.0003122	84.1	726.9	0.5168	-0.03	-0.2593
TZ44-F50	0.01	0.1724		-0.01	-0.1724		40.6	699.9		-0.03	-0.5172
TZ45											
TZ45-F50	0.03	0.3304		0.02	0.2203		590	6498		0.19	2.092
TZ46	-0.01	-0.286	-0.001806	-0.01	-0.286	-0.001032	228	6521	4.946	-0.03	-0.858
TZ46-F50	0.02	0.2006		-0.01	-0.1003		644	6459		-0.03	-0.3009
f(i)	0.008	0.008		0.014	0.014		0.0661	0.0661		0.0065	0.0065
BKGND	0	0		0	0		51.3	51.3		0	0

TEST NO.	NL(Ni), g/m	Si, ACL	Si, ppm	NL(Si), g/m	Th, ACL	Th, ppm	NL(Th), g/m	Ti, ACL	Ti, ppm	NL(Ti), g/m	U, ACL
TZ31	0.01414	44.4	278.7	0.05154	0.09	0.5648	0.0124	0.23	1.443	0.0135	1.309
TZ31-F50		19.3	242.9		-0.05	-0.6293		-0.02	-0.2517		0.407
TZ32	0.01407	43.4	283.3	0.05209	0.09	0.5875	0.01278	0.23	1.501	0.01391	1.289
TZ32-F50		19.4	243.4		-0.05	-0.6273		-0.02	-0.2509		0.38
TZ33	0.02195	53.2	351.3	0.06725	0.12	0.7924	0.01731	0.4	2.641	0.02458	1.806
TZ33-F50		26.4	266.9		-0.05	-0.5055		-0.02	-0.2022		0.511
TZ34	0.01894	78.2	337.6	0.06512	0.18	0.777	0.01721	0.55	2.374	0.0224	2.628
TZ35	0.02446	82.7	411.2	0.07997	0.21	1.044	0.02268	0.66	3.282	0.03037	2.834
TZ35-UF		10.5	388.7		-0.05	-1.851		0.07	2.592		0.382
TZ36	0.02603	81.2	417.3	0.08116	0.2	1.028	0.02229	0.67	3.443	0.03181	2.841
TZ36-F50		12.9	323.4		-0.05	-1.254		-0.02	-0.5014		0.248
TZ37	0.008583	22.7	361.2	0.06921	-0.05	-0.7956	-0.01733	0.08	1.273	0.01181	0.588
TZ37-F50		38.1	360.2		-0.05	-0.4728		-0.02	-0.1891		0.663
TZ38	0.005151	20.9	349.9	0.06662	-0.05	-0.837	-0.0182	0.03	0.5022	0.004651	0.463
TZ38-F50		37.4	361.7		-0.05	-0.4836		-0.02	-0.1934		0.657
TZ39	-0.001437	61.4	379.4	0.07364	-0.05	-0.309	-0.006769	-0.02	-0.1236	-0.001153	0.96
TZ40	-0.001421	63.2	385.4	0.07512	-0.05	-0.3049	-0.006695	-0.02	-0.122	-0.001141	1.012
TZ41	-0.003084	32.3	428.6	0.08442	-0.05	-0.6635	-0.01453	-0.02	-0.2654	-0.002475	0.395
TZ41-F50		38.3	440		-0.05	-0.5745		-0.02	-0.2298		0.391
TZ42	-0.002846	35.6	436	0.08603	-0.05	-0.6124	-0.0134	-0.02	-0.245	-0.002284	0.431
TZ42-F50		39.4	446.2		-0.05	-0.5662		-0.02	-0.2265		0.444
TZ43	-0.002022	65	562.5	0.1145	-0.05	-0.4327	-0.009523	-0.02	-0.1731	-0.001622	0.412
TZ43-F50		32.4	531.8		-0.05	-0.8206		-0.02	-0.3282		0.196
TZ44	-0.002017	65.6	567	0.1154	-0.05	-0.4321	-0.0095	-0.02	-0.1729	-0.001619	0.399
TZ44-F50		31.3	539.6		-0.05	-0.862		-0.02	-0.3448		0.181
TZ45											
TZ45-F50		83.6	920.7		-0.05	-0.5507		-0.02	-0.2203		0.0128
TZ46	-0.00667	37.2	1064	0.2251	-0.05	-1.43	-0.03142	-0.02	-0.572	-0.005352	0.0183
TZ46-F50		96.8	970.8		-0.05	-0.5015		-0.02	-0.2006		0.0383
f(i)		0.2287	0.2287		0.0023	0.0023		0.0054	0.0054		0.0164
BKGND		45.2	45.2		0	0		0	0		0

TEST NO.	U, ppm	NL(U), g/m2	Zn, ACL	Zn, ppm	NL(Zn), g/m	Zr, ACL	Zr, ppm	NL(Zr), g/m
TZ31	8.215	0.02529	-0.02	-0.1255	-0.03169	-0.05	-0.3138	-0.02264
TZ31-F50	5.123	0.01577	-0.02	-0.2517		-0.05	-0.6293	
TZ32	8.414	0.02567	-0.02	-0.1305	-0.03266	-0.05	-0.3264	-0.02333
TZ32-F50	4.767	0.01455	-0.02	-0.2509		-0.05	-0.6273	
TZ33	11.93	0.03654	0.07	0.4622	0.1161	-0.05	-0.3302	-0.0237
TZ33-F50	5.166	0.01583	-0.02	-0.2022		-0.05	-0.5055	
TZ34	11.34	0.03524	0.08	0.3453	0.08796	-0.05	-0.2158	-0.01571
TZ35	14.09	0.04293	0.05	0.2486	0.06211	0.05	0.2486	0.01775
TZ35-UF	14.14	0.04309	-0.02	-0.7405		-0.05	-1.851	
TZ36	14.6	0.04441	0.05	0.2569	0.06409	0.05	0.2569	0.01831
TZ36-F50	6.218	0.01891	-0.02	-0.5014		-0.05	-1.254	
TZ37	9.356	0.02858	-0.02	-0.3182	-0.0797	-0.05	-0.7956	-0.05693
TZ37-F50	6.269	0.01915	-0.02	-0.1891		-0.05	-0.4728	
TZ38	7.75	0.02363	-0.02	-0.3348	-0.08371	-0.05	-0.837	-0.05979
TZ38-F50	6.355	0.01938	-0.02	-0.1934		-0.05	-0.4836	
TZ39	5.932	0.01823	-0.02	-0.1236	-0.03114	-0.05	-0.309	-0.02224
TZ40	6.171	0.019	-0.02	-0.122	-0.0308	-0.05	-0.3049	-0.022
TZ41	5.242	0.01609	-0.02	-0.2654	-0.06682	-0.05	-0.6635	-0.04773
TZ41-F50	4.492	0.01379	-0.02	-0.2298		-0.05	-0.5745	
TZ42	5.279	0.0162	-0.02	-0.245	-0.06166	-0.05	-0.6124	-0.04404
TZ42-F50	5.028	0.01543	-0.02	-0.2265		-0.05	-0.5662	
TZ43	3.565	0.01101	-0.02	-0.1731	-0.04381	-0.05	-0.4327	-0.03129
TZ43-F50	3.217	0.00993	0.02	0.3282		-0.05	-0.8206	
TZ44	3.449	0.01063	-0.02	-0.1729	-0.0437	-0.05	-0.4321	-0.03122
TZ44-F50	3.12	0.00962	-0.02	-0.3448		-0.05	-0.862	
TZ45								
TZ45-F50	0.141		0.08	0.881		-0.05	-0.5507	
TZ46	0.5234	0.001613	-0.02	-0.572	-0.1445	-0.05	-1.43	-0.1032
TZ46-F50	0.3841		0.05	0.5015		-0.05	-0.5015	
F(i)	0.0164		0.0002	0.0002		0.0007	0.0007	
BKGND	0		0	0		0	0	

Table D.4. Anion Results

<u>Column Heading</u>	<u>Comment</u>
TEST No.	Test number in test matrix (Table D.1).
REACTION TIME	Actual reaction time, in days.
pH	Leachate pH measured at room temperature.
C dil. factor	Dilution factor of sample used in carbon analyses.
TC analyzed	Total carbon in sample analyzed, in parts per million of carbon.
TOC analyzed	Total organic carbon in sample analyzed, in parts per million of carbon.
TC, ppm	Total carbon corrected for dilution, in parts per million of carbon.
TOC, ppm	Total organic carbon corrected for dilution, in parts per million of carbon.
TIC, ppm	Total inorganic carbon calculated as TC-TOC, in parts per million of carbon.
ACL Number	Solution identification number.
F-, ppm	Fluoride content, in parts per million.
CHO ₂ -, ppm	Formate content, in parts per million.
Cl-, ppm	Chloride content, in parts per million.
NO ₂ -, ppm	Nitrite content, in parts per million.
NO ₃ -, ppm	Nitrate content, in parts per million.
HPO ₄ =, ppm	Hydrogen phosphate content, in parts per million.
SO ₄ =, ppm	Sulfate content, in parts per million.
C ₂ O ₄ =, ppm	Oxalate content, in parts per million.

TEST No.	Reaction Time	pH	C dil. factor	TC analyzed	TOC analyzed	TC	TOC	TIC	
BX1	7	8.21	1	23.65	1.815	23.65	1.815	21.84	
BX2	7	8.2	1	23.99	1.904	23.99	1.904	22.09	
BX3	28	8.28	1	22.5	2.108	22.5	2.108	20.39	
BX4	28	8.34	1	19.97	1.557	19.97	1.557	18.41	
BX5	94	8.26	1	26.48	3.8	26.48	3.8	22.68	
BX6	94	8.59	1	23.8	2.745	23.8	2.745	21.06	
BX7	238	8.43	1	21.6	2.319	21.6	2.319	19.28	
BX8	238	9.04	1	19.82	2.309	19.82	2.309	17.51	
BX9	600	8.57	1	27.64	2.699	27.64	2.699	24.94	
BX10	600	8.58	1						
BX11	1600	8.74	1	21.11	1.305	21.11	1.305	19.8	
BX12	1600	8.91	1	22.14	1.856	22.14	1.856	20.28	
TEST No.	ACL Number	F, ppm	CH02-, ppm	Cl-, ppm	NO2-, ppm	NO3-, ppm	HP04=, ppm	SO4=, ppm	C204=, ppm
BX1	91-2010-01	2.7	-0.5	9.2	-0.2	17	-0.5	24	-0.5
BX2	91-2010-02	2.8	-0.5	9.4	-0.2	18	-0.5	24	-0.5
BX3	91-2010-03	2.8	-0.5	9.2	-0.2	17	-0.5	24	-0.5
BX4	91-2010-04	2.7	-0.5	9.2	-0.2	17	-0.5	24	-0.5
BX5									
BX6									
BX7									
BX8									
BX9	93-2154-01	5.9	-0.3	16	1.4	21	0.5	58	-0.5
BX10	92-2102-37	4.6		11	-0.2	12	-1	31	1.4
BX11	94-2110-06	4.2	4.1	8.2	0.61	10	1.1	29	0.53
BX12	94-2110-07	4.3	3.5	8	0.25	9.7	-0.5	29	0.84

Test No.	Reaction Time	pH	C dil. factor	TC analyzed	TOC analyzed	TC	TOC	TIC
BY1	3	9.27	1	36.97	7.316	36.97	7.316	29.65
BY2	3	9.3	1	38.13	7.63	38.13	7.63	30.5
BY3	7	9.48	1	45.73	8.501	45.73	8.501	37.23
BY4	7	9.48	1	67.82	9.609	67.82	9.609	58.21
BY5	14	9.77	1	46.04	11.93	46.04	11.93	34.11
BY6	14	9.74	1	74.58	10.73	74.58	10.73	63.85
BY7	30	9.94	1	46.99	11.74	46.99	11.74	35.25
BY8	30	9.87	1	51.39	9.191	51.39	9.191	42.2
BY9	70	10.09	1	69.35	39.36	69.35	39.36	29.99
BY10	70	10.11	1	75.23	39.44	75.23	39.44	35.79
BY11	140	10.02	5	9.065	3.053	45.33	15.27	30.06
BY12	140	10.11	5	10.27	2.625	51.35	13.13	38.23
BY13	280	10.47	1	44.23	12.32	44.23	12.32	31.91
BY14	280	10.45	1	40.84	9.526	40.84	9.526	31.31
BY15	560	10.33	11	8.076	3.17	88.84	36.49	52.35
BY16	560	10.42	11	15.14	2.94	166.5	32.34	134.2
BY17	980	10.52	11	14.38	1.883	158.2	20.71	137.5
BY18	980	10.65	11	14.62	2.528	160.8	27.81	133

Test No.	ACL Number	F-, ppm	CH02-, ppm	Cl-, ppm	N02-, ppm	N03-, ppm	HPO4-, ppm	S04-, ppm	C204-, ppm
BY1	91-2010-05	2.8	-0.5	9.9	0.88	15	-0.5	26	-0.5
BY2	91-2010-06	2.9	-0.5	10	0.97	16	-0.5	26	-0.5
BY3	91-2010-07	2.9	-0.5	10	-0.2	16	-0.5	26	-0.5
BY4	91-2010-08	2.9	-0.5	10	-0.2	16	-0.5	26	-0.5
BY5	91-2010-09	3	-0.5	11	0.85	16	-0.5	27	-0.5
BY6	91-2010-10	3	-0.5	11	0.46	16	-0.5	26	-0.5
BY7	91-2010-11	3	-0.5	11	0.88	16	-0.5	26	-0.5
BY8	91-2010-12	3.1	-0.5	11	1.1	28	-0.5	29	-0.5
BY9	91-2068-01	5		13	0.4	30	5.1	26	1.6
BY10	91-2068-02	2.6	3.8	12	0.51	11	4.6	26	2.2
BY11	91-2068-03	3.1		11	0.85	11	6.1	25	1.4
BY12	91-2068-04	3	3.6	11	0.76	11	6.1	25	2.1
BY13	91-2068-05	2.8	6	10	0.54	11	8.2	28	2.6
BY14	91-2068-06	2.7	5.3	10	0.72	11	8.1	33	2.4
BY15	92-2068-04	4.1		12	-0.2	8.9	6.1	33	3.3
BY16	92-2068-04	4.1		10	-0.2	8.8	5.9	31	2.7
BY17	93-2154-05	5	-0.3	18	1.5	22	9.2	68	4.3
BY18	93-2154-06	4.7	-0.3	17	1.7	20	10	64	5

