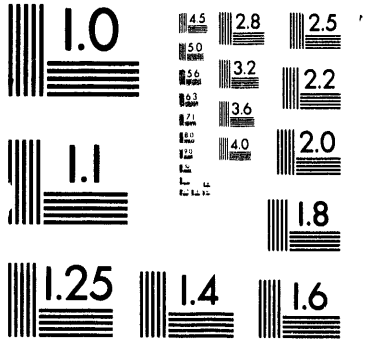
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HIGH-LEVEL WASTE GLASS COMPENDIUM; WHAT IT TELLS US CONCERNING THE DURABILITY OF BOROSILICATE WASTE GLASS

by

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ABSTRACT

Facilities for vitrification of high-level nuclear waste in the United States are scheduled for startup in the next few years. It is, therefore, appropriate to examine the current scientific basis for understanding the corrosion of high-level waste borosilicate glass for the range of service conditions to which the glass products from these facilities may be exposed. To this end, a document has been prepared which compiles worldwide information on borosilicate waste glass corrosion. Based on the content of this document, the acceptability of canistered waste glass for geological disposal is addressed.

Waste glass corrosion in a geologic repository may be due to groundwater and/or water vapor contact. The important processes that determine the glass corrosion kinetics under these conditions are discussed based on experimental evidence from laboratory testing. Testing data together with understanding of the long-term corrosion kinetics are used to estimate radionuclide release rates. These rates are discussed in terms of regulatory performance standards.

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HIGH-LEVEL WASTE GLASS COMPENDIUM; WHAT IT TELLS US CONCERNING THE DURABILITY OF BOROSILICATE WASTE GLASS

J. C. Cunnane and J. M. Allison

INTRODUCTION

Almost four decades have passed since glass was suggested as a matrix for immobilization of high-level nuclear waste (HLW) [1]. During this period, an extensive international effort has been pursued to develop the technology and scientific basis for use of glass waste forms in the management of the HLW. This effort has led to international technical consensus on the choice of borosilicate glass as a waste form. In the United States, after an extensive evaluation of alternatives, the decision was made over a decade ago to proceed with the development of facilities that will convert the high-level waste stored in underground tanks into a borosilicate glass. The fruition of this effort will occur with the planned hot startup of the Defense Waste Processing Facility (DWPF) at the Savannah River site, Aiken, South Carolina, in 1995 and at the West Valley Development Project (WVDP), West Valley, New York, in 1996.

The suitability of the canistered waste glass products to be produced by these facilities for geologic disposal is an issue of considerable regulatory and public interest. Although the formal licensing decision on these glass products will take place after hot startup of the vitrification facilities [2], it would be imprudent to start up these facilities without a sound technical basis for the expectation that the canistered glass products will be found suitable for disposal. The need to compile the pertinent technical information has motivated the preparation of the document entitled "High-Level Nuclear Waste Borosilicate Glass: A Compendium of Corrosion Characteristics." The objective of the document is to summarize available scientific information relevant to understanding and assessing waste glass corrosion and the associated radionuclide release under the environmental conditions to which the waste glass may be exposed during geological disposal.

The first draft of the compendium was summarized in a paper presented in this symposium a year ago [3]. Since then, the document has undergone extensive review and revision. Rather than present an update of the revised content, the focus of this paper is on what the compiled scientific information indicates concerning the suitability of the waste glass products for geologic disposal. More specifically, it addresses the following question: "Does the available scientific information indicate that the waste glass products will corrode and release radionuclides at an acceptably low rate when they are eventually contacted by groundwater following disposal in a geologic repository?"

The next section addresses the rates at which radionuclides immobilized in the glass matrix may be released into groundwater as a result of aqueous corrosion of the waste glass matrix. These rates are then compared with a standard for what might constitute an "acceptably low release rate." Although previous performance assessments have addressed these issues [4], the intent of this paper is to complement such studies by presenting the pertinent information in a simple fashion wherein the underlying data and assumptions are amenable to scrutiny by interested parties who may be unfamiliar with probabilistic performance assessment models.

RELEASE OF RADIONUCLIDES FROM CANISTERED WASTE GLASS IN A REPOSITORY

Figure 1 shows the waste glass composition and nominal design of the canistered waste glass to be produced by the DWPF. This canistered waste glass will be enclosed (packaged) in other components of the engineered barrier system in a geologic repository. These barriers are expected to be eventually breached, allowing the fluid in the repository subsurface to interact with the waste glass and initiate waste glass corrosion and associated radionuclide release.

