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SHORT-TERM CONSISTENCY TESTING VS. LONG-TERM BEHAVIOR

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ABSTRACT

We conducted Product Consistency Tests (PCTs) with a surrogate low-activity waste (LAW) glass to (1) evaluate the possible use of various test conditions in a specification test for LAW waste forms, (2) measure the reproducibility of the test at low temperatures, and (3) determine if the rates calculated from 7-day PCTs bound the rates measured in PCT conducted for longer durations, which represent more advanced corrosion. The effects of temperature and pH on the dissolution rate in PCTs are much less than the effects observed in dilute solutions due to the buildup of dissolved glass components in the PCTs. The precision of replicate 7-day tests at 20 and 40°C was limited by the analytical uncertainty. The dissolution rates at all temperatures decreased with the test duration initially. However, the dissolution rates in tests at 70 and 90°C increased when certain alteration phases formed after about 100 and 500 days, respectively; the rates in some tests exceeded that measured in a 7-day PCT. While the 7-day PCT does not provide a bounding rate for this glass at 70 or 90°C, tests for longer durations are needed to determine if a 7-day test provides a bounding rate at lower temperatures.

INTRODUCTION

MASTER

High-level waste (HLW) and low-activity waste (LAW) at several U.S. Department of Energy (DOE) facilities will be vitrified to immobilize radionuclides prior to disposal. The corrosion behavior of those waste glasses over long disposal times must be known to support performance assessment calculations and provide confidence that a disposal system will adequately isolate radionuclides. Glass behavior over the very long times that radionuclides must be contained by a disposal system cannot be measured directly under disposal conditions, but must be calculated by combining knowledge of the glass corrosion mechanism and likely materials interactions with anticipated environmental conditions.¹ Laboratory tests are being conducted to understand the corrosion mechanism of waste glasses and the effects of various waste form and environmental factors, such as glass composition, temperature, and groundwater chemistry. Product specifications that exist for HLW and those that are being developed for LAW do not directly measure the long-term behavior of the waste forms. Instead, specifications focus on the consistency of product behavior in short-term tests that can be conducted quickly and remotely in a production facility.

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The Product Consistency Test (PCT) was developed to monitor the consistency of the dissolution behavior of the glass products from the Defense Waste Processing Facility (DWPF).^{2,4} The Waste Acceptance Product Specifications for Vitrified High-Level Waste Forms (WAPS) were written to facilitate transfer of HLW forms from production facilities for disposal.⁵ These require that the reactivity of HLW glasses in a 7-day PCT be consistently lower than that of a benchmark glass, which is the Environmental Assessment (EA) glass. A similar approach is being considered for LAW forms, and a variation of the PCT was included in the specifications for Phase 1a of the remediation program for Hanford tank waste.⁶ Inherent to this approach is the presumption that a waste glass that is less reactive than the benchmark in a short-term test will have acceptable long-term behavior during disposal.

At Argonne National Laboratory (ANL), we are providing technical support to DOE's HLW and LAW programs by studying the relationship between the behavior of relevant glasses in short-term tests and their long-term corrosion behavior. In this paper, we compare the reactivity of a surrogate LAW glass in a 7-day PCT with its long-term corrosion behavior to determine if its dissolution rate will likely always be less than the rate measured with a 7-day PCT. In addition, we present the results of replicate PCT conducted at 20 and 40°C with the same glass to study the precision of the test at temperatures that are relevant to anticipated LAW disposal conditions.

EXPERIMENTAL

All tests were conducted with LD6-5412 glass, which was made at Pacific Northwest National Laboratory (PNNL) as a part of a glass optimization program and provided to ANL for testing.⁷ The composition is given in Table 1. The LD6-5412 composition neither represents a waste glass formulation nor is it intended to be used as a benchmark or reference glass for LAW forms. It was selected for testing because its properties were typical of the suite of glasses developed in the PNNL program. All tests discussed in this paper were conducted with glass in the -100 +200 mesh size fraction (74-140 μm) that was washed following the

Table 1. Composition of LD6-5412 Glass, in oxide mass%.