Test No.	Reaction Time	pH	C dil. factor	TC analyzed	TOC analyzed	TC	TOC	TIC
BZ1	3	10.5	2.5	56.07	23.1	140.2	57.75	82.43
BZ2	3	10.62	2.5	47.12	20.63	117.8	51.58	66.23
BZ3	7	10.73	2.5	54.15	22.12	135.4	55.3	80.08
BZ4	7	10.48	2.5	58.28	27.69	145.7	69.23	76.48
BZ5	14	10.98	2.5	84.24	27.9	210.6	69.75	140.9
BZ6	14	11.01	2.5	57.22	28.35	143.1	70.88	72.18
BZ7	28	11.02	2	77.63	39.88	155.3	79.76	75.5
BZ8	28	11.1	2	81.08	40.39	162.2	80.78	81.38
BZ9	56	11.24	1.67	138.4	93.36	231.1	155.9	75.22
BZ10	56	11.33	1.67	144.4	96.48	241.2	161.1	80.03
BZ11	98	11.26	2.5	102.4	65.59	256	164	92.03
BZ12	98	11.43	2.5	107.8	63.96	269.5	159.9	109.6
BZ13	182	10.92	11	30.63	20.11	336.9	221.2	115.7
BZ14	182	11.29	11	25.08	17.29	275.9	190.2	85.69
BZ15	364	12.03	11	13.43	7.117	147.7	78.29	69.44
BZ16	364	12.02	11	14.86	7.331	163.5	80.64	82.82
BZ17	736	11.96	11	12.58	8.903	138.4	97.93	40.45
BZ18	736	11.97	11	15.63	15.21	171.9	167.3	4.62
BZ19	484	11.86	11	9.269	8.465	102	93.12	8.844

Test No.	ACL Number	ACL F-, ppm	ACL, CHO2-, pp	Cl-, ppm	NO2-, ppm	NO3-, ppm	HPO4=, ppm	SO4=, ppm	C2O4=, ppm
BZ1	91-2010-13	3	15	12	1.7	17	7.7	34	5.1
BZ2	90-4085-11	3.6	12	12	0.84	16	7.4	35	3.6
BZ3	91-2010-14	3	14	15	1.5	19	10	39	6.4
BZ4	91-2010-15	3	18	13	0.95	18	11	38	6.4
BZ5	91-2010-16	3	19	13	1.3	17	13	37	7.7
BZ6	90-4085-12	3	17	12	1	16	12	35	7.2
BZ7	91-2010-17	3	22	14	1.5	18	18	40	11
BZ8	90-4085-13	3	23	13	1.2	15	15	39	11
BZ9	91-2045-11		27	14	-0.02	18	22	42	14
BZ10	91-2045-12		28	14	-0.02	17	23	40	14
BZ11	91-2045-13		40	15	-0.02	22	28	42	14
BZ12	91-2045-14		31	14	-0.02	16	28	39	14
BZ13	91-2068-07	5.6	35	19	1.9	251	51	44	18
BZ14	91-2068-08	2.2	26	18	1.9	151	71	39	10
BZ15	92-2005-01			-10	216	12	121	95	15
BZ16	92-2005-02			-10	171	17	257	85	17
BZ17	93-2154-07	112	-20	447	22	48	115	612	104
BZ18	92-2103-39	27		215	-15	26	103	293	56
BZ19	92-2103-40	8.7		119	-15	28	119	183	-50

Test No.	Reaction Ti	pH	C dil. fact	TC analyzed	TOC analyze	TC	TOC	TIC	Test No.	ACL Number
PX1	7	8.18	1	23.84	1.873	23.84	1.873	21.97	PX1	91-2010-18
PX2	7	8.24	1	23.49	1.662	23.49	1.662	21.83	PX2	91-2010-19
PX3	14	8.3	1	24.36	2.108	24.36	2.108	22.25	PX3	91-2010-20
PX4	14	8.24	1	24.15	2.25	24.15	2.25	21.9	PX4	91-2010-21
PX5	94	8.44	1	25.68	2.592	25.68	2.592	23.09	PX5	
PX6	94	8.31	1	24.56	2.57	24.56	2.57	21.99	PX6	
PX7	238	8.58	1	28.85	3.049	28.85	3.049	25.8	PX7	
PX8	238	9.27	1	25.76	2.926	25.76	2.926	22.83	PX8	
PX9	601	9.09	11	4.399	0.57	48.39	6.27	42.12	PX9	93-2154-08
PX10	601	9.34	11	4.438	0.582	48.82	6.402	42.42	PX10	92-2102-38
PX11	1400	8.96	1	29.47	2.402	29.47	2.402	27.07	PX11	94-2110-01
PX12	1400	9.32	1	30.89	2.613	30.89	2.613	28.28	PX12	94-2110-02
F-, ppm	CH02-, ppm	Cl-, ppm	NO2-, ppm	NO3-, ppm	HP04-, ppm	SO4-, ppm	C204-, ppm			
3.2	-0.5	9.8	-0.2	17	-0.5	25	-0.5			
2.9	-0.5	9.6	-0.2	19	-0.5	25	-0.5			
2.8	-0.5	9.8	0.41	16	-0.5	26	-0.5			
2.8	-0.5	9.7	0.93	16	-0.5	26	-0.5			
6.5	-0.3	20	0.98	24	1.4	67	0.57			
4.3		12	-0.2	12	-1	29	1.3			
4.8	5.1	11	0.73	13	0.63	37	0.68			
4.7	4.1	9.6	0.61	11	0.66	32	0.82			

Test No.	Reaction Time	pH	C dil. factor	TC analyzed	TOC analyzed	TC	TOC	TIC
PY1	3	9.93	1	32.56	4.535	32.56	4.535	28.03
PY2	3	10.07	1	47.99	4.72	47.99	4.72	43.27
PY3	7	10.26	1	39.6	5.459	39.6	5.459	34.14
PY4	7	10.25	1	37.45	5.834	37.45	5.834	31.62
PY5	14	10.45	1	36.53	10.2	36.53	10.2	26.33
PY6	14	10.45	1	39.41	12.18	39.41	12.18	27.23
PY7	30	10.47	1	39.18	9.839	39.18	9.839	29.34
PY8	30	10.52	1	41.46	10.79	41.46	10.79	30.67
PY9	70	10.61	1	71.21	30.47	71.21	30.47	40.74
PY10	70	10.66	1	69.5	33.91	69.5	33.91	35.59
PY11	140	10.72	5	9.782	2.948	48.91	14.74	34.17
PY12	140	10.72	5	8.479	1.57	42.4	7.85	34.55
PY13	280	11.44	11	7.654	2.538	84.19	27.93	56.28
PY14	280	11.48	11	6.472	1.857	71.19	20.43	50.77
PY15	560	11.44	11	6.571	1.095	72.28	12.05	60.24
PY16	560	11.49	11	6.291	1.252	69.2	13.77	55.43
PY17	980	11.74	11	6.05	1.605	66.55	17.66	48.9
PY18	980	11.72	11	8.719	2.182	95.91	24	71.91

Test No.	ACL Number	F-, ppm	CH02-, ppm	Cl-, ppm	NO2-, ppm	NO3-, ppm	HP04-, ppm	S04-, ppm	C204-, ppm
PY1	91-2010-22	2.6	-0.5	10	-0.2	16	-0.5	26	-0.5
PY2	91-2010-23	2.7	-0.5	9.7	0.93	16	-0.5	26	-0.5
PY3	91-2010-24	2.7	-0.5	11	-0.2	18	-0.5	27	-0.5
PY4	91-2010-25	2.7	-0.5	11	-0.2	18	-0.5	27	-0.5
PY5	91-2010-26	2.7	-0.5	11	-0.2	17	-0.5	27	-0.5
PY6	91-2010-27	2.7	-0.5	10	-0.2	16	-0.5	27	-0.5
PY7	91-2010-28	2.5	-0.5	10	1.3	16	-0.5	29	-0.5
PY8	91-2010-29	2.4	-0.5	10	1	16	-0.5	28	-0.5
PY9	91-2068-18	1.9	5.6	11	-0.2	10	-2	25	-1
PY10	91-2068-19	1.8	2.8	13	0.96	12	-2	29	-1
PY11	91-2068-20	1.6	2.6	11	0.39	11	-2	28	-1
PY12	91-2068-21	1.7	2.5	12	0.63	11	-2	28	-1
PY13	91-2081-09	1.7	3.1	12	0.73	12	5.6	40	-1
PY14									
PY15	92-2063-01	3.3		11	0.81	8.9	9.1	35	1.3
PY16	92-2063-02	2.8		12	0.81	8.4	8.5	32	1.1
PY17	93-2154-12	14	-5	50	-8	16	-40	164	-40
PY18	93-2154-13	14	-5	59	18	60	-40	180	-40

Test No.	Reaction Time	pH	C dil. factor	TC analyzed	TOC analyzed	TC	TOC	TIC	
PZ1	3	11.94	2.5	56.95	16.67	142.4	41.68	100.7	
PZ2	3	11.86	2.5	33.59	11.46	83.98	28.65	55.33	
PZ3	7	11.99	2.5	34.49	28.21	86.23	70.53	15.7	
PZ4	7	11.98	2.5	42.91	35.24	107.3	88.1	19.18	
PZ5	14	12.05	2.5	41.68	38.41	104.2	96.03	8.175	
PZ6	14	12.04	2.5	42.25	36.5	105.6	91.25	14.38	
PZ7	28	12.12	2.5	44.45	40.9	111.1	102.3	8.875	
PZ8	28	12.11	2.5	37.38	26.17	93.45	65.43	28.03	
PZ9	56	12.12	1.67	55.21	49.46	92.2	82.6	9.603	
PZ10	56	12.14	1.67	104.2	86.23	174	144	30.01	
PZ11	98	12.14	2.5	72.74	64.94	181.9	162.4	19.5	
PZ12	98	12.12	2.5	72.64	64.84	181.6	162.1	19.5	
PZ13	182	12.18	10	8.807	2.766	88.07	27.66	60.41	
PZ14	182	12.19	10	9.743	2.929	97.43	29.29	68.14	
PZ15	364	12.2	11	9.708	9.633	106.8	106	0.825	
PZ16	364	12.17	11	9.977	9.2	109.8	101.2	8.547	
PZ17	728	12.02	11	11.81	11.5	129.9	126.5	3.41	
PZ18	728	12.05	11	11.02	10.66	121.2	117.3	3.96	
PZ19	552	12.04	11	12.43	11.17	136.7	122.9	13.86	
PZ20	1100	12.18	11	12.15	11.59	133.7	127.5	6.2	
Test No.	ACL Number	F-, ppm	CH02-, ppm	Cl-, ppm	NO2-, ppm	NO3-, ppm	HP04=, ppm	SO4=, ppm	C204=, ppm
PZ1	91-2010-30	7.3	-0.5	28	-0.2	19	24	90	-0.5
PZ2	91-2010-31	7.3	-0.5	28	-0.2	17	26	92	-0.5
PZ3	91-2010-32	12	-0.5	50	-0.2	20	56	141	-0.5
PZ4	91-2010-33	12	-0.5	51	-0.2	19	54	136	-0.5
PZ5	91-2010-34	13	-0.5	63	-0.2	21	66	159	-0.5
PZ6	91-2010-35	13	-0.5	66	-0.2	21	66	150	-0.5
PZ7	91-2010-36	14	-0.5	76	-0.2	21	79	174	-0.5
PZ8	91-2010-37	14	-0.5	81	-0.2	21	84	180	-0.5
PZ9	91-2045-15	16	-0.5	85	-0.2	19	60	179	-1
PZ10	91-2045-16	16	-0.5	93	-0.2	21	105	181	-1
PZ11	91-2045-17	16	-0.5	88	-0.2	20	103	192	-1
PZ12	91-2045-18	16	-0.5	90	-0.2	20	99	191	-1
PZ13	93-2154-14	-3	-5	76	-8	23	166	827	-40
PZ14	93-2154-15	-3	-5	149	-8	27	190	775	-40
PZ15	92-2005-03			236	-15	14	280	1490	-0.7
PZ16	92-2005-04			210	-15	12	244	1300	-0.7
PZ17	93-2154-16	-3	-5	382	-8	-15	574	2070	-40
PZ18	93-2154-17	-3	-5	300	-8	18	460	1710	-40
PZ19	93-2154-18	-3	-5	215	-8	20	381	1250	-40
PZ20	94-2110-03	-20	-20	189	-20	18	414	1150	-50

Test No.	Reaction Time	pH	C dil. factor	TC analyzed	TOC analyzed	TC, ppm	TOC, ppm	TIC, ppm		
TW1	14	8.95	11	3.084	2.112	33.92	23.23	10.69		
TW2	14	9.31	11	2.982	1.185	32.8	13.04	19.77		
TW3	28	9.24	11	4.756	2.181	52.32	23.99	28.33		
TW4	28	9.28	11	8.037	3.153	88.41	34.68	53.72		
TW5	56	9.5	11	4.651	1.946	51.16	21.41	29.76		
TW6	56	9.44	11	3.911	1.435	43.02	15.79	27.24		
TW7	91	9.36	11	4.871	4.681	53.58	51.49	2.09		
TW8	91	9.41	11	4.183	4.064	46.01	44.7	1.309		
TW9	180	9.4	11	6.341	1.358	69.75	14.94	54.81		
TW10	180	9.54	11	6.149	1.381	67.64	15.19	52.45		
TW11	360	9.63	11	3.317	0.622	36.49	6.84	29.65		
TW12	361	9.59	11	4.828	1.108	53.11	12.19	40.92		
TW13	540	10.48	11	11.91	3.883	131	42.71	88.3		
TW14	540	9.58	11	3.223	0.976	35.45	10.74	24.72		
TW15	720	9.31	11	6.86	1.085	75.46	11.94	63.53		
TW16	720	9.75	11	9.47	1.235	104.2	13.58	90.59		
Test No.	ACL NUMBER	F, ppm	CH02-, ppm	Cl-, ppm	NO2-, ppm	NO3-, ppm	HP04=, ppm	SO4=, ppm	C204=, ppm	
TW1										
TW2										
TW3	93-2154-19	1.6	-0.3	0.95	0.59	-0.2	2.3	4.3	-0.5	
TW4	93-2154-20	1.2	-0.3	0.63	0.24	-0.2	2.3	3.6	-0.5	
TW5	93-2154-21	2.5	-0.3	0.95	0.74	-0.2	3.6	5.9	0.69	
TW6	93-2154-22	2.6	-0.3	0.98	1.5	1.2	4.1	4.7	-0.5	
TW7										
TW8	92-2103-49	2.1		1.3	2.1	0.66	3.2	2.1	-1	
TW9	93-2154-23	3.1	-0.3	1.9	0.26	-0.2	5.4	6.3	-0.5	
TW10	92-2103-50	2.1		1.1	-0.2	0.29	2.9	1.9	-1	
TW11	93-2154-24	6.9	-0.3	1.2	1.5	0.76	8.7	8.4	-0.5	
TW12	92-2103-51	3.7		0.7	-0.2	0.36	3.7	2.5	1.7	
TW13										
TW14	93-2154-25	6	-0.3	1.4	1.4	1.9	5	6	-0.5	
TW15	93-2154-26	5	-0.3	0.67	0.3	0.22	4.4	3.8	-0.5	
TW16	93-2154-27	5.7	-0.3	1.4	0.82	0.63	4.5	7.2	1.4	