Figure 2 provides a schematic illustration of some of the waste glass features that are pertinent to glass corrosion and associated radionuclide release in a repository. The environmental conditions to which the waste glass will be exposed cannot be precisely determined because neither the repository location nor the engineering design has been established. The gap region (i.e., region 4 in Fig. 2), which defines the corroding glass fluid environment, may be filled with (1) groundwater, (2) air/water vapor mixtures along with a liquid phase present as water films and/or dripping water, or (3) air/water vapor mixtures. In this paper, the waste glass corrosion due to exposure to intermittent water, humid air, and/or water vapor contact will be referred to as "weathering."

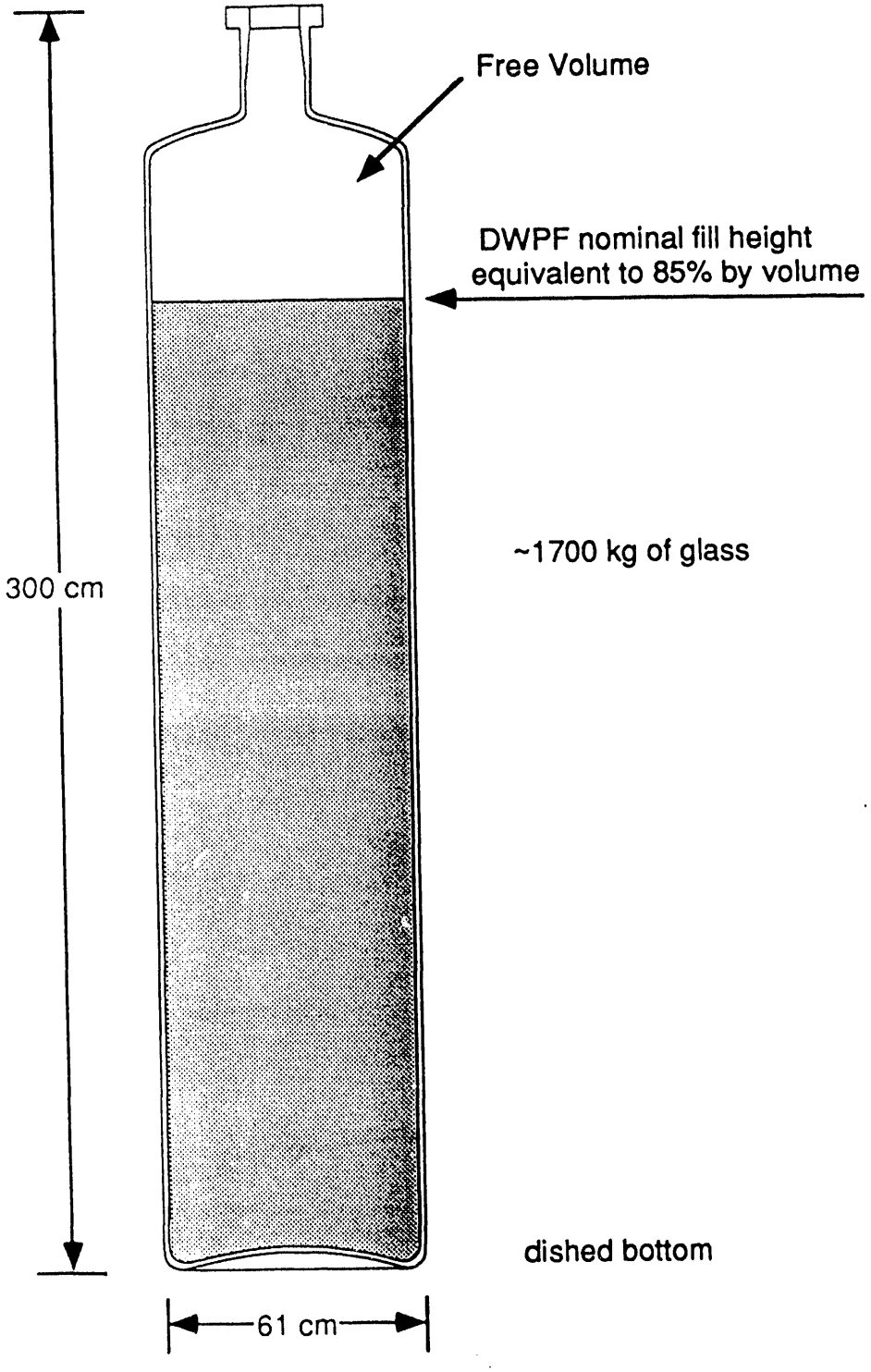
The release of radionuclides through the engineered barrier systems (EBS) can be considered to consist of the following sequential steps: 1) corrosion of the glass matrix; 2) mass transport of the radionuclides in the corroded glass through the surface alteration layers and release into region 4 as dissolved and colloidal material; and 3) mass transport through the EBS and into the host rock, i.e., external-field mass transport. The release rate can be estimated on the basis of the slowest step in a sequential process and the fastest step in a parallel process [5]. Hence, the release across the EBS boundary can be conservatively estimated by considering either the rate of release to region 4 or the rate of external-field mass transport, assuming solubility-limited concentrations for the sparingly soluble radionuclides in region 4. Although the latter approach has been adapted in recent performance assessment studies [4, 6], the focus here is on the former. The rate of radionuclide release from individual waste glass logs into region 4 is calculated by the following method.