Al_2O_3	12.5	Fe_2O_3	0.13	MnO_2	0.005	SiO_2	57.5
B_2O_3	5.19	K_2O	1.33	Na_2O	19.7	TiO_2	0.008
CaO	3.79	MgO	0.03	P_2O_5	0.12	ZrO_2	0.007
Cr_2O_3	0.05						

PCT procedure to remove fines. Tests were conducted by adding the appropriate amounts of crushed glass and water to a test vessel to achieve the desired glass surface area/solution volume ratio (S/V). The vessel was then sealed, weighed, and placed into an oven previously set at the desired temperature. The 20°C tests were conducted in a water bath. Most tests were conducted with demineralized water in either TFA Teflon or Type 304L stainless steel vessels. Some tests were conducted with a pH 12 solution (0.005 m LiCl + 0.0107 m LiOH).

At the end of the test, the vessel was removed from the water bath or oven and weighed. Vessels removed from the water bath were dried before weighing. Aliquots of the leachate were analyzed for pH with a combination microelectrode (all measurements were made at room temperature) and for cations with inductively coupled plasma-mass spectrometry (ICP-MS). Leachates from replicate tests were analyzed with ICP-MS on three separate days to measure the reproducibility of the analysis. Reacted solids from long-term tests were examined with a scanning electron microscope (SEM) and a transmission electron microscope (TEM) to identify alteration phases.

While knowledge of the absolute surface area is not required for application of the PCT for product consistency, it must be known to compare PCT results to the results of other tests and to calculate the dissolution rate. The PCT procedure recommends that the surface area of crushed glass be estimated by assuming the particles have a geometric shape that can be related to the sieve sizes, such as a sphere or a cube. By approximating glass of the -100 +200 mesh size to be cubes with an edge equal to the average mesh opening, the specific surface area is calculated to be about 0.02 m²/g for LD6-5412, which has a density of about 2.5 g/cm³. Thus, glass mass/water volume of 0.02, 0.05, 0.1, and 1 g/mL were used to attain S/V ratios of about 400, 1000, 2000 and 20,000 m⁻¹, respectively.

The results of three series of tests are discussed in this paper: short-term PCTs conducted to measure the effects of the S/V ratio, temperature, and duration on the dissolution rate; replicate PCTs conducted at 2000 m⁻¹ to measure the reproducibility of the test method at 20 and 40°C; and long-term PCTs conducted to study advanced stages of corrosion behavior. These tests provide insight into both the test method and the glass corrosion behavior.

RESULTS

The results of several PCT conducted to determine the effects of the S/V ratio (400, 1000, and 2000 m⁻¹), test duration (1, 3, and 7 days), and temperature (20 and 40°C) on the reactivity in a short-term test are summarized in Table 2. All tests were conducted in Teflon vessels. Most were conducted with demineralized water, although some were conducted with a pH 12 solution as the leachant. The measured pH and B, Na, and Si concentrations and the average dissolution rates are tabulated. The average dissolution rates, NR(i), are calculated as:

$$NR(i) = C_i / \{(S/V) \cdot f_i \cdot t\} \quad (1)$$

where C_i is the measured concentration of component i; f_i is the mass fraction of i in the glass; and t is the reaction time. The average dissolution rates based on the releases of B, Na, and Si all decrease as the S/V and reaction time increase, and increase as the temperature increases. The effect of the pH was measured directly by conducting one 7-day 20°C test at each S/V with a pH 12 solution. The final pH values in these tests were about 3, 2.5, and 2 pH units higher than the final pH values in tests conducted with demineralized water at S/V of 400, 1000, and

Table 2. Results of Modified PCT with LD6-5412 Glass.