TEST NO.	Reaction Time	pH	C dil. factor	TC analyzed	TOC analyzed	TC, ppm	TOC, ppm	TIC, ppm	
TW31	14	9.38	11	4.419	1.624	48.61	17.86	30.75	
TW32	14	9.23	11	3.381	0.808	37.19	8.888	28.3	
TW33	28	9.44	11	5.637	1.52	62.4	16.72	45.68	
TW34	28	9.45	11	5.678	1.974	62.46	21.71	40.74	
TW35	56	9.66	11	4.405	3.005	48.46	33.06	15.4	
TW36	56	9.71	11	4.147	1.562	45.62	17.18	28.44	
TW37	91	9.74	11	5.017	2.166	55.19	23.83	31.36	
TW38	91	9.63	11	4.754	4.155	52.29	45.71	6.589	
TW39	180	9.88	11	9.483	2.025	104.3	22.28	82.04	
TW40	180	9.69	11	7.275	1.527	80.03	16.8	63.23	
TW41	360	9.79	11	3.299	0.564	36.29	6.204	30.09	
TW42	360	9.79	11	3.974	0.703	43.71	7.733	35.98	
TW43	540	9.69	11	4.461	0.823	49.07	9.053	40.02	
TW44	540	9.78	11	3.834	4.434	42.17	48.77	-6.6	
TW45	720	9.24	11	6.215	0.733	68.37	8.06	60.3	
TW46	720	9.42	11	6.455	0.945	71.01	10.4	60.61	
Test No.	ACL Number	F-, ppm	CH02-, ppm	Cl-, ppm	NO2-, ppm	NO3-, ppm	HP04=, ppm	S04=, ppm	C204=, ppm
TW31									
TW32									
TW33	93-2154-28	6.3	-0.3	20	1.4	21	0.93	61	-0.5
TW34	93-2154-29	8.7	-0.3	22	1.6	24	-0.5	68	-0.5
TW35	93-2154-30	8	-0.3	20	1.9	23	1.8	62	-0.5
TW36	93-2154-31	6.8	-0.3	18	1.5	20	2.1	58	-0.5
TW37	93-2154-32	7.5	-0.3	16	1.9	19	2.5	57	-0.5
TW38	92-2103-52	4.6		12	-0.2	13	1.4	29	-1
TW39	93-2154-33	20	-0.3	44	3.3	46	9.5	155	-0.5
TW40	92-2103-53	5.7		11	-0.2	12	1.3	27	-1
TW41	93-2154-34	12	-0.3	16	2.2	16	3.9	53	-0.5
TW42	92-2103-54	7.2		10	-0.2	10	2.2	26	1.1
TW43									
TW44	93-2154-35	11	-0.3	14	2	12	3.9	47	-0.5
TW45	93-2154-36	13	-0.3	14	1.4	14	2	46	-0.5
TW46	93-2154-37	13	-0.3	15	1.4	15	4.2	48	-0.5

Test No.	Reaction Time	pH	C dil. factor	TC analyzed	TOC analyzed	TC, ppm	TOC, ppm	TIC, ppm	
TY1	3	9.96	11	2.989	1.138	32.89	12.52	20.36	
TY2	3	9.93	11	2.544	0.853	27.98	9.383	18.6	
TY3	7	9.99	11	4.714	0.901	51.85	9.911	41.94	
TY4	7	10.07	11	4.679	0.91	51.47	10.01	41.46	
TY5	14	10.02	11	5.12	1.208	56.32	13.29	43.03	
TY6	14	9.97	11	4.274	1.43	47.01	15.73	31.28	
TY7	30	10.24	11	5.303	1.849	58.33	20.34	37.99	
TY8	30	10.12	11	4.653	0.595	51.18	6.545	44.64	
TY9	70	10.32	11	5.928	1.167	65.27	12.84	52.37	
TY10	70	10.33	11	5.04	0.891	55.41	9.801	45.64	
TY11	140	10.17	11	6.75	1.584	74.25	17.42	56.83	
TY12	140	10.28	11	12.37	1.179	136.1	12.97	123.1	
TY13	280	10.39	11	6.219	1.257	68.41	13.83	54.58	
TY14	280	10.31	11	5.564	1.209	61.2	13.3	47.91	
TY15	560	10.11	11	4.867	1.105	53.54	12.16	41.38	
TY16	560	10.29	11	3.657	0.927	40.22	10.2	30.03	
TY17	980	9.95	1	63.54	6.444	63.54	6.444	57.1	
TY18	980	9.84	1	63.38	5.621	63.38	5.621	57.76	
Test No.	ACL Number	F, ppm	CH02-, ppm	Cl-, ppm	NO2-, ppm	NO3-, ppm	HP04=, ppm	S04=, ppm	C204=, ppm
TY1									
TY2									
TY3	93-2154-56	0.79	-0.3	1.3	0.16	1.3	3.4	10	-0.5
TY4	93-2154-57	1.6	-0.3	0.67	1.9	0.98	5	4.1	-0.5
TY5	93-2154-58	0.81	-0.3	0.83	0.3	-0.2	5.9	4.2	-0.5
TY6	92-2079-01	0.98		1.4	0.73	0.52	3.2	1.8	-0.3
TY7	93-2154-59	0.92	-0.3	2.6	-0.1	0.56	5.8	5.3	-0.5
TY8	92-2079-02	0.99		1.5	0.64	0.4	3.7	2	0.39
TY9	93-2154-60	1.5	-0.3	0.92	0.29	-0.2	8.3	6.2	-0.5
TY10	92-2079-03	1.2		1.2	0.27	0.35	4.7	1.6	0.46
TY11	93-2154-61	1.8	-0.3	1.8	2	2.4	9.7	5.7	-0.5
TY12	92-2079-04	1.1		3.6	1	0.79	4.8	2	0.87
TY13	93-2154-62	2	-0.3	0.7	0.74	0.7	10	4.9	-0.5
TY14	92-2079-05	1.7		0.96	0.46	0.3	6	1.6	0.9
TY15	93-2154-63	1.8	-0.3	1.8	0.28	2.4	9.7	5.7	-0.5
TY16	93-2154-64	2	-0.3	0.7	1.1	0.7	10	4.9	-0.5
TY17	94-2110-04	1.7	13	1	0.54	0.54	7.9	4.6	0.97
TY18	94-2110-05	2.2	12	0.52	0.31	0.61	7	2.2	1

Test No.	Reaction Time	pH	C dil. factor	TC analyzed	TOC analyzed	TC, ppm	TOC, ppm	TIC, ppm		
TZ1	3	11.09	11	3.901	2.027	42.91	22.297	20.613		
TZ2	3	11.14	11	2.959	1.02	32.55	11.22	21.33		
TZ3	7	10.93	11	4.933	2.297	54.26	25.267	28.993		
TZ4	7	11.21	11	4.003	1.341	44.03	14.751	29.279		
TZ5	14	11.34	11	4.117	1.88	45.29	20.68	24.61		
TZ6	14	11.33	11	3.149	1.374	34.64	15.114	19.526		
TZ7	28	11.49	11	10.87	4.632	119.6	50.952	68.648		
TZ8	28	11.46	11	8.288	4.162	91.17	45.782	45.388		
TZ9	56	11.46	11	4.583	2.382	50.41	26.202	24.208		
TZ10	56	11.5	11	4.783	2.462	52.61	27.082	25.528		
TZ11	91	11.43	11	5.871	2.772	64.58	30.492	34.088		
TZ12	91	11.5	11	5.537	2.593	60.91	28.523	32.387		
TZ13	182	11.62	11	10.93	3.668	120.2	40.348	79.852		
TZ14	182	11.69	11	8.352	2.557	91.87	28.127	63.743		
TZ15	364	11.72	11	6.233	1.281	68.56	14.091	54.469		
TZ16	364	11.76	11	6.507	0.982	71.58	10.802	60.778		
Test No.	ACL NUMBER	ACL F-, ppm	ACL, CH02-, pp	Cl-, ppm	N02-, ppm	N03-, ppm	HP04=, ppm	S04=, ppm	C204=, ppm	
TZ1	93-2154-38	1.4	-0.3	2.7	2.1	2.8	18	11	0.55	
TZ2	93-2154-39	1.9	-0.3	2	0.42	-0.2	16	7.3	1	
TZ3										
TZ4	93-2154-40	2.2	-0.3	2.1	1.5	-0.2	19	7.6	1.4	
TZ5	93-2154-41	2.4	-0.3	1.6	0.8	-0.2	24	8.5	2	
TZ6	93-2154-42	2.4	-0.3	2.2	1.1	0.82	26	9.9	2	
TZ7	93-2154-43	2.7	-0.3	2.8	0.37	0.42	34	11	1.5	
TZ8	92-2102-41		16	1.6	-0.2	0.5	21	4.3	1.3	
TZ9	93-2154-44	2.7	-0.3	2.2	0.5	0.55	39	12	2	
TZ10	92-2102-42	3.8		1.7	-0.2	0.45	26	5	1.4	
TZ11	92-2005-05	1.7	5.8	1.2	0.97	1.9	36	6.9	-0.7	
TZ12	92-2005-06	1.8	7.4	1.5	1.1	50	36	6.2	-0.7	
TZ13	93-2154-43	2.7	-0.3	3.6	0.88	-0.2	52	19	1.5	
TZ14	92-2103-43	5.4		2.5	-0.2	1	29	7.9	1.4	
TZ15	93-2154-44	22	-0.3	22	2.6	272	58	208	2	
TZ16	92-2103-44	-5	-15	-8	-15	7.4	62	138	-50	

Test No.	Reaction Time	pH	C dil. factor	TC analyzed	TOC analyzed	TC, ppm	TOC, ppm	TIC, ppm		
TZ31	3	11.02	11	4.73	0.966	52.03	10.63	41.4		
TZ32	3	11.04	11	5.075	1.168	55.83	12.85	42.98		
TZ33	7	11.03	11	5.439	1.573	59.83	17.3	42.53		
TZ34	7	11.01	11	5.609	1.183	61.7	13.01	48.69		
TZ35	14	11.23	11	5.807	1.582	63.88	17.4	46.48		
TZ36	14	11.28	11	5.561	1.417	61.17	15.59	45.58		
TZ37	28	11.37	11	10.33	2.88	113.6	31.68	81.95		
TZ38	28	11.38	11	12.13	4.392	133.4	48.31	85.12		
TZ39	56	11.37	11	6.492	2.355	71.41	25.91	45.51		
TZ40	56	11.41	11	6.417	1.715	70.59	18.87	51.72		
TZ41	91	11.4	11	7.188	2.644	79.07	29.08	49.98		
TZ42	91	11.38	11	7.708	2.422	84.79	26.64	58.15		
TZ43	182	11.63	11	10.66	2.418	117.3	26.6	90.66		
TZ44	182	11.66	11	10.72	2.135	117.9	23.49	94.44		
TZ45	364	11.09	11	7.955	1.257	87.51	13.83	73.68		
TZ46	364	11.75	11	7.552	0.716	83.07	7.876	75.2		
Test No.	ACL Number	ACL F-, ppm	ACL, CHO2-, pp	Cl-, ppm	NO2-, ppm	NO3-, ppm	HPO4=, ppm	SO4=, ppm	C2O4=, ppm	
TZ31	93-2154-47	6.1	-0.3	19	2.8	32	17	59	2.1	
TZ32	93-2154-48	5.3	-0.3	18	2.2	17	13	51	2	
TZ33	93-2154-49	5.2	-0.3	17	1.5	18	17	50	1.2	
TZ34	93-2154-50	6.3	-0.3	20	2.2	23	19	57	1.2	
TZ35	93-2154-51	5.4	-0.3	16	2.4	15	19	51	2.5	
TZ36	93-2154-52	5.7	-0.3	16	2	17	20	53	2	
TZ37	93-2154-53	5.5	-0.3	17	2.4	15	26	55	1.7	
TZ38	92-2103-45	5.6		12	-0.2	9.9	18	25	1.5	
TZ39	93-21549-54	5.8	-0.3	16	2.1	21	34	57	1.7	
TZ40	92-2103-46	5.8		11	-0.2	9.6	22	25	1.6	
TZ41	92-2005-07	2.7	9.2	8.5	1.1	9.8	28	2.5	0.82	
TZ42	92-2005-08	2.8	9.2	9.3	1.1	10	30	2.6	0.81	
TZ43	93-2154-55	5.1	-0.3	17	2.2	21	59	62	2.1	
TZ44	92-2103-47	7.4		10	-0.2	8.1	33	24	2.3	
TZ45										
TZ46	92-2103-48	11		33	-15	79	77	180	-50	

Table D.5. Actinide Results

<u>Column Heading</u>	<u>Comment</u>
SAMPLE (FRACTION)	Test number in test matrix (Table D.1). Identification of sample treatment.
UF	Unfiltered leachate.
F.45	Filtrate from 0.45 μm filtration.
F50	Filtrate from filtration through Amicon model CF50A filter.
AS	Solution from acid soak of reaction vessel.
ELEC	Sample prepared by electroplating
RXN TIME	Reaction time, in days.
SOL'N ml	Volume of solution into which actinides were dissolved, in milliliters.
ALIUOT ml	Volume of solution evaporated or electroplated onto planchet.
SPEC NO. (DET)	Spectrum number of alpha counting (detector number).
LIVE TIME	Time detector was active during analysis, in seconds.
EFFICIENCY	Measured efficiency of detector, in counts/disintegration.
Np bkgnd	Background counts per second in Np region.
Pu bkgnd	Background counts per second in Pu region.
Am bkgnd	Background counts per second in Am region.
Np COUNTS	Number of counts assigned to neptunium-237 peak.
Np ng	Mass of neptunium-237 calculated, in nanograms.
Pu COUNTS	Number of counts assigned to plutonium-239 peak.
Pu ng	Mass of plutonium-239 calculated, in nanograms.
Am COUNTS	Number of counts assigned to americium-241 peak.
Am pg	Mass of americium-241 calculated, in picograms.