The fraction (F) of a canistered waste glass that would corrode per year after it is contacted by groundwater is given by:

$$F = \frac{RA}{W} \tag{1}$$

where R is the glass corrosion rate ($g/m^2/yr$), W is the weight (g) of the glass in a canister (~1.7×10^6 g, see Figure 1), and A is the surface area (m^2) of the glass contacted by water. Because radionuclides can only be released from properly formulated waste glass as a result of breakdown of the glass network, the fractional release of individual radionuclides cannot exceed the fraction of the glass corroded. The ratio of the cumulative corrosion (as measured by the normalized release of boron) to the cumulative release of individual radionuclides is given by the retention factor $(RF)_i$. Hence, the fraction of the individual radionuclides expected to be released per year from a corroding glass log is given by:

Fig. 1 Schematic of the DWPF Canistered Waste Showing Nominal Physical Characteristics and Relevant Glass Compositions.

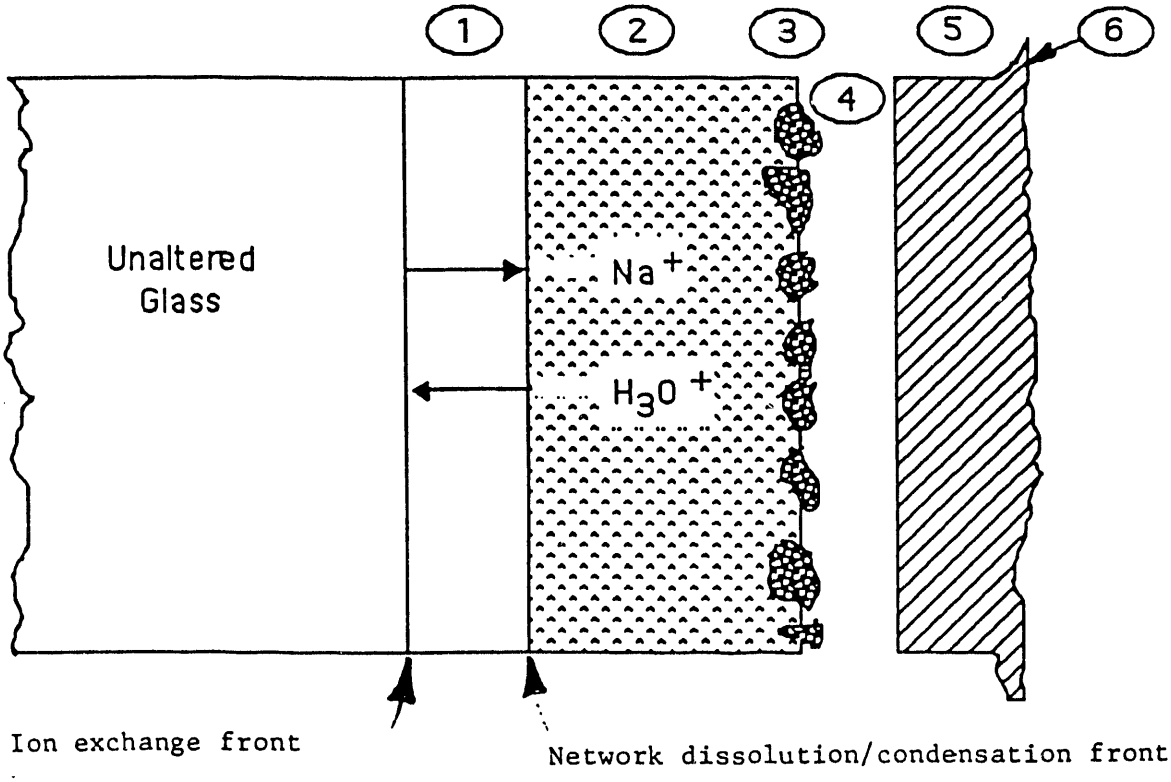


DWPF Design-Basis Waste Glass Composition

	Composition (wt.%)[a]	
Component	Blend	SRL 202
SiO_2	50.2	49.0
B_2O_3	8.0	8.0
Al_2O_3	4.0	3.8
Fe_2O_3	10.4	11.4
Na_2O	8.7	8.9
K_2O	3.9	3.7
Li_2O	4.4	4.7
U_3O_8	2.1	1.9
CaO	1.0	1.2
MgO	1.4	1.3
MnO	2.0	2.0
TiO_2	0.9	0.9
NiO	0.9	0.82

[a] Components <0.5 wt. % not tabulated.