Test No.	Temp. °C	S/V, m ⁻¹	Time, d	pH	[B] mg/L	NR(B), g/(m ² •d)	[Na] mg/L	NR(Na) g/(m ² •d)	[Si] mg/L	NR(Si), g/(m ² •d)
1	20	400	1	9.22	0.03	0.0045	1.93	0.032	0.261	0.0024
2	20	400	3	7.93	0.08	0.0040	3.28	0.018	0.38	0.0012
3	20	400	7	9.50	0.11	0.0024	2.84	0.0068	2.74	0.0036
4*	20	400	7	12.46	1.23	0.027	11.5	0.027	18.5	0.024
5	40	400	3	9.22	0.21	0.011	4.05	0.023	2.95	0.0089
6	40	400	7	9.78	0.39	0.0084	6.20	0.015	5.57	0.0072
7	20	1000	3	9.37	0.19	0.0038	6.28	0.014	1.22	0.0015
8	20	1000	7	9.93	0.23	0.0020	6.74	0.0064	2.13	0.0011
9*	20	1000	7	12.45	1.57	0.014	15.8	0.015	25.1	0.013
10	40	1000	3	9.79	0.45	0.0090	9.8	0.022	6.17	0.0075
11	40	1000	7	10.07	0.58	0.0050	11.8	0.011	8.41	0.0044
12	20	2000	3	9.75	0.30	0.0030	10.6	0.012	2.11	0.0013
13	20	2000	7	10.28	0.44	0.0019	13.5	0.0064	3.78	0.00098
14*	20	2000	7	12.44	1.85	0.0080	23.2	0.011	29.5	0.0077
15	40	2000	3	9.69	0.66	0.0066	17.7	0.020	8.81	0.0053
16	40	2000	7	10.48	0.85	0.0037	20.8	0.0099	11.8	0.0031
f _i						0.0166		0.150		0.275

* Tests conducted with pH 12 leachant solution.

2000 m⁻¹, respectively. The NL(B) and NL(Si) values were about four to twelve times higher and the NL(Na) values were about two to four times higher in tests conducted in the pH 12 solution than in tests conducted in demineralized water. While in qualitative agreement, the effects of temperature and pH in the PCTs are smaller than in dilute solutions (see below).

The average pH, B, Na, and Si concentrations of replicate 7-day PCTs conducted at 2000 m⁻¹ at 20 and 40°C are summarized in Table 3. For each test condition, the mean, standard deviation (s.d.), and percent relative standard deviation (%rsd) for each of three ICP-MS analyses of the leachates from nine replicate tests are given. These results show that the uncertainty in the Si concentration (as %rsd) is significantly higher than the uncertainties in the B and Na concentrations at both temperatures. Based on the variation in the results of triplicate ICP-MS analyses, the uncertainty in the test results is due primarily to the analysis of the dilute leachate solutions. That is, the difference in the mean values of analyses conducted on different days is usually greater than that in replicate tests measured on any single day. Day-to-day variations in the measured concentrations result from minor differences in the instrumental operating conditions, small variations in calibration for each element, etc. The impact of these minor differences are significant when the solution concentrations are low. Small variations in the execution of the test (mass determination, dilutions, small temperature differences within an oven, and intangibles) have a small effect compared to the uncertainty in the solution analysis for tests at low temperatures. Nevertheless, the difference in the concentrations of each element in tests at 20 and 40°C is greater than the analytical uncertainty, so a 7-day PCT at 2000 m⁻¹ is sensitive to differences in the dissolution rates at 20 and 40°C.

The results of replicate 7-day PCTs conducted at several laboratories at 90°C with the EA glass are included in Table 3 for comparison.⁸ The average results of 36 tests are presented. The %rsd values of the pH and element concentrations for these tests are less than those for the tests conducted at 20 and 40°C. This is probably due to the higher precision that is possible with the higher solution concentrations generated in those tests. The test precision that can be achieved at lower temperatures should be taken into account when specifying a benchmark level of performance in a LAW acceptance test.

The long-term behavior of LD6-5412 glass was characterized with PCTs conducted for up to 2 years. The range of final pH values measured in different test series are summarized in Table 4. The normalized mass losses (NL) based on B, Na, and Si for tests conducted at 2000 and 20,000 m⁻¹ are displayed in Fig. 1. All tests conducted at 20 and 40°C show the release of

Table 3. Summary of Results of Replicate PCT.

Analysis number	pH			[B], mg/L			[Na], mg/L			[Si], mg/L		
	mean	s.d.	%rsd	mean	s.d.	%rsd	mean	s.d.	%rsd	mean	s.d.	%rsd
LD6-5412 at 20°C												
1	8.84	0.72	8	0.629	0.051	8	15.2	0.92	6	5.22	1.47	28
2				0.416	0.030	7	14.6	0.78	5	5.14	1.48	29
3				0.440	0.038	9	13.4	0.97	7	8.27	2.21	27
avg.	8.84	0.72	8	0.495	0.10	21	14.4	1.16	8	6.21	2.25	36
LD6-5412 at 40°C												
1	9.72	0.57	6	1.38	0.14	10	29.1	1.66	6	15.6	3.51	22
2				1.22	0.065	5	28.7	1.21	4	15.9	3.58	23
3				1.28	0.072	6	27.5	1.36	5	16.0	3.69	23
avg.	9.72	0.57	6	1.29	0.12	9	28.4	1.54	5	15.8	3.46	22
EA at 90°C												
avg.	11.85	0.10	1	587	43	7	1662	112	7	893	86	10