SAMPLE	RXN TIME	SOL'N ml	ALIQVOT ml	SPEC NO.	LIVE TIME	EFFICIENCY	Np bkgnd	Pu bkgnd
BX1(UF)	7	16.62	0.05	A129(2)	80646	0.27	0.00004	0.00002
BX1(F50)		16.62	0.03	A149(1)	185048	0.22	0.00003	0
BX1(AS)		19	0.05	2038	11977	0.24	0	0
BX2(UF)	7	16.66	0.05	A230(4)	181587	0.15	0	0.00001
BX2(F50)		16.66	0.45	A231(1)	167072	0.21	0.00002	0.00001
BX2(AS)		19	0.05	2039	12144	0.24	0	0
BX3(UF)	28	17.42	0.05	A233(3)	151368	0.17	0	0.00002
BX3(F50)		17.42	0.5	A175(1)	101937	0.27	0.00038	0.00031
BX3(AS)		19	0.05	A330(4)	81720	0.24	0	0.00002
BX4(UF)	28	16.9	0.05	A130(3)	80646	0.21	0	0
BX4(F50)		16.9	0.45	A150(2)	185047	0.27	0.00038	0.00031
BX4(AS)		19	0.05	A232(2)	151368	0.18	0.00005	0.00001
BX5(UF)	91	17.41	0.05	A117(3)	175263	0.21	0	0
BX5(F50)		17.41	0.4	A101(1)	137887	0.2	0	0
BX5(AS)		19	0.05	A102(2)	137887	0.27	0.00004	0.00001
BX6(UF)	91	16.48	0.05	A116(2)	175250	0.27	0.00004	0.00002
BX6(F50)		16.48	0.35	A104(4)	137887	0.26	0.00006	0.00002
BX6(AS)		19	0.05	A103(3)	137887	0.23	0.00004	0.00004
BX7(UF)	238	16.4	0.05	A580(1)	248819	0.26	0.00007	0.00005
BX7(F50)		16.4	1	A581(2)	248819	0.27	0.00005	0.00003
BX7(AS)		19	0.05	A567(4)	93387	0.24	0.00003	0.00002
BX8(UF)	238	16.23	0.05	A582(3)	248818	0.24	0.00013	0.00015
BX8(F50)		16.23	0.05	A583(4)	248818	0.24	0.00003	0.00002
BX8(AS)		19	0.05	A566(3)	93388	0.24	0.00013	0.00015
BX9(UF)	600	16.67	0.05	A996(2)	343939	0.25	0.00031	0.00072
BX9(F50)		16.67	0.1	A999(3)	162908	0.29	0.00005	0.00004
BX9(AS)		19	0.05	A997(3)	180985	0.29	0.00005	0.00004
BX10(UF)	600	16.93	0.05	C001(4)	162908	0.28	0.00008	0.00007
BX10(F50)		16.93	0.1	A995(1)	343939	0.25	0.00004	0
BX10(AS)		19	0.05	A998(4)	180984	0.28	0.00008	0.00007
BX11(UF)	1400	17.06	0.1	C1006(1)	172800	0.28	0.00003	0.00017
BX11(F50)		17.06	0.1	C2006(2)	172800	0.26	0.00006	0
BX11(AS)		19	0.1	C3006(3)	172800	0.26	0.00005	0.00016
BX12(UF)	1400	16.44	0.1	C4006(4)	172800	0.27	0.00007	0.0001
BX12(F50)		16.44	0.1	C5006(5)	172800	0.23	0.0001	0.00023
BX12(AS)		19	0.1	C6006(6)	172800	0.24	0	0.00004

SAMPLE	Am bkgnd	Np COUNTS	Np ng	Pu COUNTS	Pu ng	Am COUNTS	Am pg
BX1(UF)	0.00002	0	-1.88606	0	-0.01074	0	-0.19378
BX1(F50)	0.00001	72	34.63276	56	0.33226	44	4.51409
BX1(AS)	0	0	0	0	0	44	45.58751
BX2(UF)	0	0	0	2	0.00098	1	0.09564
BX2(F50)	0	16	0.50439	2	0.00015	0	0
BX2(AS)	0	0	0	5	0.28305	11	11.24015
BX3(UF)	0.00006	4	2.1242	3	-0.00017	1	-0.88193
BX3(F50)	0.00035	51	0.60126	38	0.00357	29	-0.06727
BX3(AS)	0.00003	0	0	0	-0.01398	0	-0.37856
BX4(UF)	0	0	0	1	0.00854	0	0
BX4(F50)	0.00035	23	-1.36224	46	-0.00372	7	-0.34173
BX4(AS)	0.00002	5	-1.40313	11	0.059	22	2.1298
BX5(UF)	0	78	27.73436	345	1.39647	452	33.02462
BX5(F50)	0	6	0.36269	1	0.00069	0	0
BX5(AS)	0.00002	206	78.37474	552	2.45039	1231	98.66321
BX6(UF)	0.00002	10	0.79769	0	-0.01064	35	1.72656
BX6(F50)	0.00006	58	2.46349	41	0.02157	4	-0.0435
BX6(AS)	0.00002	23	8.12993	337	1.75462	309	29.26
BX7(UF)	0.00003	10	-1.43482	8	-0.00978	7	-0.01847
BX7(F50)	0.00002	24	0.10888	12	0.00049	16	0.02134
BX7(AS)	0.00002	5	1.45174	78	0.57232	151	20.23632
BX8(UF)	0.00018	14	-3.80363	84	0.11016	94	2.09653
BX8(F50)	0.00002	16	1.80709	14	0.02175	25	0.87111
BX8(AS)	0.00018	298	184.8532	1074	7.80305	1104	144.46269
BX9(UF)	0.00186	0	-15.83383	3505	5.50677	3706	93.56852
BX9(F50)	0.00007	0	-1.09326	0	-0.00996	0	-0.3145
BX9(AS)	0.00007	591	160.26665	2557	7.99363	4974	280.75732
BX10(UF)	0.00006	0	-3.66597	1278	4.05586	1284	73.65108
BX10(F50)	0.00008	13	-0.05667	26	0.02214	26	-0.02329
BX10(AS)	0.00006	404	110.68453	2568	8.26591	6945	404.87817
BX11(UF)	0.0011	4473	607.69342	12317	19.02588	6437	174.59474
BX11(F50)	0	19	1.27999	11	0.01857	233	7.09947
BX11(AS)	0.00035	1532	249.61923	8158	15.16605	18606	624.43964
BX12(UF)	0.0003	389	50.49155	6268	9.53248	4874	132.74112
BX12(F50)	0.00084	23	0.92224	53	0.02433	156	0.3594
BX12(AS)	0.00059	1100	195.45778	9817	19.84362	20390	740.75824

SAMPLE	RXN TIME	SOL'N ml	ALIQOT ml	SPEC NO.	LIVE TIME	EFFICIENCY	Np bkgnd	Pu bkgnd
BY1(UF)	3	10	0.05	A223(3)	152264	0.22	0.00004	0.00004
BY1(F.45)		10	0.05	A012(4)	141698	0.21	0.00006	0.00002
BY1(F50)		10	0.3	A119(1)	142557	0.22	0.00003	0
BY1(AS)		19	0.05	A018(1)	85968	0.22	0	0
BY2(UF)	3	9.96	0.05	2032	20715	0.24	0	0
BY2(F.45)		9.96	0.05	2033	20704	0.24	0	0
BY2(F50)		9.96	0.4	A125(2)	105740	0.27	0.00004	0.00002
BY2(AS)		19	0.05	A078(2)	86658	0.27	0.00004	0.00001
BY3(UF)	7	10	0.05	A087(3)	75119	0.22	0.00004	0.00004
BY3(F.45)		10	0.05	A074(2)	84724	0.27	0.00004	0.00001
BY3(F50)		10	0.3	A082(2)	101090	0.27	0.00004	0.00001
BY3(AS)		19	0.05	A077(1)	86554	0.22	0	0
BY4(UF)	7	9.99	0.05	A017(4)	74790	0.21	0.00006	0.00002
BY4(F.45)		9.99	0.05	A016(3)	74039	0.22	0.00004	0.00004
BY4(F50)		9.99	0.35	A067(3)	88157	0.22	0.00004	0.00004
BY4(AS)		19	0.05	A003(3)	170983	0.22	0.00004	0.00004
BY5(UF)	14	9.95	0.05	2006	20654	0.24	0	0
BY5(F.45)		9.95	0.05	2007	31640	0.24	0	0
BY5(F50)		9.95	0.15	2008	54050	0.24	0	0
BY5(AS)		19	0.05	2009	32223	0.24	0	0
BY6(UF)	14	10.05	0.05	A222(2)	11336	0.18	0.00005	0.00001
BY6(F.45)		10.05	0.05	A221(1)	11337	0.21	0.00002	0.00001
BY6(F50)		10.05	0.4	A081(1)	101090	0.22	0	0
BY6(AS)		19	0.05	A019(2)	85967	0.27	0.00004	0.00001
BY7(UF)	30	10.01	0.1	1989	57976	0.24	0	0
BY7(F.45)		10.01	0.1	1987	18958	0.24	0	0
BY7(F50)		10.01	0.36	1991	33967	0.24	0	0
BY7(AS)		19	0.05	1992	52860	0.24	0	0
BY8(UF)	30	10.09	0.1	1993	18713	0.24	0	0
BY8(F.45)		10.09	0.1	1994	13584	0.24	0	0
BY8(F50)		10.09	0.45	1995	51921	0.24	0	0
BY8(AS)		19	0.05	1996	52000	0.24	0	0
BY9(UF)	70	10.29	0.05	A255(1)	10571	0.21	0.00002	0.00001
BY9(F.45)		10.29	0.05	A256(2)	10571	0.18	0.00005	0.00001
BY9(F50)		10.29	0.35	A685(4)	95747	0.29	0.0001	0.00008
BY9(AS)		19	0.05	A267(3)	53409	0.17	0	0.0002
BY10(UF)	70	10.02	0.05	A257(3)	10570	0.17	0	0.0002
BY10(F.45)		10.02	0.05	A258(4)	10570	0.15	0.00003	0.00003
BY10(F50)		10.02	0.45	A684(2)	95747	0.26	0.00011	0.00021
BY10(AS)		19	0.05	A268(4)	53413	0.29	0.0001	0.00008
BY11(UF)	140	9.96	0.05	A491(3)	12011	0.26	0.00005	0
BY11(F.45)		9.96	0.05	A496(4)	73351	0.24	0	0
BY11(F50)		9.96	0.05	A404(3)	83805	0.26	0.00005	0
BY11(AS)		9.96	0.2	A395(2)	142724	0.27	0	0
BY12(UF)	140	9.96	0.2	A697(4)	15944	0.3	0.0001	0.00008
BY12(F.45)		19	0.1	A388(3)	95908	0.25	0.00005	0
BY12(F50)		10.22	0.05	A503(3)	82030	0.26	0.00005	0
BY12(AS)		10.22	0.05	A504(4)	82030	0.24	0	0
BY13(UF)	280	10.22	0.2	A397(4)	142732	0.25	0	0
BY13(F.45)		10.22	0.2	A696(3)	15944	0.23	0.0001	0.00004
BY13(F50)		19	0.1	A389(4)	95910	0.25	0	0
BY13(AS)		10.03	0.05	A515(3)	156177	0.26	0.00005	0
BY14(UF)	280	10.03	0.08	A0257(1)	172800	0.24	0.00002	0.00011
BY14(F.45)		10.03	0.05	A514(2)	156178	0.27	0	0
BY14(F50)		10.03	0.35	A519(3)	101485	0.26	0.00005	0
BY14(AS)		19	0.05	A517(1)	101485	0.25	0	0.00002
BY15(UF)	560	10	0.05	A516(4)	156177	0.24	0	0
BY15(F.45)		10	0.08	A0258(2)	172800	0.22	0.00004	0.00008
BY15(F50)		10	0.05	A513(1)	156178	0.25	0	0.00002
BY15(AS)		10	0.45	A520(4)	101484	0.24	0	0
BY16(UF)	560	19	0.1	A518(2)	101485	0.27	0	0
BY16(F.45)		9.99	0.05	A929(4)	251634	0.28	0.0001	0.00001
BY16(F50)		9.99	0.05	A930(1)	87972	0.28	0.00006	0.00002
BY16(AS)		9.99	0.05	A0259(3)	172800	0.25	0.00004	0.00029
BY17(UF)	980	9.99	0.15	A933(4)	87972	0.28	0.00004	0.00001
BY17(F.45)		19	0.05	A935(2)	79780	0.25	0.00026	0.00063
BY17(F50)		10.02	0.05	A934(1)	79780	0.28	0.00004	0.00003
BY17(AS)		10.02	0.05	A931(2)	87972	0.25	0.00026	0.00063
BY18(UF)	980	5	0.05	A0260(4)	172800	0.27	0.00007	0.00001
BY18(F.45)		10.02	0.1	A932(3)	87972	0.29	0.00006	0.00002
BY18(F50)		19	0.05	A936(3)	79780	0.29	0.00006	0.00002
BY18(AS)		10	0.5	C279(1)	69087	0.26	0.00008	0.00006
BY19(UF)	980	10	0.08	A0262(6)	172800	0.23	0.00002	0.00003
BY19(F.45)		10	0.05	C281(3)	158973	0.27	0.00078	0.00046
BY19(F50)		19	0.1	C280(2)	69087	0.22	0.00046	0.00055
BY19(AS)		10	0.5	C275(3)	243107	0.27	0.00078	0.00046
BY20(UF)	980	10	0.08	A0261(5)	172800	0.24	0.00017	0.00019
BY20(F.45)		10	0.05	C273(1)	246108	0.26	0.00008	0.00006
BY20(F50)		19	0.1	C274(2)	243108	0.22	0.00046	0.00055