Fig. 2 Schematic of HLW Borosilicate Glass Corrosion Following Geologic Disposal



1 Reaction Zone (ion exchange and network hydrolysis reactions occur in this layer as does interdiffusion of the reactants and exchanged cations)

2 Gel Layer

3 Precipitated Layer

4 Fluid-Filled Gap

5 Corroded Engineered Barriers

6 Boundary of the Engineered Barrier System

$$f_i = \frac{F}{(RF)_i} = \frac{RA}{W(RF)_i} \quad (2)$$

Hence, estimation of f_i reduces to estimation of A, R, and RF_i.

As discussed below, the values of each of these parameters are uncertain, and hence, each is best represented as a distributed probabilistic variable. Also, each is either known to be or is potentially a function of time. We thus selected conservative estimates for each of the parameters based on short-term experimental data and information pertinent to long-term extrapolation.

The American Society for Testing and Materials [7] has developed a standard practice for prediction of the long-term behavior of waste package materials including waste forms used in geologic disposal of high-level nuclear waste. In simplified terms, this practice entails three important steps in the development of a credible basis for long-term prediction: (1) identifying the alteration processes, (2) conducting a variety of tests to develop quantitative models based, if possible, on a mechanistic understanding of the separate and combined effects of these processes, and (3) conducting tests for model validation. The scientific basis for extrapolating the long-term behavior of the above parameters is discussed below in light of these three steps.

The Corroding Glass Surface Area (A)

The surface area of the glass log that may be contacted by water can vary from a fraction of its geometric surface area (for weathering conditions) to the full surface area of its fracture surfaces. Fractures in canistered glass result from stresses induced in the glass during cooldown after pouring into the canisters [8]. The extent of cracking (and hence, the fracture surface area) is determined primarily by the rate of cooling below the glass transition temperature. Measurements of the surface area for full-scale canistered DWPF glass, cooled at a prototypical cooling rate, showed that the total surface area was a factor of 18 greater than the geometric surface area of the glass monolith [9]. Short-term leaching tests [10, 11] indicate that the surface area in tight cracks does not contribute to the corrosion. However, observations from the corrosion of natural basaltic glass suggest that over the long term, even the glass surfaces in tight cracks corrode at a rate comparable to the exposed surfaces [12]. Because there is no evidence that significant additional cracking will occur after repository emplacement, a reasonably conservative estimate for A, under immersion corrosion conditions, is 20 times the geometric surface area, i.e., 96 m^2. Of course, A may be considerably less than this value for weathering conditions.

The Rate Of Waste Glass Corrosion (R).

Silicate glass is metastable and, therefore, does not reach thermodynamic equilibrium with aqueous environments; its chemical durability derives from the slow kinetics of the corrosion and weathering processes that lead to breakdown of the glass network. Corrosion and weathering occur as a result of ion exchange and a number of network hydrolysis reactions that occur at glass surfaces exposed to groundwater and/or water vapor [2]. The corrosion and weathering of waste glass rates in a repository are complex functions of the waste glass composition and environmental exposure conditions.

The important processes (i.e., sequences of chemical reactions and associated mass transport steps) that contribute to the corrosion of waste glasses have been identified [6] to be the following [13]:

- Diffusion-controlled ion exchange and extraction of alkali ions. This process causes the interface between the glass and the reaction zone to move inward as the corrosion proceeds. The diffusion resistance is provided by the reaction zone (see region 2 in Fig. 2).

- Dissolution-controlled hydrolysis of the glass matrix. This process causes the interface between the gel and reaction zone to advance inward as corrosion progresses. Under some circumstances, the rate of the dissolution reaction is controlled by mass transport of the reaction products outward through the surface layers (i.e., through regions 2 and 3 in Fig. 2).

The first of these processes includes network-hydrolysis-controlled diffusion of water into the glass at the glass/water interface, ion exchange, and release of boron, while leaving much of the affected network near the glass surface intact. The second results in complete degradation of the glass network in the affected region near the glass surface due to glass network hydrolysis and dissolution reactions occurring at the surface of the reaction zone (see Fig. 2). While these processes occur simultaneously, their relative importance changes with time. In fact, the corrosion behavior exhibited by the evolution of the surface layers (which represent progressive stages in the glass degradation) can be interpreted in terms of the relative rates and effects of these processes. Waste glass corrosion, including the development of surface alteration layers and leaching of waste glass components, is discussed briefly below to illustrate the point that the underlying processes are sufficiently well understood to support comparison of short-term corrosion data with the rates that can be expected over the long term. Also, studies of very old basaltic glasses [12, 13] suggest that the processes observed in short-term laboratory tests are also important in long-term corrosion of glass.