Na to occur preferentially to the release of B and Si, which are released congruently with each other. The curves in Figs. 1a, 1b, and 1c are drawn through the NL(Na) and NL(B) results for tests at 20,000 m⁻¹ to show the trend over time. The dissolution behavior in short-term tests at 70 and 90°C is similar to that seen at 20 and 40°C, namely the values of NL appear to approach limiting values as glass components accumulate in solution. However, tests conducted for longer than about 500 days at 70°C and longer than about 100 days at 90°C show the NL values to increase significantly. The increase occurs coincidentally with the formation of zeolite alteration phases. Note that the y-axes in Figs. 1c and d are logarithmic, and that the values of NL(B) increase about 1000X after zeolites form. Examination of the solids with SEM and TEM indicate that zeolite phases similar to analcime and phillipsite form in these tests.⁹ The scatter in the 90°C results arises because the zeolites form after different reaction times in different tests, so dissolution is accelerated at different points in time in the different tests. Notice that the value of NL(Na) is greater than NL(B) before alteration phases form, but

Table 4. Range of Final pH Values for Long-Term PCT

	20°C	40°C	70°C	90°C
2000 m ⁻¹	9.6-10.3	10.2-11.0	10.5-11.7	10.8-12.0
20,000 m ⁻¹	10.6-11.4	11.0-12.0	11.7-12.3	11.9-12.4