SAMPLE	Am bkgnd	Np COUNTS	Np ng	Pu COUNTS	Pu ng	Am COUNTS	Am ng
BY1(UF)	0.00002	509	116.59008	3033	7.98838	4054	192.97682
BY1(F.45)	0.00006	533	136.31563	2711	8.01244	3831	204.1385
BY1(F50)	0.00001	960	38.90437	2228	1.03245	2898	24.2852
BY1(AS)	0	138	106.19448	841	7.36727	1349	213.30978
BY2(UF)	0	224	342.32147	1000	17.39698	1285	403.52014
BY2(F.45)	0	233	356.26465	1170	20.36528	1384	434.83936
BY2(F50)	0.00002	5078	169.48288	3787	1.43925	886	6.0669
BY2(AS)	0.00002	51	29.56728	624	4.41244	989	126.18869
BY3(UF)	0.00002	276	128.28453	2457	13.12746	3901	376.53369
BY3(F.45)	0.00002	158	51.77249	4014	15.2979	5828	400.89233
BY3(F50)	0.00002	1725	80.4962	2639	1.40464	3631	34.87915
BY3(AS)	0	79	60.38091	924	8.03956	1561	245.16101
BY4(UF)	0.00006	117	55.34629	4126	23.09653	6744	681.22693
BY4(F.45)	0.00002	887	421.0611	5124	27.76641	6968	681.81378
BY4(F50)	0.00002	1670	95.23096	2628	1.7073	2825	33.15148
BY4(AS)	0.00002	106	38.89638	1012	4.48841	1632	131.26678
BY5(UF)	0	325	497.63852	8805	153.47855	11633	3660.14502
BY5(F.45)	0	500	499.76785	3500	39.82484	5000	1026.93811
BY5(F50)	0	419	81.72066	1204	2.6732	1131	45.32694
BY5(AS)	0	0	0	1100	23.4682	1700	654.67267
BY6(UF)	0.00002	235	904.56848	1021	44.84227	1245	986.935
BY6(F.45)	0	200	636.87537	1200	43.54567	1676	1097.91077
BY6(F50)	0	2612	113.01788	1790	0.88169	2204	19.59573
BY6(AS)	0.00002	83	49.88716	1238	8.83067	1881	242.13324
BY7(UF)	0	0	0	16500	51.53938	25000	1409.55994
BY7(F.45)	0	646	542.07056	39000	372.54233	28776	4961.69043
BY7(F50)	0	769	100.04202	963	1.42617	1087	29.05773
BY7(AS)	0	49	55.98018	1763	22.92863	2161	507.30469
BY8(UF)	0	0	0	9703	94.65057	7211	1269.69946
BY8(F.45)	0	0	0	7526	101.134	5800	1406.85449
BY8(F50)	0	1347	92.44532	8690	6.7893	17795	250.95238
BY8(AS)	0	47	54.58331	1765	23.33428	2476	590.86542
BY9(UF)	0	303	1059.9563	1930	76.90768	2406	1730.69299
BY9(F.45)	0.00002	243	1027.24963	1842	88.83167	1413	1229.89636
BY9(F50)	0.00004	5447	221.26686	12667	5.86438	9239	77.22251
BY9(AS)	0.00006	34	55.81343	1339	24.82274	3271	1102.27673
BY10(UF)	0.00006	132	577.41406	2477	123.24133	2261	2031.73999
BY10(F.45)	0.00007	367	1763.33667	2312	126.54954	2639	2606.98267
BY10(F50)	0.0005	9612	331.47195	36328	14.26914	37272	264.06427
BY10(AS)	0.00004	15	9.1066	2575	27.59188	5741	1111.83252
BY11(UF)	0.00002	0	-1.45598	3352	92.51228	3829	1907.39978
BY11(F.45)	0	879	380.94275	17267	85.18742	25493	2270.21899
BY11(F50)	0.00002	854	295.28311	24996	98.87241	28506	2035.18359
BY11(AS)	0.00003	13696	682.95178	68486	38.87638	60819	623.1333
BY12(UF)	0.00004	0	-0.64656	11471	52.94799	8206	683.72668
BY12(F.45)	0.00002	0	-1.47906	9181	32.23592	20671	1309.9646
BY12(F50)	0.00002	1243	451.27512	19244	79.7974	25629	1918.16296
BY12(AS)	0	1531	608.79474	24074	108.9762	27728	2265.63647
BY13(UF)	0	13397	726.08148	72836	44.93778	67367	750.24133
BY13(F.45)	0.00003	0	-0.65897	12203	57.41133	9683	822.30017
BY13(F50)	0	0	0	6618	22.59344	17351	1069.22339
BY13(AS)	0.00002	1620	302.71075	43301	92.55437	54050	2085.24878
BY14(UF)	0.00002	5955	697.82617	107455	143.40198	162378	3911.82275
BY14(F.45)	0.00003	27475	5062.16455	51199	107.38602	59111	2237.73389
BY14(F50)	0.00002	3809	157.02202	6360	2.98864	8529	72.3268
BY14(AS)	0.00003	0	0	28008	182.88531	32791	3864.8335
BY15(UF)	0	0	0	43172	100.43621	56661	2379.36255
BY15(F.45)	0.00017	5666	715.92474	135002	194.40364	177220	4606.14551
BY15(F50)	0.00003	0	0	52560	117.37785	62709	2527.79736
BY15(AS)	0	4497	157.14491	2858	1.13691	3311	23.77457
BY16(UF)	0.00003	0	0	29257	89.44514	35612	1965.05383
BY16(F.45)	0.0001	0	-2.7136	97365	119.49621	133950	2967.65137
BY16(F50)	0.00009	0	-1.65755	38595	137.96501	41150	2654.80249
BY17(UF)	0.00027	2388	415.13773	80903	160.47227	114151	4087.84961
BY17(F.45)	0.00004	5264	540.88489	29182	34.14689	36970	781.02234
BY17(F50)	0.00185	0	-15.25823	13215	110.23934	17520	2625.86621
BY17(AS)	0.00005	0	-1.10835	18480	73.05604	34703	2476.37109
BY18(UF)	0.00185	0	-8.04671	34828	139.25954	45454	3274.09033
BY18(F.45)	0.00006	2109	169.62233	74730	68.79973	103213	1715.42297
BY18(F50)	0.00007	3465	516.86884	18420	31.32384	28036	860.47015
BY18(AS)	0.00007	0	-2.99055	13244	94.18029	17903	2297.58594
BY19(UF)	0.00011	0	0	0	0	0	0
BY19(F.45)	0.00006	2172	259.02945	68389	92.96471	87398	2144.38623
BY19(F50)	0.00058	960	149.74762	9419	19.05727	10703	390.55035
BY19(AS)	0.00167	0	-15.4259	26499	146.21445	30231	3003.75244
BY20(UF)	0.00058	0	0	0	0	0	0
BY20(F.45)	0.00068	2193	244.13025	66590	85.52636	86809	2010.75049
BY20(F50)	0.00011	2047	245.52197	11130	15.32414	11444	284.11563
BY20(AS)	0.00167	0	-15.4259	84172	131.96486	106275	3000.80786

SAMPLE	RXN TIME	SOL'N ml	ALICUOT ml	SPEC NO.	LIVE TIME	EFFICIENCY	Np bkgnd	Pu bkgnd
BZ1(UF)	3	4.99	0.05	A122(2)	9359	0.27	0.00004	0.00002
BZ1(F.45)		4.99	0.05	A121(1)	9359	0.22	0.00003	0
BZ1(F50)		4.99	0.45	2035	1818	0.24	0	0
BZ1(AS)		19	0.05	A020(3)	85967	0.22	0.00004	0.00004
BZ2(UF)	3	5.03	0.05	A011(3)	141699	0.22	0.00004	0.00004
BZ2(F.45)		5.03	0.05	A009(1)	141699	0.22	0	0
BZ2(F50)		5.03	0.1	2034	7107	0.24	0	0
BZ2(AS)		19	0.05	A021(4)	85967	0.21	0.00006	0.00002
BZ3(UF)	7	5	0.05	A015(2)	74479	0.27	0.00004	0.00001
BZ3(F.45)		5	0.05	A014(1)	74126	0.22	0	0
BZ3(F50)		5	0.25	A084(4)	101089	0.26	0.00006	0.00002
BZ3(AS)		19	0.05	A001(1)	170983	0.22	0	0
BZ4(UF)	7	5.01	0.05	A071(3)	169261	0.22	0.00004	0.00004
BZ4(F.45)		5.01	0.05	A695(2)	15945	0.26	0.0001	0.00007
BZ4(F50)		5.01	0.05	A075(3)	84723	0.22	0.00004	0.00004
BZ4(AS)		5.01	0.15	A083(3)	101090	0.22	0.00004	0.00004
BZ5(UF)	14	4.97	0.05	A002(2)	170983	0.27	0.00004	0.00001
BZ5(F.45)		4.97	0.05	2010	53303	0.24	0	0
BZ5(F50)		4.97	0.05	2011	11447	0.24	0	0
BZ5(AS)		19	0.05	2012	52882	0.24	0	0
BZ6(UF)	14	4.98	0.05	2013	30144	0.24	0	0
BZ6(F.45)		4.98	0.05	A179(3)	60524	0.24	0.00003	0.00004
BZ6(F50)		4.98	0.05	A694(1)	15945	0.27	0.00008	0.00002
BZ6(AS)		19	0.05	A227(1)	14517	0.21	0.00002	0.00001
BZ7(UF)	28	4.98	0.1	A229(3)	30220	0.24	0	0.00002
BZ7(F.45)		4.95	0.1	A228(2)	30220	0.26	0.00005	0.00001
BZ7(F50)		4.95	0.1	1998	3267	0.24	0	0
BZ7(AS)		19	0.25	A053(3)	34397	0.23	0.00004	0.00004
BZ8(UF)	28	5.02	0.1	2004	15801	0.24	0	0
BZ8(F.45)		5.02	0.1	2005	18741	0.24	0	0
BZ8(F50)		5.02	0.25	A274(2)	70045	0.26	0.00003	0
BZ8(AS)		19	0.05	A276(4)	70044	0.24	0	0
BZ9(UF)	56	4.99	0.05	A275(3)	70044	0.24	0.00003	0.00003
BZ9(F.45)		4.99	0.05	A273(1)	70045	0.26	0	0
BZ9(F50)		4.99	0.45	A154(2)	73032	0.27	0.00005	0.00001
BZ9(AS)		19	0.05	A155(3)	106198	0.24	0.00003	0.00004
BZ10(UF)	56	5.01	0.05	A159(3)	141542	0.24	0.00003	0.00004
BZ10(F.45)		5.01	0.05	A263(3)	15533	0.24	0	0.00002
BZ10(F50)		5.01	0.45	A153(1)	72942	0.27	0.0038	0.0031
BZ10(AS)		19	0.05	A148(4)	83528	0.26	0.00004	0.00002
BZ11(UF)	98	4.97	0.05	A160(4)	141609	0.26	0.00004	0.00002
BZ11(F.45)		4.97	0.05	A265(3)	16706	0.24	0.00003	0.00003
BZ11(F50)		4.97	0.05	A247(1)	8538	0.26	0	0
BZ11(AS)		19	0.4	A248(2)	169434	0.26	0.00003	0
BZ12(UF)	98	4.98	0.05	A254(4)	141149	0.24	0.00007	0
BZ12(F.45)		4.98	0.05	A252(4)	19766	0.24	0.00007	0
BZ12(F50)		4.98	0.45	A250(4)	8537	0.24	0.00007	0
BZ12(AS)		19	0.05	A249(3)	169434	0.24	0.00003	0.00003
BZ13(UF)	182	4.98	0.05	A253(1)	141138	0.26	0	0
BZ13(F.45)		4.98	0.05	A251(1)	19774	0.26	0	0
BZ13(F50)		4.98	0.1	A584(1)	174201	0.26	0	0
BZ13(AS)		19	0.05	A585(2)	174201	0.27	0.00005	0
BZ14(UF)	182	5	0.05	A683(2)	95747	0.26	0.00011	0.00021
BZ14(F.45)		5	0.05	A564(1)	93388	0.26	0.00007	0.00005
BZ14(F50)		5	0.05	A586(3)	174200	0.24	0	0
BZ14(AS)		19	0.1	A587(4)	174200	0.24	0.00003	0.00001
BZ15(UF)	364	5	0.05	A682(1)	95747	0.26	0.00003	0.00002
BZ15(F.45)		5	0.05	A565(2)	93388	0.27	0.00005	0
BZ15(F50)		5	0.05	A661(3)	163045	0.3	0.0001	0.00004
BZ15(AS)		19	0.05	A653(2)	87832	0.26	0.00011	0.00021
BZ16(UF)	364	5	0.05	A651(4)	87832	0.29	0.0001	0.00008
BZ16(F.45)		5	0.05	A654(1)	87832	0.26	0.00003	0.00002
BZ16(F50)		5	0.05	A652(3)	87832	0.3	0.0001	0.00004
BZ16(AS)		19	0.05	A650(4)	116822	0.29	0.00011	0.00021
BZ17(UF)	728	5.02	0.05	A649(3)	116823	0.3	0.00003	0.00002
BZ17(F.45)		5.02	0.05	C081(3)	82553	0.28	0.00007	0.00003
BZ17(F50)		5.02	0.05	C085(3)	173255	0.28	0.00007	0.00003
BZ17(AS)		19	0.05	C093(3)	90413	0.28	0.00007	0.00003
BZ18(UF)	728	5.02	0.05	C088(2)	256024	0.25	0.00027	0.00072
BZ18(F.45)		5.02	0.05	C094(4)	90413	0.28	0.00009	0.00012
BZ18(F50)		5.02	0.05	C098(4)	82734	0.28	0.00009	0.00012
BZ18(AS)		19	0.05	C091(1)	4854	0.26	0.00007	0.00001
BZ19(UF)	504	5.03	0.05	C090(4)	256023	0.28	0.00009	0.00012
BZ19(F.45)		5.03	0.05	C092(2)	90413	0.25	0.00027	0.00072
BZ19(F50)		5.03	0.05	C095(1)	82734	0.26	0.00007	0.00001
BZ19(AS)		19	0.05	C082(4)	82553	0.28	0.00009	0.00012
				C084(2)	173255	0.25	0.00027	0.00072