Upon initial immersion in water, both diffusion-controlled alkali extraction and dissolution-controlled corrosion occur in parallel. The former causes an alkali-depleted zone (referred to as the reaction zone or diffusion layer) to develop at the glass surface. The latter causes the outermost surface of the reaction zone to move inward toward the unaltered glass and causes a residue of insoluble materials (referred to as the gel layer) to develop atop the diffusion zone (see Fig. 2). Initially, the former process is dominant because it proceeds at a higher rate. Both processes slow with time, but diffusion-controlled ion exchange slows faster because of the increasing thickness of the diffusion layer or reaction zone. When the rate of dissolution equals the rate of diffusion, a steady-rate condition occurs wherein the diffusion layer retains a constant thickness. Thereafter, the rate of diffusion and the overall corrosion rate are controlled by dissolution because the dissolution rate now controls the thickness of the diffusion layer. When the contacting solution becomes supersaturated with respect to

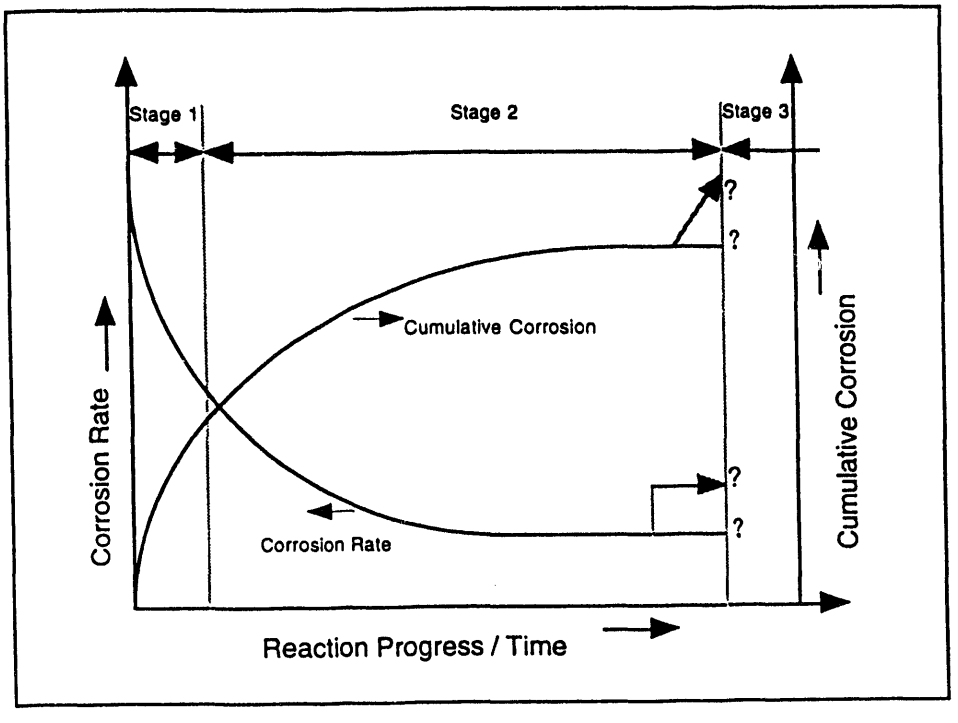
secondary mineral phases, these precipitate to form the precipitated layer shown in Fig. 2. Some evidence indicates that when the dissolution step slows sufficiently, the diffusion-controlled alkali-extraction process may again become rate limiting [13]. For a more complete understanding of the general concepts discussed above, complicating factors must be considered -- such as the coupling between processes and the interdependencies among the various steps in each process. For example, although the rate of ion exchange depends on the rate of dissolution because the latter affects the diffusion layer thickness, the rate of dissolution is influenced by ion exchange because of its effect on pH. As an example of the interdependency of the steps in the dissolution process, precipitation of iron and aluminosilicates lowers the silicic acid concentration, which, in turn, increases the dissolution rate [15, 16].

Figure 3 illustrates some of the typical characteristics of the variation in glass corrosion as a function of time as measured in static leaching tests. The two curves represent the normalized release of boron (a measure of cumulative corrosion) and the corresponding corrosion rate. The time axis is divided into three parts to illustrate three stages in the progression of the glass corrosion. Stage 1 is characterized by a period when the corrosion is dominated by the diffusion-controlled ion exchange process. Stage 2 is characterized by the occurrence of ion exchange, network hydrolysis, and network dissolution, with the overall rate of corrosion being controlled by the dissolution step. Stage 3 represents the long-term corrosion beyond experimentally accessible time; the processes that control the corrosion rate during this stage are less well established.