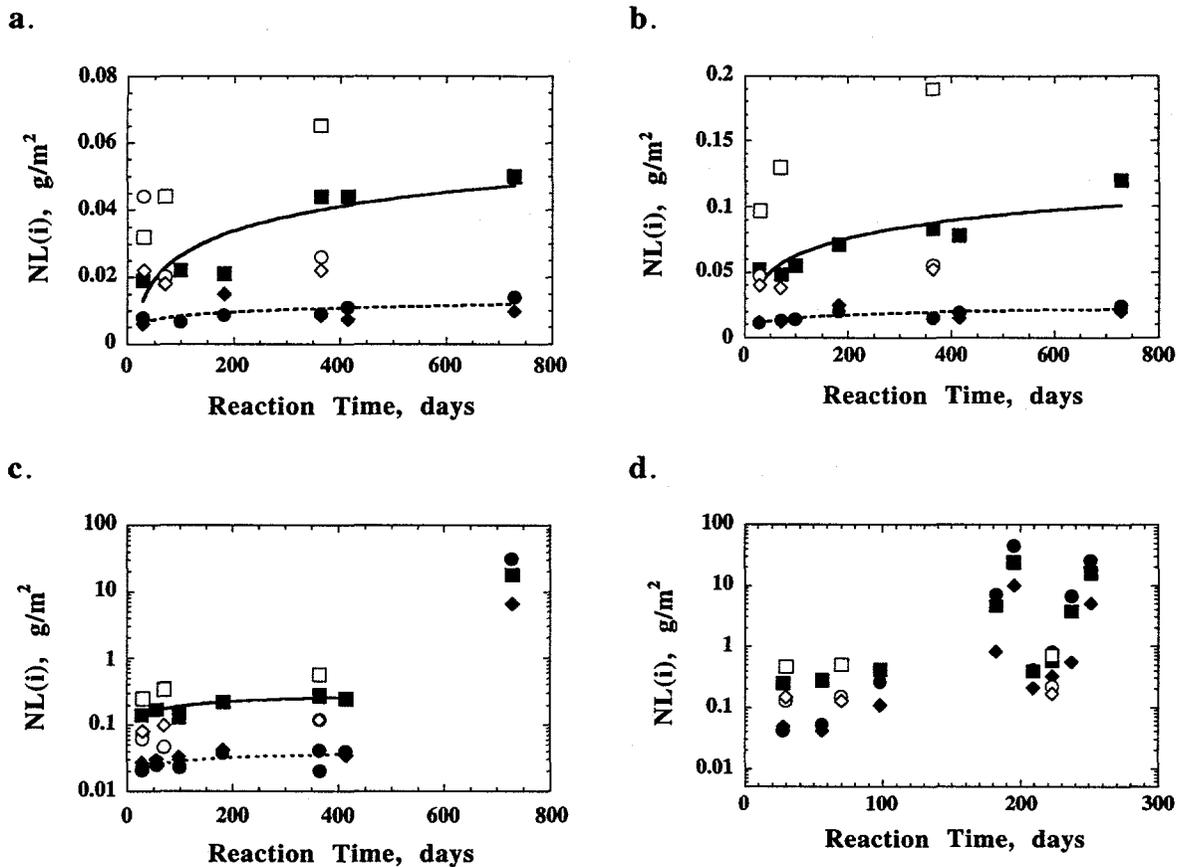


Figure 1. Results of PCT at (a) 20, (b) 40, (c) 70, and (d) 90°C. Open symbols for tests at 2000 m⁻¹, filled symbols for tests at 20,000 m⁻¹: NR(B)-(O, ●); NR(Na)-(□, ■); and NR(Si)-(◇, ◆). Curves show trends in NL(Na) (solid curve) and NL(B) (dotted curve) in tests at 20,000 m⁻¹ prior to alteration phase formation.

that NL(B) is greater than NL(Na) after alteration phases form. This is because some of the Na (and Si) that is released from the glass becomes incorporated into the alteration phases, while essentially all the B is dissolved.

As the glass dissolves, the surface area of glass that is available for continued reaction decreases significantly.¹⁰ In fact, almost all of the available glass had reacted in some tests in which zeolites formed. The decrease in the surface area of the glass affects the calculation of NL in Eq. 1. The normalized mass loss values in Fig. 1 were calculated with the average of the initial and final surface areas to account for the loss of surface area during the test. The final surface area was calculated based on the boron concentration. Only the NL values for tests in which alteration phases formed were significantly affected by this adjustment.

DISCUSSION

The kinetics of glass dissolution are known to depend primarily on the glass composition, temperature, solution pH, and the activity of orthosilicic acid.¹⁰ Hydrolysis of silane bonds is the rate-controlling reaction for network dissolution; these bonds account for about half of the glass network bonds in LD6-5412 glass. An expression used to calculate the glass corrosion rate in computer simulations is¹⁰

$$\text{rate} = k_0 10^{b \cdot \text{pH}} e^{-E_a/RT} \{1 - (Q/K)\} \quad (2)$$

where k_0 is the rate coefficient, b is the power law coefficient for the pH, E_a is the activation energy, R is the gas constant, T is absolute temperature, Q is the ion activity product, and K is the equilibrium constant of the rate-controlling phase. The term in braces is usually referred to as the reaction affinity term. The values of Q and K are usually approximated with the activity of orthosilicic acid and the equilibrium constant of a surrogate phase for the glass, such as chalcedony, respectively.¹¹ Values of k_0 , b , and E_a can be determined experimentally under test conditions in which the value of (Q/K) is maintained near zero. These values were measured to be $b = 0.4$, $E_a = 85$ kJ/mol, and $k_0 = 10^{8.87}$ g/(m²•d) for LD6-5412 glass.¹²

The accumulation of dissolved glass components in the PCTs has a strong effect on the glass dissolution rate and moderates the effects of temperature and pH. The use of different S/V ratios provides some control of how fast the concentrations of dissolved glass components and the pH increase. Note that the S/V ratio does not occur explicitly in the rate expression; it is solely a testing parameter. The S/V ratio will affect the pH and the value of Q in Eq. 2.

The very high ionic strength of the PCT leachate solutions complicates direct application of Eq. 2 to these test results. However, the influence of the affinity term can be estimated by a simple, empirical analysis of the present test results. The average dissolution rates calculated from the averages of replicate 7-day tests with LD6-5412 glass at 20 and 40°C are summarized in Table 5. The results of 90°C PCTs run at PNNL are included in the table. Consider first the reactivities in 7-day 2000 m⁻¹ tests conducted at 20 and 40°C. Dissolution occurs 2.5, 2.0, and 2.6 times faster at 40°C than at 20°C based on the release of B, Na, and Si, respectively. Based on an activation energy of 85 kJ/mol, the rate at 40°C should be about nine times greater than the rate at 20°C. The slightly higher final pH value of the 40°C test also