SAMPLE	Am bkgnd	Np COUNTS	Np ug	Pu COUNTS	Pu ng	Am COUNTS	Am ng
BZ1(UF)	0.00002	226	0.34129	3773	64.96643	4676	1.45335
BZ1(F.45)	0.00001	705	1.30826	4228	89.35109	5416	2.06597
BZ1(F50)	0	447	0.4351	1154	12.78719	1514	0.30282
BZ1(AS)	0.00002	81	0.05969	2063	18.04224	3131	0.49482
BZ2(UF)	0.00002	42	0.00449	68006	95.67667	73152	1.85777
BZ2(F.45)	0	394	0.0487	77585	109.16232	90218	2.29127
BZ2(F50)	0	793	0.89566	16000	205.72018	24000	5.57002
BZ2(AS)	0.00006	113	0.08694	18346	168.35255	35587	5.89435
BZ3(UF)	0.00002	0	-0.00057	56168	121.7789	71289	2.78991
BZ3(F.45)	0	1576	0.37014	73583	196.73006	77609	3.74536
BZ3(F50)	0.00006	3387	0.09853	3742	1.24082	3611	0.02159
BZ3(AS)	0	192	0.07429	6950	30.61112	11974	0.95197
BZ4(UF)	0.00002	5650	0.58159	127084	149.08789	153441	3.24934
BZ4(F.45)	0.00016	0	-0.00148	13742	144.80083	13253	2.52043
BZ4(F50)	0.00002	2831	0.58219	73604	172.50894	78648	3.32734
BZ4(AS)	0.00002	1419	0.08139	2725	1.78166	2924	0.03454
BZ5(UF)	0	20000	0.0672	3725	13.36227	6370	0.41243
BZ5(F.45)	0	4000	5.95185	56000	189.71339	56000	3.42441
BZ5(F50)	0	380	5.54297	11000	173.52522	11000	3.13221
BZ5(AS)	0	115	0.11399	1323	4.51766	1762	0.1086
BZ6(UF)	0.00004	7741	0.23135	771	17.65681	1360	0.56219
BZ6(F.45)	0.00004	2572	2.03242	62499	186.83714	63664	3.43536
BZ6(F50)	0	3148	2.27785	15605	157.40279	16989	3.09312
BZ6(AS)	0.00006	684	3.93873	16587	236.27278	18608	4.78451
BZ7(UF)	0.00002	66	0.17988	1821	5.44974	2242	0.12105
BZ7(F.45)	0.00002	160	0.11945	834	17.57968	1635	0.62208
BZ7(F50)	0.00002	1435	0.38687	7670	211.11867	7132	3.54349
BZ7(AS)	0	6400	0.34355	8231	22.45036	8897	0.43807
BZ8(UF)	0	82	1.27982	2400	5.46345	2900	0.11916
BZ8(F.45)	0.00003	0	0.26533	2800	103.13928	5400	3.59044
BZ8(F50)	0.00003	5105	-0.00022	139207	167.30186	156786	3.40118
BZ8(AS)	0.00007	3212	0.58387	64979	84.60208	76433	1.79629
BZ9(UF)	0	204	0.14685	21824	11.36477	29194	0.2744
BZ9(F.45)	0.00002	664	0.16303	5524	50.2543	11814	1.94001
BZ9(F50)	0.00004	1635	0.128	8201	18.0954	8730	0.34767
BZ9(AS)	0.00004	5455	0.24472	3198	5.45244	4114	0.12665
BZ10(UF)	0.00006	56	0.06815	7872	1.11957	8111	0.02082
BZ10(F.45)	0.0035	0	0.21863	805	35.76282	1838	1.47373
BZ10(F50)	0.00003	1445	-0.05401	20000	43.86364	280097	11.20504
BZ10(AS)	0.00003	5423	0.25476	5508	11.07672	6536	0.23724
BZ11(UF)	0.00007	81	0.06274	6757	0.89047	6078	0.01445
BZ11(F.45)	0	1936	0.29221	1017	42.00425	2301	1.71542
BZ11(F50)	0.00003	6982	0.16687	1587	1.56126	1596	0.02825
BZ11(AS)	0	106	0.09794	1982	0.31695	814	0.00235
BZ12(UF)	0	1619	0.32096	1645	57.45211	3080	1.94169
BZ12(F.45)	0.00007	6853	0.1514	825	0.8756	1322	0.02525
BZ12(F50)	0	0	0.07916	2164	0.28454	988	0.00234
BZ12(AS)	0.00001	0	0	2540	81.85325	4853	2.82293
BZ13(UF)	0	1206	0	328637	315.09265	434204	7.51454
BZ13(F.45)	0.00005	988	0.09711	1117	1.0313	1425	0.02375
BZ13(F50)	0.00003	587	0.07489	1982	1.71117	176	0.00202
BZ13(AS)	0.00004	16518	0.34793	8333	56.82816	17032	2.09743
BZ14(UF)	0	619	1.5132	59602	62.1567	41473	0.78056
BZ14(F.45)	0.00002	676	0.05623	1400	1.45819	1802	0.03392
BZ14(F50)	0	0	0.05178	221	0.19185	300	0.00471
BZ14(AS)	0.00003	28	-0.0027	6344	41.68486	1256	0.14897
BZ15(UF)	0.00005	1118	0.00308	3	-0.02949	0	-0.00151
BZ15(F.45)	0.00004	236	0.63379	18349	119.30685	1326	0.15527
BZ15(F50)	0.00002	61	0.03303	63	0.09843	25	0.00067
BZ15(AS)	0.00003	2659	0.00544	0	-0.03157	0	-0.00136
BZ16(UF)	0.00002	57	1.10275	18410	87.02	29499	2.51699
BZ16(F.45)	0.00005	37	0.00852	25	0.04265	7	0.0001
BZ16(F50)	0.00005	25	0.00197	13	0.00704	5	-0.00006
BZ16(AS)	0.0017	163	0.00284	8	0.00914	3	-0.00005
BZ17(UF)	0.00009	1608	0.02135	102196	264.05792	204873	9.5521
BZ17(F.45)	0.00009	19	0.24301	688	1.17089	221	0.00664
BZ17(F50)	0.00008	23	0.00192	5	-0.00931	2	-0.00019
BZ17(AS)	0.00009	3259	0.06904	11	0.37986	7	0.00414
BZ18(UF)	0.0017	89	0.65697	88125	203.60139	165547	6.90529
BZ18(F.45)	0.00008	11	0.01101	86	0.04056	186	0.00113
BZ18(F50)	0.00009	9	0.00093	3	0.00443	4	-0.0001
BZ18(AS)	0.0017	2562	0.00026	6	-0.00741	0	-0.00025
BZ19(UF)	0.0017	0	0.84515	15790	59.92138	39636	2.71633

SAMPLE	RXN TIME	SOL'N ml	ALIQOT ml	SPEC NO.	LIVE TIME	EFFICIENCY	Np bkgnd	Pu bkgnd
PX1(UF)	7	17.19	0.05	A217(1)	178877	0.26	0.00002	0.00001
PX1(F50)		17.19	0.45	A209(1)	139716	0.26	0.00002	0.00001
PX1(AS)		19	0.05	A220(4)	178877	0.24	0	0.00001
PX2(UF)	7	17.1	0.05	A219(3)	178877	0.24	0	0.00002
PX2(F50)		17.1	0.5	A218(2)	178877	0.26	0.00005	0.00001
PX2(AS)		19	0.05	A224(4)	152255	0.24	0	0.00001
PX3(UF)	28	16.78	0.05	A073(1)	84724	0.2	0	0
PX3(F50)		16.78	0.45	A068(4)	88156	0.26	0.00006	0.00002
PX3(AS)		19	0.05	A079(3)	86658	0.23	0.00004	0.00004
PX4(UF)	28	16.78	0.05	A076(4)	84723	0.26	0.00006	0.00002
PX4(F50)		16.78	0.45	A086(2)	75120	0.27	0.00004	0.00001
PX4(AS)		19	0.05	A060(2)	162374	0.27	0.00004	0.00001
PX5(UF)	91	16.79	0.05	A161(3)	171173	0.24	0.00003	0.00004
PX5(F50)		16.79	0.45	A165(3)	99484	0.24	0.00003	0.00004
PX5(AS)		19	0.05	A259(1)	274065	0.26	0	0
PX6(UF)	91	16.79	0.05	A162(4)	171162	0.26	0.00004	0.00002
PX6(F50)		16.79	0.45	A126(4)	105733	0.26	0.00004	0.00002
PX6(AS)		16.79	0.45	A166(4)	99484	0.26	0.00004	0.00002
PX7(UF)	238	16.51	0.05	A260(2)	274064	0.27	0.00003	0
PX7(F50)		16.51	0.1	A643(3)	242622	0.3	0.00003	0.00003
PX7(AS)		19	0.05	A645(1)	242622	0.27	0.00003	0.00002
PX8(UF)	238	17.17	0.05	A630(2)	184138	0.27	0.00011	0.00021
PX8(F50)		17.17	0.1	A646(4)	242622	0.29	0.0001	0.00008
PX8(AS)		19	0.05	A642(2)	242622	0.27	0.00011	0.00021
PX9(UF)	600	18.51	0.05	A647(1)	116823	0.27	0.00003	0.00002
PX9(F50)		18.51	0.05	C013(4)	344308	0.28	0.00008	0.00007
PX9(AS)		19	0.1	C014(4)	191249	0.28	0.00008	0.00007
PX10(UF)	600	17.11	0.05	C011(2)	153059	0.25	0.00034	0.00072
PX10(F50)		17.11	0.05	C015(1)	320854	0.25	0.00004	0
PX10(AS)		19	0.1	C012(3)	344308	0.29	0.00005	0.00004
				C010(1)	153059	0.25	0.00004	0

SAMPLE	Am bkgnd	Np COUNTS	Np ng	Pu COUNTS	Pu pg	Am COUNTS	Am pg
PX1(UF)	0	34	8.61333	0	-5.76526	0	0
PX1(F50)	0	149	5.88853	128	58.04621	92	0.76139
PX1(AS)	0	4	1.35605	38	139.74866	52	3.6224
PX2(UF)	0.00006	8	2.4409	0	-12.426	1	-0.61019
PX2(F50)	0.00002	81	2.0294	61	18.98405	63	0.34389
PX2(AS)	0	0	0	15	61.10757	29	2.37341
PX3(UF)	0	20	15.171	2	17.27038	0	0
PX3(F50)	0.00006	175	10.57456	6	3.00529	0	-0.06772
PX3(AS)	0.00002	3	-0.34051	16	104.18668	14	1.84058
PX4(UF)	0.00006	10	2.86888	6	28.5997	5	-0.01
PX4(F50)	0.00002	93	6.33692	7	5.00891	0	-0.02174
PX4(AS)	0.00002	8	0.49963	31	111.01619	35	2.16599
PX5(UF)	0.00004	107	31.89019	2	-17.27375	2	-0.3118
PX5(F50)	0.00004	450	26.75439	6	1.37673	3	-0.01204
PX5(AS)	0	15	3.0637	54	125.55585	91	3.8192
PX6(UF)	0.00003	59	15.0724	4	1.8975	0	-0.30493
PX6(F50)	0.00003	1	-0.16787	8	3.48267	9	0.06225
PX6(AS)	0.00003	174	9.39317	44	26.4213	1	-0.02253
PX7(UF)	0.00007	222	37.3078	70	156.73007	82	2.98174
PX7(F50)	0.00002	416	39.45292	11	7.36057	5	-0.42784
PX7(AS)	0.00005	19	-0.36744	20	16.64494	0	-0.09625
PX8(UF)	0.00004	39	2.75489	93	181.05508	101	5.52155
PX8(F50)	0.0005	55	2.8421	2	-37.04686	0	-0.37277
PX8(AS)	0.00002	756	347.21143	2	-55.93981	0	-2.50237
PX9(UF)	0.00006	133	15.50953	648	3391.44043	308	28.98076
PX9(F50)	0.00006	42	7.06953	19	-8.54124	343	9.74142
PX9(AS)	0.00186	2208	410.00974	3	-31.30933	19	0.40942
PX10(UF)	0.00008	1232	199.20219	773	1434.90149	16858	647.64752
PX10(F50)	0.00007	1440	186.75526	343	637.98895	290	8.87476
PX10(AS)	0.00008	768	144.89011	25	16.77689	25	0.02423
				14768	31971.49023	12693	495.53525

SAMPLE	RXN TIME	SOL'N ml	ALIQOT ml	SPEC NO.	LIVE TIME	EFFICIENCY	Np bkgnd	Pu bkgnd
PY1(UF)	3	10.23	0.05	2036	37249	0.24	0	0
PY1(F.45)		10.23	0.05	A176(2)	101936	0.27	0.00005	0.00001
PY1(F50)		10.23	0.2	A193(1)	170977	0.27	0.00038	0.00031
PY1(AS)		19	0.05	A181(1)	53998	0.27	0.00038	0.00031
PY2(UF)	3	10.56	0.05	2037	37246	0.24	0	0
PY2(F.45)		10.56	0.05	A128(1)	80646	0.22	0.00038	0.00031
PY2(F50)		10.56	0.3	A191(3)	75424	0.24	0.00003	0.00004
PY2(AS)		19	0.05	A182(2)	53997	0.27	0.00005	0.00001
PY3(UF)	7	9.88	0.05	A185(1)	96931	0.27	0.00038	0.00031
PY3(F.45)		9.88	0.05	A186(2)	96930	0.27	0.00005	0.00001
PY3(F.45)		9.88	0.05	A278(2)	675242	0.26	0.00003	0
PY3(F50)		9.88	0.35	A194(2)	170977	0.27	0.00005	0.00001
PY3(AS)		19	0.05	A183(3)	53997	0.24	0.00003	0.00004
PY4(UF)	7	9.85	0.05	A188(4)	96930	0.26	0.00004	0.00002
PY4(F.45)		9.85	0.05	A187(3)	96930	0.24	0.00003	0.00004
PY4(F50)		9.85	0.4	A195(3)	170977	0.24	0.00003	0.00004
PY4(AS)		19	0.05	A184(4)	53997	0.26	0.00004	0.00002
PY5(UF)	14	9.94	0.05	A008(4)	356107	0.26	0.00006	0.00002
PY5(F.45)		9.94	0.05	A163(1)	99485	0.27	0.00038	0.00031
PY5(F50)		9.94	0.5	A196(4)	170977	0.26	0.00004	0.00002
PY5(AS)		19	0.05	A189(1)	96931	0.27	0.00038	0.00031
PY6(UF)	14	10.04	0.05	A204(4)	84044	0.26	0.00004	0.00002
PY6(F.45)		10.04	0.05	A056(2)	173928	0.27	0.00004	0.00001
PY6(F50)		10.04	0.45	A126(3)	105733	0.24	0.00003	0.00004
PY6(AS)		19	0.05	A190(2)	75425	0.27	0.00005	0.00001
PY7(UF)	30	10.14	0.05	A178(4)	101935	0.26	0.00004	0.00002
PY7(F.45)		10.14	0.05	A072(4)	169261	0.26	0.00006	0.00002
PY7(F50)		10.14	0.45	A192(4)	75424	0.26	0.00004	0.00002
PY7(AS)		19	0.05	A180(3)	28698	0.24	0.00003	0.00004
PY8(UF)	30	10.04	0.05	A055(1)	173928	0.2	0	0
PY8(F.45)		10.04	0.05	A051(3)	86261	0.23	0.00004	0.00004
PY8(F50)		10.04	0.45	A280(4)	86772	0.24	0.00007	0
PY8(AS)		19	0.05	A054(4)	34396	0.26	0.00006	0.00002
PY9(UF)	70	9.92	0.05	A240(2)	76247	0.26	0.00005	0.00001
PY9(F.45)		9.92	0.05	A245(3)	66282	0.24	0.00003	0
PY9(F50)		9.92	0.4	A243(1)	66283	0.26	0.00007	0
PY9(AS)		9.92	0.4	A261(3)	72010	0.24	0.00003	0
PY10(UF)	70	19	0.05	A246(4)	66282	0.24	0.00003	0.00003
PY10(F.45)		10	0.05	A242(1)	76247	0.26	0.00007	0
PY10(F50)		10	0.05	A239(2)	76248	0.26	0.00005	0.00001
PY10(AS)		10	0.45	A244(4)	66283	0.24	0.00003	0.00003
PY11(UF)	140	10	0.05	A262(3)	72010	0.24	0.00003	0
PY11(F.45)		10	0.05	A241(2)	76247	0.26	0.00005	0.00001
PY11(F50)		10	0.05	A403(4)	83805	0.25	0	0
PY11(AS)		10	0.05	A496(4)	85472	0.25	0	0
PY12(UF)	140	10	0.05	A709(4)	233282	0.29	0.00001	0.00008
PY12(F.45)		10	0.2	A396(3)	142723	0.25	0.00005	0
PY12(F50)		10	0.1	A386(3)	80405	0.25	0.00005	0
PY12(AS)		10	0.05	A402(1)	83806	0.27	0	0.00002
PY13(UF)	280	10.09	0.05	A480(4)	86032	0.21	0.00002	0.00001
PY13(F.45)		10.09	0.05	A483(3)	103662	0.26	0.00009	0.00011
PY13(F50)		10.09	0.2	A394(1)	142742	0.27	0	0.00002
PY13(AS)		19	0.05	A387(4)	80408	0.25	0	0
PY14(UF)	280	10.09	0.05	A624(2)	183482	0.26	0.00006	0.00004
PY14(F.45)		10.09	0.1	A0251(1)	236433	0.26	0	0
PY14(F50)		10.09	0.5	A648(2)	116823	0.27	0.00011	0.00021
PY14(AS)		19	0.05	A613(1)	234720	0.26	0.00007	0.00001
PY15(UF)	560	10.09	0.05	A629(1)	184138	0.26	0.00007	0.00001
PY15(F.45)		10.09	0.05	A625(1)	253256	0.26	0.00007	0.00001
PY15(F50)		10.09	0.05	A0252(2)	236433	0.22	0	0
PY15(AS)		19	0.05	A623(1)	183482	0.26	0.00007	0.00001
PY16(UF)	560	10.09	0.05	A627(1)	83816	0.26	0.00007	0.00001
PY16(F.45)		10.09	0.05	A644(4)	84156	0.29	0.00001	0.00008
PY16(F50)		10.09	0.08	A973(1)	410106	0.27	0.00004	0.00003
PY16(AS)		19	0.05	A0253(3)	236433	0.25	0	0
PY17(UF)	980	10.4	0.05	A977(2)	167698	0.25	0.00026	0.00063
PY17(F.45)		10.4	0.05	A976(1)	167698	0.27	0.00004	0.00003
PY17(F50)		10.4	0.05	A972(4)	104297	0.28	0.00004	0.0001
PY17(AS)		19	0.03	A975(4)	410105	0.28	0.00004	0.0001
PY18(UF)	980	10.1	0.05	A0254(4)	236433	0.27	0	0
PY18(F.45)		10.1	0.05	A974(3)	410105	0.3	0.00006	0.00002
PY18(F50)		10.1	0.05	A978(3)	167697	0.3	0.00006	0.00002
PY18(AS)		19	0.08	A971(3)	104298	0.3	0.00006	0.00002
PY19(UF)		10.4	0.05	A0256(6)	236433	0.23	0	0
PY19(F.45)		10.4	0.05	C341(3)	269888	0.25	0	0
PY19(F50)		10.4	0.1	C343(1)	159105	0.25	0	0
PY19(AS)		19	0.08	C354(2)	104865	0.24	0	0
PY20(UF)		10.1	0.05	A0255(3)	236433	0.25	0	0
PY20(F.45)		10.1	0.05	C353(1)	104865	0.25	0	0
PY20(F50)		10.1	0.1	C342(4)	269888	0.24	0	0
PY20(AS)		19		C344(2)	159105	0.24	0	0

