

BSC

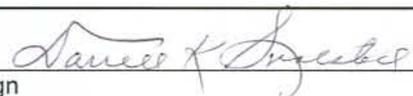
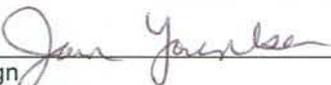
Model Administrative Change Notice

QA: QA

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Complete only applicable items.

1. Document Number:	ANL-EBS-MD-000016	2. Revision:	02	3. ACN:	01
4. Title:	Defense HLW Glass Degradation Model				
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6. Approvals:		
Preparer:	<u>Emma Thomas</u>  Print name and sign	<u>9/15/05</u> Date
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7. Affected Pages	8. Description of Change:
5-1	Citation update (Correct DIRS as appropriate) Section 5.2 "A pH VALUE FOR CONDENSED WATER ON GLASS OF 10 IS USED ABOVE 100°C", 2 nd paragraph, 1 st sentence, change: BSC 2004 [DIRS 167621] To BSC 2005 [DIRS 174583] This change is associated with CR-4185
6-5	Citation update (Correct DIRS as appropriate) Section 6.3.1 "Mechanism for Glass Dissolution", 1 st paragraph on page 6-5, last line, change: BSC 2004 [DIRS 167621] To BSC 2005 [DIRS 174583] This change is associated with CR-4185

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4. Title:	Defense HLW Glass Degradation Model				
6-5	<p>Citation update</p> <p><i>Footnote² on page 6-5, 1st line, change:</i></p> <p>BSC 2004 [DIRS 167621] To <i>BSC 2005 [DIRS 174583]</i></p> <p>This change is associated with CR-4185</p>				
6-8	<p>Citation update (Correct DIRS as appropriate)</p> <p>Section 6.3.3 “Uncertainties in the Subsystem and Environment”, 1st paragraph, line 8, change:</p> <p>BSC 2004 [DIRS 167621] To <i>BSC 2005 [DIRS 174583]</i></p> <p>This change is associated with CR-4185</p>				
6-22	<p>Citation update (Correct DIRS as appropriate)</p> <p>Section 6.5.2.3 “Effects of the Affinity Term on the Glass Degradation Rate”, last paragraph on page 6-22, line 5, change:</p> <p>BSC 2004 [DIRS 167621] To <i>BSC 2005 [DIRS 174583], Table 8-2</i></p> <p>This change is associated with CR-4185</p>				
6-59	<p>Citation update (Correct DIRS as appropriate)</p> <p>Section 6.8.1 “Uncertainty in Surface Area of Glass Contacted by Water”, 2nd paragraph on page 6-59, line 10, change:</p> <p>BSC 2004 [DIRS 167621] To <i>BSC 2005 [DIRS 174583]</i></p> <p>This change is associated with CR-4185</p>				

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1. Document Number:	ANL-EBS-MD-000016	2. Revision:	02	3. ACN:	01
4. Title:	Defense HLW Glass Degradation Model				
6-61	<p>Citation update (Correct DIRS as appropriate)</p> <p>Section 6.8.5 "Impacts of Uncertainty on Scientific Analysis Output", 1st paragraph, line 3, change:</p> <p>BSC 2004 [DIRS 167621] To <i>BSC 2005 [DIRS 174583]</i></p> <p>This change is associated with CR-4185</p>				
9-3	<p>Citation update</p> <p>Section 9.1 "DOCUMENTS CITED", add new citation 174583 on page 9-3, add:</p> <p><i>"BSC 2005. In-Package Chemistry Abstraction. ANL-EBS-MD-000037 REV 04.Las Vegas, Nevada: Bechtel SAIC Company. ACC: DOC.20050714.0003."</i></p> <p>This change is associated with CR-4185</p>				

5. ASSUMPTIONS

Several assumptions were made in the development of this model.

5.1 WATER HAS ACCESS TO SOME OF THE CRACK SURFACES

Assumption: It is assumed that water has free access to some of the crack surfaces of the glass and that the reaction rate in the cracks is less than for a free glass surface.