This time-dependent behavior of the cumulative corrosion and corrosion rate curves in Fig. 3 has been explained as arising from changes in the leachate (e.g., increased silicic acid concentration) that are produced by the corroding glass. These changes, in turn, have a feedback effect that reduces the rate during Stage 2 of the corrosion process. In general, the time dependence of the corrosion rate for a particular glass composition is attributed to a variety of feedback effects associated with the reaction progress, e.g., the effects of the increasing diffusion layer thickness on the rate of diffusion in Stage 1 and the effects of the increasing concentrations of silicic acid (and other glass constituents) during Stage 2. Since the time dependence can be attributed to the effects of reaction progress, it is useful to consider the corrosion rate as a function of a generalized reaction progress parameter, as illustrated in Fig. 3. The corrosion rate exhibits a generally decreasing trend with reaction progress. Two characteristic rates (i.e., a "forward reaction rate" and a "saturation rate") can be identified, corresponding to infinite dilution and silicon saturation conditions, respectively. A discontinuity in the rate curve (change in the slope of the cumulative corrosion curve) is indicated by the dashed lines near the end of Stage 2 in Fig. 3. This decrease in the corrosion rate may not be monotonic for some glasses at very high values of the reaction progress parameter. Such effects may be due to nucleation and precipitation of secondary phases that establish a lower saturation concentration of silicic acid (or other solution species) and thereby increase the affinity for glass dissolution. Although it may be possible to avoid such effects by proper formulation of waste glass compositions, these effects need to be fully understood to support long-term extrapolation of experimental results.

Forward and saturation rates for a variety of reference waste glasses measured at 90°C and at different ratios of surface area to solution volume (S/V) are compiled in Table 1 [17-20]. Factors such as temperature, radiation dose, and interactions with other materials may cause the values to vary somewhat from the values in Table 1. For extrapolation of these experimental results into Stage 3, the current theoretical understanding and empirical evidence indicate that glass corrosion rates should generally decrease with time. This occurs because the buildup of corrosion products in solution and on the surface of the glass will, in general, inhibit the corrosion process. However, effects such as onset of nucleation and

Fig. 3 Schematic Showing Qualitative Behavior of Glass Corrosion v. Time



precipitation of secondary phases, increased water flow rate, and cracking of protective layers may increase the rate of the dissolution-controlled process for some glass compositions. While such effects introduce uncertainties in accurate long-term corrosion predictions, it appears that predictions based on the assumption that the measured "saturation rates" provide an upper bound on the long-term rates can be made with some confidence [21]. Hence, based on the saturation corrosion rates in Table 1, a reasonably conservative estimate for the long-term corrosion rates of SRL202 glass and the French R7T7 glass is about $2.5 \times 10^{-3} g/m^2/d$ at 90°C.

Radionuclide Retention in Surface Alteration Layers (RF_i)

For many radioelements, only a fraction of the radionuclides in the corroded glass is released to the contacting solution as solutes and colloids. As the glass corrodes, the sparingly soluble radionuclides do not enter solution due to their low solubilities or due to retention in the surface layers on the glass as a result of such processes as sorption or incorporation of solids from the solution into secondary mineral phases on the glass surface.

The retention of any element i during glass corrosion can be quantified by its retention factor, which is equal to NL(B)/NL(i), where NL(i) is the normalized mass loss of element i and NL(B) is the normalized release of boron [22]. Boron is assumed to be totally released into solution and to represent the extent of glass corrosion [23]. High RF values indicate that the radionuclides involved remain largely immobile as the glass corrodes, while low values indicate that a significant fraction of the radionuclides associated with the corroded glass is released into the contacting solution as potentially mobile solute or colloidal forms. Figure 4 illustrates the retention factors that have been measured for U, Np, Pu, and Am when R7T7 glasses were leached in distilled water at 90°C. This figure illustrates that only a fraction (~ 10% for U and Np, ~ 3 % for Pu and ~ 0.3% for Am) of the sparingly soluble radionuclides associated with the corroded glass is released. Similar results have been obtained by other investigators [24]. While these empirical data are not supported by a quantitative model based on a mechanistic understanding of the underlying sorption and precipitation or coprecipitation processes, the values cited above provide a reasonable, albeit uncertain, estimate for RF.

CONCLUDING REMARKS

Based on the values given for the parameters discussed above, the fraction of a canistered waste glass log that would corrode per year is given by:

$$F = [2.5 \times 10^{-3}\ g/m^2/d][96\ m^2][365\ d/yr]/[1.7 \times 10^6\ g] = 5.2 \times 10^{-5}\ yr^{-1}$$

Using Eq. 2 and the values for RF in Fig. 4, we determined that the fractional release rates (f_i) are $5.2 \times 10^{-6}\ yr^{-1}$ for U and Np, $1.6 \times 10^{-6}\ yr^{-1}$ for Pu, and $1.6 \times 10^{-7}\ yr^{-1}$ for Am. The question of whether or not these represent "acceptably low release rates" is discussed briefly below.