SAMPLE	Am bkgnd	Np COUNTS	Np ng	Pu COUNTS	Pu ng	Am COUNTS	Am pg
PY1(UF)	0	384	336.59662	453	4.52027	575	103.43563
PY1(F.45)	0.00002	667	188.45493	541	1.75016	616	35.87383
PY1(F50)	0.00035	1280	51.56197	691	0.30821	684	5.43577
PY1(AS)	0.00035	87	66.36466	433	4.73036	640	127.24089
PY2(UF)	0	514	465.1199	470	4.84158	580	107.7094
PY2(F.45)	0.00035	495	211.70779	408	1.9878	462	40.58583
PY2(F50)	0.00004	1004	74.60612	639	0.5392	780	11.87561
PY2(AS)	0.00002	96	93.13894	654	7.42601	1181	241.72708
PY3(UF)	0.00035	1009	281.12518	706	2.22517	824	46.88683
PY3(F.45)	0.00002	910	261.74954	876	2.88055	614	36.32307
PY3(F.45)	0.00003	4345	186.42888	7868	3.86105	5624	49.57411
PY3(F50)	0.00002	2624	61.25377	338	0.08966	234	1.10824
PY3(AS)	0.00004	146	162.14696	728	9.27962	675	155.07335
PY4(UF)	0.00003	1197	357.20587	1260	4.28769	1473	90.32372
PY4(F.45)	0.00004	1101	356.15112	1227	4.51599	1666	110.63243
PY4(F50)	0.00004	4542	104.27555	369	0.09476	202	0.92054
PY4(AS)	0.00003	0	-2.23908	1132	13.34624	1248	265.16345
PY5(UF)	0.00006	3320	271.26599	4682	4.37642	3885	65.20502
PY5(F.45)	0.00035	1022	278.98135	1516	4.79242	1014	56.96134
PY5(F50)	0.00003	7436	127.24592	3408	0.66383	1130	3.95394
PY5(AS)	0.00035	1020	546.74249	692	4.19054	824	90.16698
PY6(UF)	0.00003	1396	490.14066	1104	4.41649	730	52.5444
PY6(F.45)	0.00002	1591	259.41562	2362	4.40025	1008	33.76076
PY6(F50)	0.00004	1	-0.07314	8	0.00145	0	-0.02923
PY6(AS)	0.00002	111	76.63277	3400	27.65499	4784	701.42499
PY7(UF)	0.00003	949	276.92764	2153	7.17613	1094	65.61385
PY7(F.45)	0.00006	1027	179.46964	3799	7.62617	2194	79.101
PY7(F50)	0.00003	3219	141.53215	4475	2.24117	1131	10.19431
PY7(AS)	0.00004	8	15.08553	2356	56.64621	1833	794.38983
PY8(UF)	0	1537	339.81052	3533	8.89189	1242	56.35187
PY8(F.45)	0.00002	625	240.93279	1459	6.42295	555	44.01339
PY8(F50)	0	3458	141.64153	3645	1.70261	959	8.07554
PY8(AS)	0.00006	110	175.65817	2266	41.96786	1095	365.02255
PY9(UF)	0.00002	279	105.48077	4616	20.13845	2802	220.29256
PY9(F.45)	0.00003	721	343.45468	4033	21.93055	2204	215.86238
PY9(F50)	0	1989	109.36931	10207	6.40414	4146	46.89516
PY9(AS)	0.00003	3366	184.8765	8214	5.13912	5362	60.45358
PY10(UF)	0.00007	0	-1.81925	8019	83.49793	10025	1881.40649
PY10(F.45)	0.00002	650	249.09511	3540	15.57126	2428	192.53297
PY10(F50)	0.00007	779	299.52548	4165	18.31682	2913	230.86804
PY10(F50)	0.00003	2327	124.39359	8748	5.32685	3739	41.00274
PY10(AS)	0.00003	5363	264.00674	20014	11.22028	20801	210.20601
PY11(UF)	0.00002	0	-2.79885	5027	42.00655	5666	853.43359
PY11(UF)	0	1132	413.87125	3557	14.80439	2166	162.51785
PY11(UF)	0	1322	473.91049	3536	14.42995	2053	151.035
PY11(F.45)	0.00004	992	112.05692	13121	16.8883	5581	129.46686
PY11(F50)	0.00002	2569	137.49625	11146	6.80991	4559	50.18295
PY11(AS)	0.00002	323	115.47618	13224	54.49791	10908	810.27826
PY12(UF)	0.00003	1120	379.14722	5051	19.45856	2575	178.71732
PY12(UF)	0	1110	469.8927	4669	22.53103	1760	153.13904
PY12(F.45)	0.00009	1061	298.89282	7027	22.69803	3874	225.40944
PY12(F50)	0.00003	3518	174.80307	18444	10.43107	8402	85.63243
PY12(AS)	0	0	0	12083	99.58767	16182	2404.35767
PY13(UF)	0.00005	1119	179.51031	2631	4.83892	2607	86.37478
PY13(UF)ele	0	289	18.16797	1429	1.02265	2062	26.60241
PY13(F.45)	0.00005	1459	35.4357	2020	0.55662	1261	6.0474
PY13(F50)	0.00003	2735	344.29996	2865	4.12717	3252	84.33919
PY13(AS)	0.00003	0	-3.91838	26319	91.07356	16636	1037.51428
PY14(UF)	0.00003	1186	135.22679	2800	3.68614	3420	81.05929
PY14(UF)ele	0	231	33.848	727	1.21267	994	29.89037
PY14(F.45)	0.00003	572	8.1213	477	0.07856	367	1.0775
PY14(F50)	0.00003	692	239.97157	599	2.38154	351	25.01267
PY14(AS)	0.00004	0	-5.01862	20708	140.53482	12763	1561.56934
PY15(UF)	0.00005	287	11.80487	53	0.02021	25	0.04024
PY15(UF)ele	0	102	13.33743	95	0.14141	80	2.14677
PY15(F.45)	0.00185	188	26.62041	141	0.07419	333	0.86104
PY15(F50)	0.00005	97	15.41267	25	0.0388	9	0.02155
PY15(AS)	0.00004	0	-2.07914	19294	109.4026	16213	1657.78381
PY16(UF)	0.00004	1350	150.93726	1522	1.90815	991	22.63709
PY16(UF)ele	0	540	65.9627	1484	2.0636	1042	26.12138
PY16(F.45)	0.00007	318	18.59563	129	0.08716	101	0.94033
PY16(F50)	0.00007	79	10.68538	17	0.02408	16	0.13555
PY16(AS)	0.00007	0	-1.81925	26648	88.18272	26329	1570.37305
PY17(UF)elec	0	106	15.52859	92	0.15343	152	4.56976
PY17(F.45)	0	0	0	0	0	0	0
PY17(F50)	0	0	0	0	0	0	0
PY17(AS)	0	0	0	44369	182.55257	43521	3228.07227
PY18(UF)elec	0	531	69.50188	42000	62.5805	87588	2352.72095
PY18(F.45)	0	0	0	0	0	0	0
PY18(F50)	0	0	0	0	0	0	0
PY18(AS)	0	0	0	56265	48411	48411	

SAMPLE	RXN TIME	SOL 'N ml	ALIQOT ml	SPEC NO.	LIVE TIME	EFFICIENCY	Np bkgnd	Pu bkgnd
PZ1(UF)	3	5.05	0.05	A069(1)	169300	0.21	0	0
PZ1(F.45)		5.05	0.05	A024(1)	117091	0.21	0	0
PZ1(F50)		5.05	0.2	A211(3)	139716	0.24	0.00003	0.00004
PZ1(AS)		19	0.05	A200(4)	87992	0.26	0.00004	0.00002
PZ2(UF)	3	5.04	0.05	A070(2)	169261	0.27	0.00004	0.00001
PZ2(F.45)		5.04	0.05	A026(2)	117091	0.27	0.00004	0.00001
PZ2(F50)		5.04	0.25	A212(4)	139716	0.26	0.00004	0.00002
PZ2(AS)		19	0.05	A199(3)	87992	0.24	0.00003	0.00004
PZ3(UF)	7	5.02	0.05	A205(1)	31919	0.27	0.00038	0.00031
PZ3(F.45)		5.02	0.05	A206(2)	31918	0.27	0.00005	0.00001
PZ3(F.45)e1e		5.02		A0304(4)	172800	0.27	0	0
PZ3(AS)		19	0.05	A198(2)	87992	0.27	0.00005	0.00001
PZ4(UF)	7	5.05	0.05	A208(4)	31918	0.26	0.00004	0.00002
PZ4(F.45)		5.05	0.05	A207(3)	31918	0.24	0.00003	0.00004
PZ4(AS)		19	0.05	A197(1)	87993	0.27	0.00038	0.00031
PZ5(UF)	14	5	0.05	A235(2)	279392	0.26	0.00005	0.00001
PZ5(F.45)		5	0.05	A234(1)	204537	0.26	0.00007	0
PZ5(F50)		5	0.15	A005(1)	356108	0.2	0	0
PZ5(AS)		19	0.05	A201(1)	84045	0.27	0.00038	0.00031
PZ6(UF)	14	5.09	0.05	A131(4)	80645	0.26	0.00004	0.00002
PZ6(F.45)		5.09	0.05	A124(1)	105747	0.27	0.00038	0.00031
PZ6(F50)		5.09	0.25	A006(2)	356108	0.27	0.00004	0.00001
PZ6(AS)		19	0.05	A202(2)	84044	0.27	0.00005	0.00001
PZ7(UF)	28	5.01	0.05	A088(4)	75119	0.26	0.00006	0.00002
PZ7(F.45)		5.01	0.05	A118(4)	32685	0.26	0.00006	0.00002
PZ7(F.45)e1e		5.01	0.008	A0306(6)	172800	0.24	0	0
PZ7(F50)		5.01	0.4	A085(1)	75120	0.2	0	0
PZ7(AS)		19	0.05	A080(4)	86657	0.26	0.00006	0.00002
PZ7(AS)		19	0.05	A279(3)	86772	0.24	0.00003	0
PZ8(UF)	28	4.99	0.05	A237(4)	279391	0.24	0.00003	0.00003
PZ8(F.45)		4.99	0.05	A236(3)	279392	0.24	0.00003	0
PZ8(F50)		4.99	0.15	A210(2)	139716	0.27	0.00005	0.00001
PZ8(AS)		19	0.05	A203(3)	34044	0.24	0.00003	0.00004
PZ9(UF)	56	5.03	0.05	A156(4)	106198	0.26	0.00004	0.00002
PZ9(F.45)		5.03	0.05	A157(1)	204502	0.27	0.00038	0.00031
PZ9(F.45)		5.03	0.05	A164(2)	99484	0.27	0.00005	0.00001
PZ9(F.45)e1e		5.03	0.033	A0268(6)	172800	0.23	0	0
PZ9(F50)		5.03	0.4	A151(3)	185047	0.24	0.00003	0.00004
PZ9(AS)		19	0.05	A264(4)	15539	0.24	0.00003	0.00003
PZ9(AS)		19	0.05	A686(1)	241271	0.26	0.00003	0.00002
PZ10(UF)	56	5.03	0.05	A158(2)	204484	0.27	0.00005	0.00001
PZ10(F.45)		5.03	0.05	A277(1)	675243	0.26	0.00007	0
PZ10(F50)		5.03	0.45	A152(4)	185047	0.26	0.00004	0.00002
PZ10(AS)		19	0.05	A266(4)	16713	0.24	0.00003	0.00003
PZ11(F.45)	98	4.99	0.05	A272(4)	83809	0.24	0.00003	0.00003
PZ11(F.45)e1e		4.99	0.031	A0263(1)	172800	0.24	0	0
PZ11(F50)		4.99	0.4	A271(3)	83806	0.24	0.00003	0
PZ11(AS)		19	0.05	A270(4)	32632	0.24	0.00003	0.00003
PZ12(F.45)	98	4.99	0.05	A281(3)	588475	0.24	0.00003	0
PZ12(F50)		4.99	0.4	A282(4)	588480	0.24	0.00003	0.00003
PZ12(AS)		19	0.05	A269(3)	32628	0.24	0.00003	0.00003
PZ13(UF)	182	4.98	0.05	A343(4)	144140	0.24	0	0
PZ13(F.45)		4.98	0.05	A334(3)	143189	0.25	0.00005	0
PZ13(F.45)e1e		4.98	0.032	A0263(2)	172800	0.22	0	0
PZ13(F50)		4.98	0.05	A342(3)	144139	0.25	0.00005	0
PZ13(AS)		19	0.1	A332(1)	143190	0.27	0	0.00002
PZ14(UF)	182	5.01	0.05	A335(4)	143188	0.24	0	0
PZ14(F.45)		5.01	0.05	A340(1)	144141	0.27	0	0.00002
PZ14(F50)		5.01	0.05	A341(2)	144140	0.27	0	0
PZ14(AS)		19	0.1	A333(2)	143189	0.27	0	0
PZ15(UF)	364	5.02	0.05	A702(1)	173930	0.26	0.00008	0.00002
PZ15(F.45)		5.02	0.05	A703(2)	173929	0.26	0.0001	0.00007
PZ15(F.45)e1e		5.02	0.033	A0264(3)	172800	0.25	0	0
PZ15(F50)		5.02	0.1	A704(3)	173929	0.3	0.00004	0.00001
PZ16(UF)	364	5.02	0.05	A708(3)	233283	0.3	0.00004	0.00001
PZ16(F.45)		5.02	0.05	A706(4)	233284	0.29	0.00006	0.00009
PZ16(F50)		5.02	0.05	A707(1)	233283	0.26	0.00008	0.00002
PZ16(AS)		19	0.1	A710(2)	23035	0.26	0.0001	0.00007
PZ17(UF)	728	5.05	0.05	C122(4)	250872	0.28	0.00009	0.00012
PZ17(F.45)	728	5.05	0.05	C118(4)	111926	0.28	0.00009	0.00012
PZ17(F.45)e1e		5.05	0.033	A0265(3)	172800	0.25	0	0
PZ17(F50)		5.05	0.05	C117(3)	111926	0.3	0.00007	0.00003
PZ17(AS)		19	0.1	C120(2)	250873	0.24	0.00027	0.00072
PZ18(UF)	728	5.09	0.05	C119(1)	250873	0.26	0.00007	0.00001
PZ18(F.45)	728	5.09	0.05	C126(4)	93358	0.28	0.00009	0.00012
PZ18(F50)		5.09	0.05	C121(3)	250872	0.3	0.00007	0.00003
PZ18(AS)		19	0.1	C124(2)	93358	0.24	0.00027	0.00072
PZ19(UF)			0.05	C125(3)	93358	0.3	0.00007	0.00003
PZ19(F.45)	552	4.99	0.05	C123(1)	93358	0.26	0.00007	0.00001
PZ19(F.45)e1e		4.99	0.033	A0267(5)	172800	0.24	0	0
PZ19(F50)		4.99	0.05	C115(1)	111927	0.26	0.00007	0.00001
PZ19(AS)		19	0.1	C116(2)	111926	0.24	0.00027	0.00072
PZ20(F.45)e1e	1000	5	0.032	A0274(6)	259200	0.23	0	0