contributes to the increase in the dissolution rate. From Eq. 2, a one unit increase in the pH causes a 2.5X increase in the dissolution rate. The difference in the rates of the PCTs at 20 and 40°C is only about one-third that expected based on the individual effects of temperature and pH. This is because of the effect of the affinity term. Likewise, dissolution in PCTs occurs only about four times faster at 90°C than at 40°C, while the rate at 90°C is predicted to be about 90X higher than the rate at 40°C due to temperature alone. Part of the increase in the rate of PCTs is due to the higher pH. The measured difference in the rates of from PCTs conducted at 40 and 90°C is much smaller than that predicted based on the temperature and pH differences because the effect of the affinity term in Eq. 2 is very large in PCTs. Consideration of the other results summarized in Tables 1 and 5 indicates that the influence of the affinity term on the rate measured with a PCT increases with test temperature, duration, and S/V ratio.

The average dissolution rates calculated from the measured concentrations of B, Na, and Si accumulated over the entire duration of long-term PCTs run at 2000 and 20,000 m⁻¹ are plotted against the reaction time in Fig. 2. The horizontal lines show the rates calculated from 7-day PCT at 2000 m⁻¹ (see Table 5). For all tests at 20 and 40°C, the average rates from the longer-term tests are much lower than the average rate from the 7-day PCTs conducted at the same temperature. The dissolution rates in tests run at 70 and 90°C are less than the 7-day PCT rates before zeolite alteration phases form (i.e., before about 500 and 100 days, respectively), but the rates calculated for some tests in which alteration phases formed are greater than the 7-day PCT rate. Notice that the values of NR(Si) for the long-term tests remain less than the values of NR(Si) for the 7-day test. This is because much of the silicon released from the glass becomes incorporated into alteration phases.

Table 5. Measured Average Dissolution Rates from Replicate 7-day PCT at 2000 m⁻¹.

Temperature, °C	pH	NR(i), g/(m ² •d)		
		NR(B)	NR(Na)	NR(Si)
20 ^a	10.28	0.0018	0.0060	0.0016
40 ^a	10.48	0.0046	0.012	0.0042
70	-	0.011 ^b	0.034 ^b	0.0098 ^b
90 ^c	11.39	0.016	0.054	0.015

^a Average of replicate tests conducted at ANL.

^b Interpolated from 40 and 90°C results.