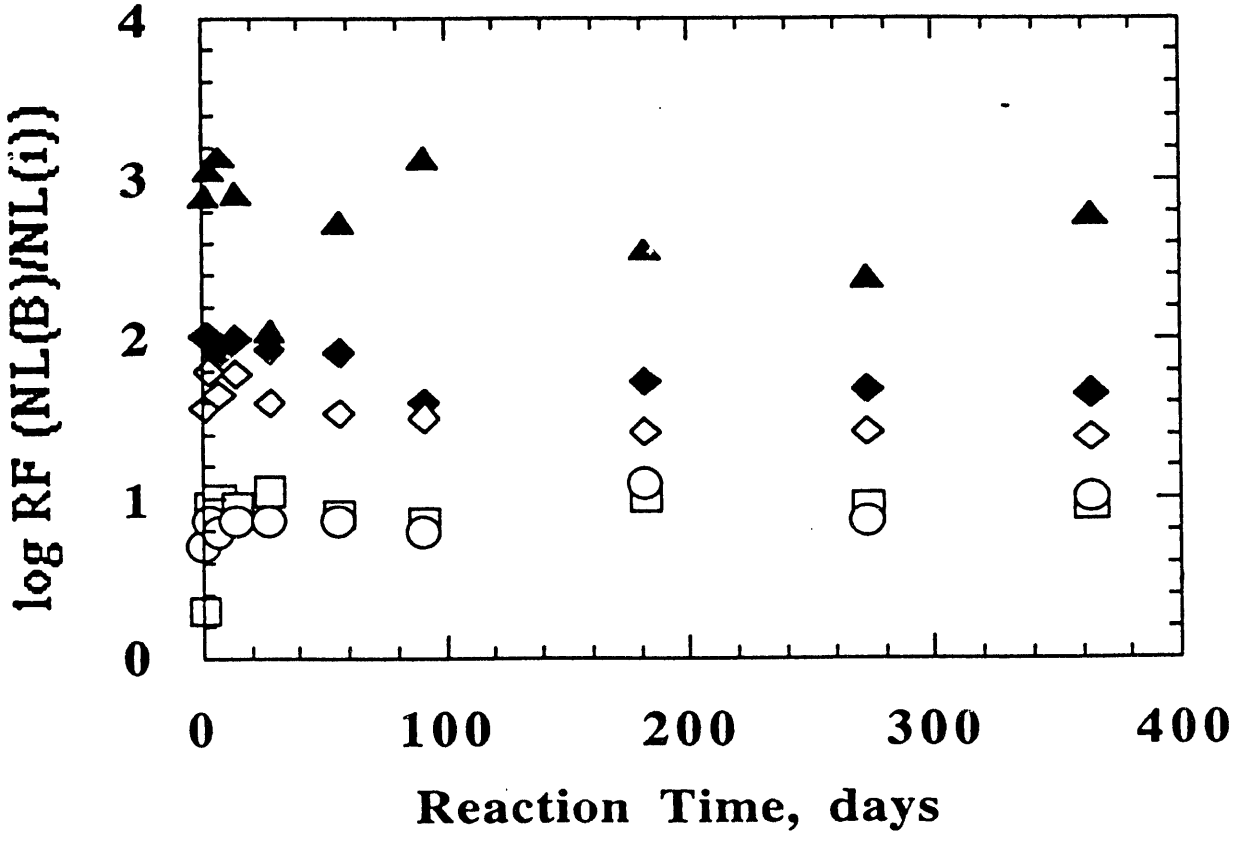
There is no established standard for what constitutes an acceptably low rate of radionuclide release from the individual waste glass canisters in a repository. Although it does not directly apply, the most pertinent standard is the performance objective for the (EBS) "controlled release rate" performance objective [22]. This performance objective establishes a standard for the rate (expressed as an annual fraction of the 1000-yr inventory) at which radionuclides may be released from the EBS, which includes the whole ensemble of waste

Table 1. Experimentally Determined Reaction Rates at 90°C, in g glass per m^2/day

Glass/Leachant	S/V (m^{-1})	Forward Rate	Saturation Rate	Reference
Static Tests				
PNL 76-68/DIW	2000	1.6	0.08[a]	[17]
SRL 165/DIW	2000	0.80	0.024[a]	[17]
EMS-11/DIW	2000	0.083	0.0016[a]	[17]
JSS-A/DIW	10[b]	1.5	0.0025	[18]
PNL 76-68/DIW	10[b]	1.8	0.0075	[18]
SRL 131/DIW	10[b]	3.0	0.033	[18]
SRL 131/J-13[c]	10	0.14		[19]
SRL 131/J-13	2000	0.24	0.021	[19]
SRL 131/J-13	20,000	0.84	0.053	[19]
SRL 202/J-13	10	0.10		[19]
SRL 202/J-13	2000	0.025	0.0016	[19]
SRL 202/J-13	20,000	0.04	0.0025	[19]
R7T7/DIW	50		0.0083	[20]
R7T7/Volvic[d]	50		0.0133	[20]
R7T7/DIW	400		0.0045	[20]
R7T7/Volvic	400		0.025	[20]
R7T7/Volvic	2000		0.0006	[20]
R7T7/Volvic	8000		0.0006	[20]
R7T7/Volvic	20,000		<0.0001	[20]