Rationale: As a glass cools in the canister, the combination of thermal and hoop stresses causes the glass to crack. The assumption is made in this model that not all the crack surfaces are accessible by water. While the data with artificial cracks suggest that the water access is limited (Perez and Westsik 1981 [DIRS 111044]), data from basaltic glasses at the ocean trenches suggests that the surfaces of bubbles in the glass, including the cracks that connect them to the surface, are accessed by water. Reaction rates within the cracks were lower than those at a free surface. Sené et al. (1999 [DIRS 163283]) established that the expected surface area contacted by water is less than the calculated surface area including the contribution from cracks. The reaction rate between water in the cracks and glass is reduced relative to the free surface because $[H_4SiO_4]$ is higher in the crack due to the low transport of dissolved material from the crack. Thus, in the model, the high exposed surface area factor assumes free access of water to all cracks and the low value assumes 50% of the crack surfaces are contacted with a reaction rate that is 50% of the rate at the free surface of the glass.

5.2 A pH VALUE FOR CONDENSED WATER ON GLASS OF 10 IS USED ABOVE 100°C

Assumption: It is assumed that condensed water on the surface of glass will have a pH of 10 for temperatures above 100°C.

Rationale: When the temperature rises above 100°C, most of the liquid phase is converted to water vapor and the limit for pH from *In-Package Chemistry Abstraction* (BSC 2005 [DIRS 174583]) is reached. At these temperatures and humidities, the glass reacts with the water vapor to form a thin layer of water on the surface of the glass. Because the ratio of surface area to volume is very high (about 4,000 m⁻¹), the pH can be very high. As the temperature increases in the repository at constant pressure, the relative humidity decreases. The functional dependence on the relative humidity is not known, but alteration rate is known to decrease to zero at 44%. Both the effect of CO₂ and the relative humidity cannot be accurately calculated as a function of temperature. However, conservatively assuming that only Na⁺ is released from the glass, the condensed water would become a NaOH solution. Reacting this solution with CO₂ gas from a reservoir of unlimited volume at constant $pCO_2 = 10^{-1.5}$ kPa ($10^{-3.5}$ atm) converts the solution to one containing NaCO₃, which, under these conditions, has a pH of about 10.2 to 10.3 (pK_a for $HCO_3^- + H_2O = CO_3^{2-} + H_3O^+$ (Stumm and Morgan 1981 [DIRS 100829], pp. 204 to 206). Therefore, the rate of glass alteration increases as the temperature increases as calculated from the activation energy and the pH is fixed at the high value of 10 for the purposes of calculating an alteration rate. Using a fixed pH is conservative because the calculated rate increases with temperature (activation energy), the actual pH of the

solution on the glass is lower than 10 because of CO₂ availability, and the equilibrium constant for water shifts to lower values

exchange and hydrolysis reactions can occur at sites on the surface of the glass, water also diffuses into the glass and reacts at interior sites (Grambow and Muller 2001 [DIRS 171412]). Alkali metals, alkaline earth metals, and boron are initially released from the glass preferentially to silicon and aluminum in acidic and neutral solutions (Abraitis et al. 2000 [DIRS 163195]). Ion exchange appears to take place independent of matrix dissolution in alkaline solutions (McGrail et al. 2001 [DIRS 171165]). The rates of ion exchange and water diffusion decrease with time and the dissolution becomes controlled by matrix dissolution. Solubility limits with respect to several minerals are quickly reached and they precipitate. The ion exchange and the precipitation of minerals leads to a solution that is incongruent with respect to the glass composition. Because the precipitating minerals remove less alkali, the solution in contact with most nuclear waste glasses becomes alkaline. Although experimentally the glass dominates the solution in contact with glass, the corrosion of the package components will dictate the composition of the solutions that will come in contact with the glass. Thus, implementation of the glass degradation model will utilize the solution pH that is calculated in *In-Package Chemistry Abstraction* (BSC 2005 [DIRS 174583]).²

Because boron is not incorporated into alteration phases, whereas most of the other elements are, the extent of glass dissolution in most laboratory tests is provided by the boron solution concentration. The boron concentration is used to determine the parameter values in the glass degradation model, which is then used to calculate the release rates of radionuclides from the glass. An exception is the use of the vapor hydration test results, which are discussed in Section 6.5.3.3. In these tests, a thin film of water, the thickness of which has only been estimated, contacts glass. The small volume of water promotes the formation of alteration phases. These alteration products are the same as those that are formed in tests in which bulk water is in contact with the test specimen and are formed in the long-term on natural materials (Bates and Steindler 1983 [DIRS 104261]). The extent of glass corrosion is measured using the thickness of an alteration layer (Ebert 2003 [DIRS 164518]) or the thickness of the unreacted glass (Jirička et al. 2001 [DIRS 163262]). With either of these methods, the extent of reaction is determined from the amount of glass converted to alteration phases.