SAMPLE	Am bkgnd	Np COUNTS	Np ng	Pu COUNTS	Pu ng	Am COUNTS	Am pg
PZ1(UF)	0						
PZ1(F.45)	0	34	5.3488	3	0.00537	0	0
PZ1(F50)	0.00004	130	3.62838	19	0.0044	6	0.00244
PZ1(AS)	0.00003	116	71.5554	1337	9.6697	2194	286.45444
PZ2(UF)	0.00002						
PZ2(F.45)	0.00002	44	4.80115	3	0.00254	0	-0.05876
PZ2(F50)	0.00003	110	2.21931	7	0.00102	8	0.01663
PZ2(AS)	0.00004	148	100.17828	1333	10.43032	2147	303.54523
PZ3(UF)	0.00035	56	19.57468	135	0.63545	68	5.21027
PZ3(F.45)	0.00002	0	-0.7121	0	-0.00162	0	-0.05853
PZ3(F.45)ele	0	83		25		29	
PZ3(AS)	0.00002	0	-2.69518	4598	32.05893	7596	955.95111
PZ4(UF)	0.00003						
PZ4(F.45)	0.00004	6	2.54634	0	-0.00734	0	-0.13248
PZ4(AS)	0.00035	205	105.0977	3059	21.14207	6105	764.60242
PZ5(UF)	0.00002	367	18.61331	61	0.03494	33	0.29698
PZ5(F.45)	0	44	2.13773	0	0	5	0.07399
PZ5(F50)	0	136	2.43784	16	0.00326	8	0.02947
PZ5(AS)	0.00035	171	89.19037	3201	23.18093	5468	716.75134
PZ6(UF)	0.00003	24	3.86296	2	0.00082	1	-0.05423
PZ6(F.45)	0.00035	942	123.14952	406	0.58018	247	5.89232
PZ6(F50)	0.00002	241	1.83903	22	0.0017	0	-0.01187
PZ6(AS)	0.00002	223	140.33139	2023	14.76437	3632	478.44568
PZ7(UF)	0.00006	231	44.50388	103	0.22703	33	1.15042
PZ7(F.45)	0.00006	3	0.46916	0	-0.00336	0	-0.18198
PZ7(F.45)ele	0	121	69.98048	52	0.34236	358	42.5452
PZ7(F50)	0	31	0.98981	4	0.00145	0	0
PZ7(AS)	0.00006	48	27.64745	2936	21.57708	6291	834.33783
PZ7(AS)	0.00003	0	-1.81925	2872	22.84881	6113	877.47931
PZ8(UF)	0.00007						
PZ8(F.45)	0.00003	78	3.96851	6	0.00389	0	-0.09818
PZ8(F50)	0.00002	44	1.25016	3	0.00062	0	-0.01939
PZ8(AS)	0.00004	143	252.90282	2582	52.32935	3700	1353.78088
PZ9(UF)	0.00003	779	108.11098	318	0.50178	69	1.88713
PZ9(F.45)	0.00035	139	4.27681	78	0.0116	47	-0.35238
PZ9(F.45)	0.00002	43	5.45454	2	0.00164	0	-0.05865
PZ9(F.45)ele	0	88	12.92599	27	0.04515	35	1.05639
PZ9(F50)	0.00004	159	1.66409	13	0.00069	2	-0.01204
PZ9(AS)	0.00007	108	419.65573	1391	61.77565	2818	2258.90186
PZ9(AS)	0.00002	0	-1.67931	24085	63.59914	50730	2418.26221
PZ10(UF)	0.00002						
PZ10(F.45)	0	3148	68.04993	16587	4.14399	18608	83.91489
PZ10(F50)	0.00003	255	2.20316	27	0.00236	4	-0.00284
PZ10(AS)	0.00007	69	248.54124	857	35.37782	1762	1312.83496
PZ11(F.45)	0.00007	13	1.99263	0	-0.00544	0	-0.22908
PZ11(F.45)el	0	261	38.79923	92	0.15569	66	2.01606
PZ11(F50)	0.00003	40	0.89047	9	0.00243	0	-0.01227
PZ11(AS)	0.00007	198	366.13397	7163	151.51329	11291	4310.70117
PZ12(F.45)	0.00003	79	1.66025	11	0.00339	2	-0.08706
PZ12(F50)	0.00007	272	0.86044	36	0.00071	18	-0.01612
PZ12(AS)	0.00007	0	-1.81925	4294	90.83044	7676	2930.63916
PZ13(UF)	0	446	49.18098	41	0.05147	33	0.74774
PZ13(F.45)	0.00002	86	8.40152	28	0.03397	0	-0.06271
PZ11(F.45)el	0	17330	2717.12891	15308	27.32234	8198	264.11652
PZ13(F50)	0.00002	79	7.60009	22	0.02651	2	-0.0192
PZ13(AS)	0.00003	616	115.94622	10736	22.99801	19912	769.96863
PZ14(UF)	0	357	39.8673	86	0.10933	54	1.23913
PZ14(F.45)	0.00003	68	6.70539	8	0.00574	0	-0.08762
PZ14(F50)	0.00003	79	7.79014	13	0.01459	0	-0.08762
PZ14(AS)	0.00003	126	23.71644	14721	31.54307	28484	1101.51489
PZ15(UF)	0.00004	562	46.60506	158	0.14958	86	1.38109
PZ15(F.45)	0.00016	128	9.40525	60	0.04629	23	-0.08437
PZ13(F.45)el	0	123	16.58861	903	1.38637	754	20.89547
PZ15(F50)	0.00002	193	6.85523	22	0.0085	0	-0.02634
PZ16(UF)	0.00002	336	17.94881	73	0.0442	14	0.10539
PZ16(F.45)	0.00004	46	1.81903	11	-0.00647	27	0.20636
PZ16(F50)	0.00004	43	1.54294	16	0.00818	20	0.13898
PZ16(AS)	0.00016	313	377.50888	1730	23.9067	3091	770.81049
PZ17(UF)	0.00009	0	-1.24338	0	-0.01887	0	-0.25549
PZ17(F.45)	0.00009	0	-1.24338	0	-0.01887	0	-0.25549
PZ17(F.45)el	0	98	13.29593	950	1.46725	234	6.52355
PZ17(F50)	0.00005	0	-0.9026	0	-0.0044	0	-0.13248
PZ17(AS)	0.0017	2741	323.09415	73103	100.33113	114445	2831.64331
PZ18(UF)	0.00008	0		0		0	
PZ18(F.45)	0.00009	0	-1.25323	0	-0.01902	0	-0.25752
PZ18(F50)	0.00005	0	-0.90975	0	-0.00444	0	-0.13353
PZ18(AS)	0.0017	2558	822.6012	12552	46.1592	21655	1434.59424
PZ19(UF)	0.00005	0		0		0	
PZ19(F.45)	0.00008	0	-1.02909	0	-0.00167	0	-0.24167
PZ19(F.45)el	0	33	4.60834	71	0.11287	157	4.50512
PZ19(F50)	0.00008	0	-1.02909	0	-0.00167	0	-0.24167
PZ19(AS)	0.0017	1068	281.13535	26622	81.85061	40295	2232.44946
PZ20(F.45)el	0	27	2.71031	53	0.06056	57	1.17573

Table D.6. Spreadsheet Calculations Performed in Tables D.2 through D.5

Table D.2. Test Matrices:

Dilution factors (DF) are calculated for the unfiltered leachates (UF), the filtrates from the 0.45 μm filtered solutions (F.45), and the filtrates from the CF50A filtered solutions (F50). The dilution factors are calculated from the mass of the empty solution bottle (LPE), the mass of the solution bottle plus the leachate solution (+SOLN), and the mass of deionized water and concentrated nitric acid added (+DIW+HNO₃) to dilute the leachate as:

$$DF = \frac{((+SOLN) - (LPE) + (+DIW + HNO_3))}{((+SOLN) - (LPE))}$$

For example, the dilution factor for the unfiltered leachate of test BX1 is:

$$(UF \text{ LPE}) = 24.27 \text{ g}; (+UF \text{ SOLN}) = 38.78 \text{ g}; (+DIW+HNO_3) = 10.97 \text{ g}$$

$$DF = (38.78 - 24.27 + 10.07) / (38.78 - 24.27) = 1.6940$$

Table D.3. Cation Results:

The analytical results of the diluted solutions (given in the "I, ACL" columns) are corrected for the dilution by multiplying them by the dilution factor. The dilution-corrected concentrations are given in the "I, ppm" columns for each element.

For example, the dilution-corrected aluminum concentration for test BX1 is:

$$DF = 1.694; Al, ACL = 0.54 \text{ ppm}$$

$$Al, \text{ ppm} = (1.694) * (0.54) = 0.9148 \text{ ppm}$$

The normalized mass loss is calculated from the dilution-corrected concentration of the element of interest, the background concentration of the element of interest, the SA/V of the test, and the weight fraction of the element of interest. The background concentration and the weight fraction of each element, $f(i)$, are given in the tables.

For example, the normalized mass loss based on aluminum, NL(Al), in test BX1 is:

$$Al, \text{ ppm} = 0.9148 \text{ ppm}, Al \text{ background} = 0.56 \text{ ppm}, SA/V = 10.01 \text{ m}^{-1}, f(Al) = 0.0203$$

$$NL(Al) = (0.9148 - 0.56) / (10.01 * 0.0203) = 1.75 \text{ g/m}^2.$$

Table D.4. Anion Results:

The dilution factors are calculated using the leachate volume and the volume of deionized water used to dilute the leachates prior to analysis for carbon analysis (data not included). Solutions analyzed for other anions were not diluted.

The total carbon (TC) and total organic carbon (TOC) results were corrected for dilution by multiplying by the dilution factor (C dil factor). The dilution-corrected results are given in the TC and TOC columns. The total inorganic carbon (TIC) concentration was calculated as:

$$\text{TIC} = \text{TC} - \text{TOC}$$

Table D.5. Actinide Results:

The masses of neptunium, plutonium, and americium measured to be in the various fractions were determined from the number of counts under each peak in the alpha spectra. The efficiencies of the detectors and the number of background counts were determined using standards and blanks. The mass of each actinide is calculated as follows:

$$\text{An, } \mu\text{g} = (\text{An counts/LT} - \text{An bkgnd}) * (1/\text{EFF}) * (\text{SOLN ml/ALQ ml}) * \text{AN}$$

where (LT = LIVE TIME, ALQ = ALIQUOT)

and AN is 3.83×10^{-2} mg•s/dissociation for Np-237

4.36×10^{-4} mg•s/dissociation for Pu-239

7.87×10^{-6} mg•s/dissociation for Am-241

For example, the mass of plutonium in the F50 fraction of test BX1 is:

Pu = 56 counts

LIVE TIME = 185048 seconds

Pu bkgnd = 0 counts for detector 1

EFFICIENCY = 0.22 counts/disintegrations for detector 1

SOL'N ml = 16.62 ml

ALIQUOT ml = 0.03 ml

$$(56/185048 - 0) * (1/0.22) * (16.62/0.03) * 4.36 \times 10^{-4} = 3.32 \times 10^{-4} \mu\text{g}$$

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