[a]Estimated.
[b]Values determined from results of both static and dynamic tests.
[c]Tuff groundwater. Major components are 45 ppm Si, 55 ppm Na, and 120 ppm HCO_3^-.
[d]Granite groundwater. Major components are 11 ppm Si, 9.8 ppm Ca, 9.2 ppm Na, and 66 ppm HCO_3^-.

Fig. 4. Logarithm of the Retention Factors for Actinides Released from R7T7 Glass at 90°C (data from [VERNAZ-1992b]): (○) U, (□) ^{237}Np, (◊) ^{238}Pu, (♦) ^{239}Pu, and (▲) ^{241}Am.



packages to be emplaced in the repository. Assuming that the allowed fractional release rate is apportioned equally among each type of waste in the repository, we determined the fractions of the individual radionuclide inventory in the WVDP waste that could be released at the EBS boundary. Table 2 tabulates the fraction of the inventory of individual radionuclides (whose inventory is great enough that they could not be released in a single year) that may be released at 1,000 years and at 10,000 years after closure of the repository. Comparison of the allowed release rates in Table 2 with the estimates for corrosion and radionuclide release rates presented above indicates that the waste glass will be an effective barrier for inhibiting release of radionuclides in a repository. In fact, this comparison suggests that the release rate of glass alone (i.e., without assistance from other components of the EBS) may be sufficient to establish an acceptably low release rate. Moreover, while the annual fractional release from each individual canister is lower than NRC allowed fractional release from the EBS would, for most scenarios, be sufficient, it is not necessary for compliance with the controlled-release-rate performance objective. That is because other components of the EBS (e.g., the waste package containers) will contribute to controlling the release from the EBS [4]. On the other hand, this fractional release may not suffice if, for example, the breached waste glass packages were exposed to weathering conditions for a number of years and were subsequently contacted by groundwater.

The available data and current understanding of waste glass performance described herein indicate that properly formulated borosilicate glass should be effective in inhibiting the release of radionuclides under repository service conditions. However, this analysis alone is not sufficient to take credit in licensing for the level of performance indicated. Given the uncertainty in the available experimental data and in long-term extrapolation, others may feel that more conservative estimates than those selected here should be utilized. Probabilistic performance assessment is needed to support more definitive conclusions for a specific repository location and EBS design.

Table 2. Allowed Release Rates from the EBS for Mp, Pu and Am Isotopes in WVDP Glass[(a)]

Isotope	1000-Yr Postclosure Inventory[c] (Ci)	10,000-Yr Postclosure Inventory (Ci)	NRC Release Rate Limit per Year (Ci)	Allowed Fractional Release Rate at 1000 Yr^{-1}	Allowed Fractional Release Rate at 10,000 Yr^{-1}
U-233	9.95E+00	9.6E+00	2.13E-04	2.1E-05	2.2E-05
U-234	7.16E+00	7.0E+00	2.13E-04	3.0E-05	3.0E-05
U-235	1.02E-01	1.0E-01	2.13E-04	2.0E-03	2.1E-03
U-236	3.40E-01	3.4E-01	2.13E-04	6.3E-04	6.3E-04
U-238	8.50E-01	8.5E-01	2.13E-04	2.5E-04	2.5E-04
Np-237	2.33E+01	2.61E+01	2.33E-04	1.0E-05	8.9E-06
Pu-238	1.60E+00	--	2.13E-04	1.3E-04	1.0
Pu-239	1.65E+03	1.3E+03	1.65E-02	1.3E-05	1.3E-05
Pu-240	1.22E+02	4.7E+02	1.22E-02	1.3E-05	2.6E-05
Pu-241	8.12E+00	--	2.13E-04	2.6E-05	1.0
Pu-242	1.70E+00	1.7E+00	2.13E-04	1.2E-04	1.3E-04
Am-241	1.36E+04	7.3E-03	1.36E-01	1.0E-05	1.0
Am-242m	1.12E-01	--	2.13E-04	1.9E-03	1.0
Am-243	2.17E+03	9.3E+02	2.17E-02	1.0E-05	2.3E-05

[a]Source: adapted from [SCP-1988].
[b]Radionuclides with a release that must be controlled at 1 part in 100,000 of their own 1,000-yr postclosure inventory are underlined.
[c]E indicates exponential notation.
[d]Grow-in affects this ratio.

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