As a result of the mechanistic studies, the glass dissolution rate can generally be expressed with the formula given by Aagaard and Helgeson (1982 [DIRS 101530]):

$$r_{glass} = k_r \times \left[\prod_i a_i^{-\nu_i} \right] \times \left[1 - \exp\left(\frac{A_r}{\sigma RT} \right) \right] \quad (\text{Eq. 5})$$

where

- r_{glass} = glass dissolution rate
- k_r = rate constant that carries the units of rate
- a_i = the chemical activity of the species i
- ν_i = the stoichiometric coefficient for species i in the mechanistic step

² Although the pH is passed from *In-Package Chemistry Abstraction* (BSC 2005 [DIRS 174583]), there is no continuous water when the temperature is above 100°C. In this case, Assumption 5.2, in which the pH is assumed to be 10, is used.

6.3.3 Uncertainties in the Subsystem and Environment

Except in the case where the relative humidity is less than 44%, the amount of water contacting the exposed glass is not specified directly in the model. Rather, it is treated as an uncertainty that is taken into account through the range and distribution of the effective rate constant. The lower end of the range of values of the effective rate constant is selected to yield a calculated value that matches the rate measured in tests with glass contacted by dripping water (when the solution is acidic) or by humid air (when the solution is alkaline) at the pH and temperatures of those tests. The pH of the solution accumulated in a breached waste package is provided by *In-Package Chemistry Abstraction* (BSC 2005 [DIRS 174583]). The upper end of the range of values of the effective rate constant is selected to yield a calculated rate that matches the rate measured for immersed glass. The most likely value of the effective rate constant is selected to be the lower limit. This is because the environment within a breached waste package is anticipated to be hydrologically unsaturated for most of the service life.

The composition of the waste glass in a breached container is also treated as an uncertainty. The radionuclide inventory is averaged over all HLW glass packages. However, an average glass composition is not used in this report. Instead, the results of tests conducted with glasses having a range of compositions are used to provide a corresponding range of degradation rates, and the upper range for immersion conditions and lower range for dripping water conditions is used to determine model parameter values.

6.4 CONSIDERATION OF ALTERNATIVE CONCEPTUAL MODELS

A summary of the alternative conceptual models considered is provided in Table 6-2.

Table 6-2. Alternative Conceptual Models Considered

Alternative Conceptual Models	Key Concepts	Screening Assessment and Basis
Diffusion-controlled release	Release rate of radionuclides determined by solid-state diffusion rates.	Not incorporated into TSPA-LA model Not supported by data for waste glasses
Composition independent effective rate constant	Intrinsic rate constants vary over a small interval for different compositions and the very low flow rates in the repository compared to those used in the laboratory mean that the affinity term will be low.	Not incorporated into TSPA-LA model Current approach provides a much more robust range of values for use in the TSPA-LA

6.4.1 Diffusion-Controlled Release

An alternative conceptual model that is considered for the process controlling glass degradation was developed for HLW glass in France. Researchers at the Commissariat à l'Énergie Atomique (CEA) in France attribute the residual dissolution rate to diffusion-controlled release of silica (Jégou et al. 2000 [DIRS 163260]; Vernaz et al. 2001 [DIRS 163328]; Gin, Jollivet et al. 2001 [DIRS 163256]; Gin, Ribet, and Couillard 2001 [DIRS 163255]). They have concluded that the reaction-control model in Equation 5 is inconsistent with the results of various tests they have conducted with the reference glass R7T7 and simple analog glasses, and that the results are better described with a diffusion-based model. They point to the fact that a range of saturation

p. 573; McGrail, Martin, and Lindenmeier 1997 [DIRS 111040], p. 257). Other processes that remove dissolved silica from solution can impact the glass degradation rate in the same way. This has been observed in tests conducted in the presence of various clays (Gin, Jollivet et al. 2001 [DIRS 163256]). Tests in the presence of ductile iron and various iron oxides have shown a similar effect (McVay and Buckwalter 1983 [DIRS 101728]; Werme et al. 1990 [DIRS 163346]). The removal of silica from solution due to sequestration in glass alteration phases or sorption to other phases has the same effect on the glass degradation rate. The long-term impact of these processes in the repository will depend on the precipitation rates of the alteration phases and sorption capacities of iron corrosion products. The possibility that the precipitation rate or sorption capacity will limit the glass degradation rate is not included in the model, thereby making the model conservative.

Phases like analcime that can cause enhanced dissolution through consumption of silica are unlikely to form under most repository conditions. One exception is the case where the relative humidity is greater than 44% and the temperature of the glass is greater than 99°C. Under these conditions, similar to those of a vapor hydration test, a thin layer of water can form on the surface of the glass because the reaction kinetics are high enough and the initial solution that forms has a water vapor pressure less than the surrounding atmosphere. Unlike the vapor hydration test, however, the repository provides essentially an infinite amount of CO₂ gas at constant $p\text{CO}_2 = 10^{1.5}$ kPa ($10^{-3.5}$ atm). It will react with the solution as it forms initially and limit the rise in pH. Because these conditions have not been investigated (i.e., VHTs with constant partial pressure of CO₂), a limit based on other information is used in this model.

A measure of the pH of the solution that forms on the test specimen in a vapor hydration test is given by Vienna et al. (2001 [DIRS 163331]), Jiříčka et al. (2001 [DIRS 163262]) and Ebert et al. (1991 [DIRS 111026]), where the pH(25°C) either measured on the test specimen or in the solution that dripped from the test specimen can be as high as 12. The high pH(25°C) of the solution that is calculated in *In-Package Chemistry Abstraction* (BSC 2005 [DIRS 174583], Table 8-2) is 8.5. The pH at elevated temperature is lower than at 25°C because of the shift of the water dissociation constant to lower values with increasing temperature. The pH shift is about 0.8 at 90°C and 0.6 at 70°C. Therefore, above 99°C, it is assumed that the pH in this model is fixed at 10 and the increase in the rate is calculated with Equation 13. In practical terms, alteration of the glass in humid air is limited to the temperature range 100°C to 125°C because for pure water, the relative humidity at 1 atmosphere total pressure falls below 44% at 125°C. If salt solutions from the evaporation of groundwater elsewhere in the drift or waste package control the relative humidity, then the upper temperature at which vapor phase alteration would occur is less than 125°C. Both 125°C and a pH value of 10 represent conservative values.

6.5.2.4 Maximum Value of k_E for Glass Degradation in Alkaline Solutions

Triplicate 7-day PCTs were conducted with the nine glasses used to evaluate the effect of glass composition on the value of k_0^{\rightarrow} (Section 6.5.2.2) (DTN: MO0308ANLGPC01.528 [DIRS 164790]). The 7-day PCTs were conducted at 90°C. The averages of tests with each glass were used to extract values of k_{PCT} using Equation 13 with parameter values $\eta = 0.49$ and

$E_a = 69$ kJ/mol (Appendix B, Table B-3). The averages of the measured 7-day PCT rates (based on the release of boron), the measured final pH, and the extracted value of $\log_{10}(k_{\text{PCT}})$ are

The extent of fracturing is modeled to be proportional to the geometric surface area of the glass. Waste glasses will fracture as they cool because of the thermal stresses in the glass and the hoop stresses from the difference in expansion between the glass and the pour canister. Additional fracturing may occur during handling. Although fractures at the surface can be measured and quantified, fractures in the bulk glass cannot. In addition, not all of the fractures that are formed may be accessible to water. The modeled surface area for a glass used in this report is an average value that accounts for the thermal cracking of all glass and cracking of a small percentage of the glass from handling. Data quantifying the effect of thermal cracking and impact cracking are combined with estimates of accessibility and reactivity of glass in tight cracks. A factor of 12 is used to account for thermal cracking in all canisters (Smith and Baxter 1981 [DIRS 102089]) and a factor of 40 is used to account for impact cracking of 1% of the canisters (Smith and Ross 1975 [DIRS 102088]). A maximum value of $f_{\text{exposure}} = 17$ is obtained by assuming that all glass surfaces formed by fractures are freely accessible to water and that glass within cracks has the same reactivity as glass at free surfaces (Section 6.5.4). A minimum value of $f_{\text{exposure}} = 4$ is obtained by assuming that one-half of the glass surfaces formed by fractures are freely accessible to water and that glass within cracks has the one-half the reactivity of glass at free surfaces (Section 6.5.4).

The relative humidity and the range of values for the effective dissolution rate constant, k_E , are used to represent the uncertainty in the glass surface area in contact with water vapor, dripping water, and bulk water. If the relative humidity is less than or equal to 44%, no glass degradation occurs. If the relative humidity is greater than 44%, glass alteration occurs at a rate determined from the rate expression given in this report. The dependence of the rate on relative humidities above 44% and the presence of water (under either dripping or immersion conditions) is contained implicitly in k_E . Between 100°C and 125°C, the pH of the water on the surface of the glass is assigned a value of 10. This value is higher than the mildly acid values noted for the case where the layer drips from the surface of the glass and equal to the high value calculated in *In-Package Chemistry Abstraction* (BSC 2005 [DIRS 174583]). Practically, the lower limit to the relative humidity limits the temperature range over which glass is altered and, hence, the model applied. From the ambient temperatures prior to waste acceptance to the boiling point of water, the alteration rate is given by Equation 13. From 100°C to 125°C, the glass alteration rate is given by Equation 57.

$$\log_{10}(\text{rate}) = 6.35 + \log_{10} \left[\exp \left(\frac{-69 \text{ kJ} / \text{mol}}{(0.008314 \text{ kJ} / \text{mol} \times \text{K}) \times T} \right) \right] \quad (\text{Eq. 57})$$

If salt solutions from the evaporation of groundwater elsewhere in the drift or waste package control the relative humidity, then the upper temperature at which vapor phase alteration would occur is less than 125°C.

Another uncertainty related to the exposed glass surface area is the size of the breached canister. Both “short” canisters (about 3-m long) and “long” canisters (about 4.5-m long) will be used. The glasses in long and short canisters will provide different initial surface areas and initial masses. The initial surface area of glass in a long canister is about 1.5 times that in a short canister. It is anticipated that about two-thirds of the waste glass will be in long canisters and one-third in short canisters (DOE 2002 [DIRS 158873]). This is taken into account by assigning

glass composition, solution composition, and water contact conditions are combined in the ranges of the effective rate constant, k_E , for dissolution in acidic and alkaline solutions. The bounding values are determined from experimentally measured dissolution rates under relevant conditions. The maximum values are determined from immersion tests in acidic or alkaline solutions and the minimum values are determined from tests in dripping water that resulted in acidic conditions and tests in humid air that resulted in alkaline conditions. Since the values of k_E were extracted from the measured reaction rates with constant pH and temperature parameters, the effects of glass and solution composition and processes are not modeled explicitly. The uncertainties in these processes are captured in the range of values of k_E .

6.8.3 Uncertainty in Stoichiometric Releases of Radionuclides

In the model, glass dissolution is treated as a constant rate at a given temperature and pH. The chemistry associated with the dissolution process and the variation with time even at fixed temperature and pH is not calculated. Hence, all radionuclides whether they are controlled by the solubility of a solid phase, such as Np or Pu, or not controlled by solubility, such as Cs or Tc are released congruently with the glass matrix. This approach is taken because the rate a radionuclide is released from the glass matrix cannot be distinguished from the effects of solubility control in most test results. The effects of solubility control are imposed in the TSPA-LA with a different model.

6.8.4 Impact of Input Variable Uncertainty

The uncertainty in the exposure conditions is provided through the input variables pH, temperature, and relative humidity. Therefore, the sensitivity of the glass alteration rate is obtained through the variability in the individual input parameter. The assignment of constant values to the model parameters for the pH and temperature dependence provides a link between the uncertainty in the pH and temperature and the uncertainty in the rate that can be readily calculated. For example, the degradation rate calculated by the model increases by about a factor of 3.1 for every unit increase in pH in acidic or alkaline solutions. When the temperature increases, for example, from 35°C to 45°C in alkaline solutions, the alteration rate increases by a factor of 2.7 or decreases by a factor of 2.9 when the temperature decreases from 35°C to 25°C (Equations 50 and 51). Smaller temperature effects occur for acidic solutions. Uncertainty in the relative humidity impacts this model only with regard to whether the relative humidity is greater than or less than 44%.

6.8.5 Impacts of Uncertainty on Scientific Analysis Output

Uncertainty in the individual effects of glass composition, solution composition, temperature, and pH on the glass degradation rate is not propagated into other models, such as *In-Package Chemistry Abstraction* (BSC 2005 [DIRS 174583]). This is because the glass dissolution model provides only the calculated glass degradation rate, not individual parameter values. The assignment of single values, rather than ranges of values, for the pH and temperature parameters and assigning the uncertainties in the parameter values to the ranges of k_E for degradation in acidic and alkaline solutions reduce the uncertainty in the calculated glass dissolution rate. This approach avoids the possible calculation of unrealistically high or

extremely low rates that would be calculated if unrealistic combinations of parameter values were selected. The range of rates

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