^c Average of duplicate tests conducted at PNNL.⁷

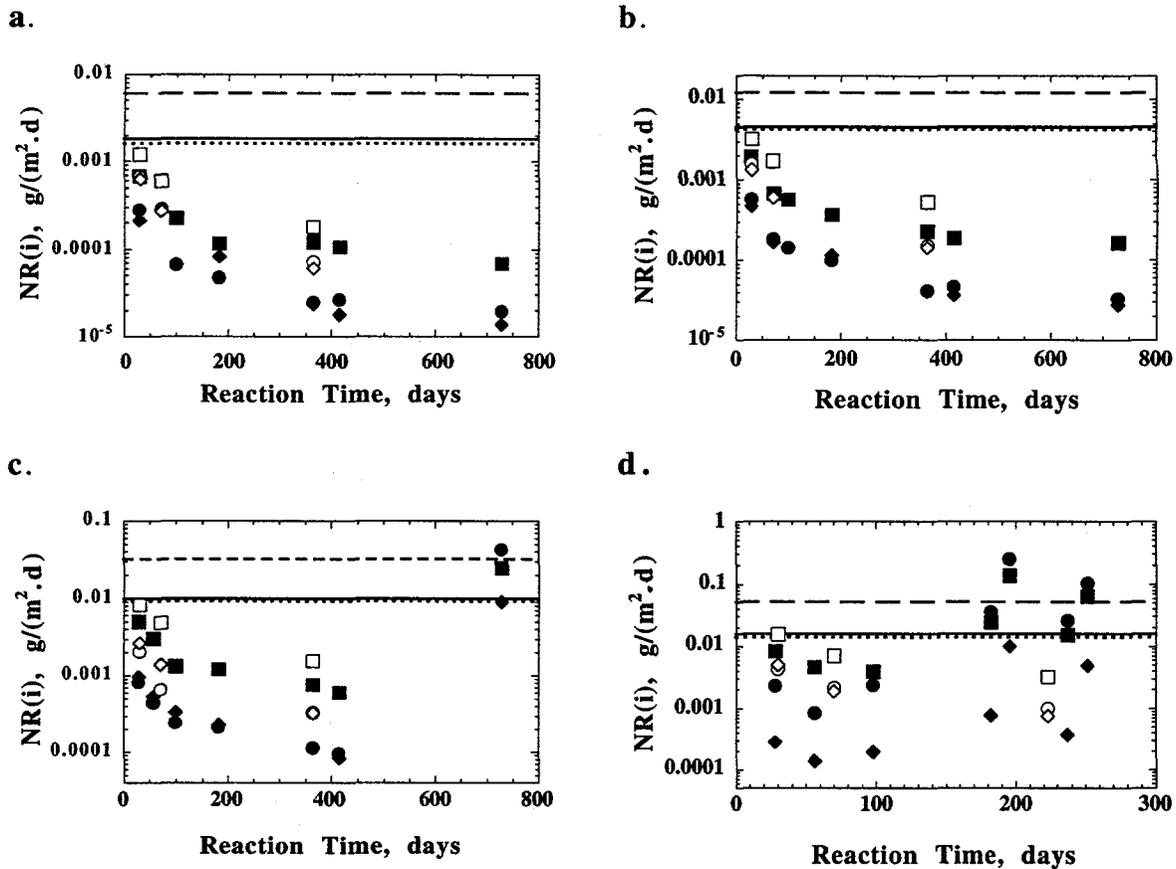


Figure 2. Results of long-term PCT at (a) 20°C, (b) 40°C, (c) 70°C, and (d) 90°C. Open symbols for tests at 2000 m^{-1} , filled symbols for tests at 20,000 m^{-1} : NR(B)-(O, ●); NR(Na)-(□, ■); and NR(Si)-(◇, ◆). Lines show rates from 7-day tests: NR(B)-(solid); NR(Na)-(dashed); and NR(Si)-(dotted).

Slightly higher average dissolution rates were measured in the tests at 2000 m^{-1} than in tests at 20,000 m^{-1} before alteration phases formed, although the rates in tests at 2000 m^{-1} were all much lower than the 7-day PCT rates. This is because more concentrated solutions are generated in the tests at 20,000 m^{-1} than at 2000 m^{-1} , so that the values of the affinity terms in tests at 20,000 m^{-1} are significantly less than the values in tests at 2000 m^{-1} . This simply means that the tests at 20,000 m^{-1} represent a greater extent of corrosion than tests at 2000 m^{-1} . The S/V ratio used in a test does not itself represent a particular disposal environment. Rather, it is the solution chemistries attained in tests at different S/V ratios that must be compared to the chemistries attained in a disposal site. Whether or not alteration phases eventually form in tests conducted at 2000 m^{-1} is irrelevant. What is important is the fact that when certain alteration phases form, the glass dissolution rate increases to a value that may be higher than the value measured in a 7-day PCT. Whether or not solutions in a disposal environment can become

concentrated enough that those phases can form must be determined by coupling knowledge of glass dissolution behavior with site characteristics.

Finally, it is important to distinguish the average dissolution rates shown in Fig. 2 from the rates at which the glass is actually dissolving at a particular point in time. Several PCTs conducted for different durations are required to determine whether the rate is increasing, decreasing, or constant at a particular time. The dissolution rates measured in tests conducted for times less than 7 days and at S/V ratios less than 2000 m^{-1} show that the dissolution rate is initially very much higher than the rate measured in a 7-day PCT at 2000 m^{-1} , while the rates in PCTs conducted for longer than 7 days continue to decrease as the affinity term becomes smaller until the formation of alteration phases changes the dissolution rate, and perhaps the rate-controlling phase. The formation of certain alteration phases increases the dissolution rate by providing a sink for dissolved silicon that increases the reaction affinity. In a disposal site, the presence of other materials may also affect the long-term corrosion behavior of waste glasses. Short-term tests may not provide an upper bound to the glass dissolution rate if alteration phases form or if other phenomena affect the solution chemistry. How the formation of silicon-consuming phases affects the glass corrosion behavior must be better understood and accounted for in model calculations to forecast long-term behavior of waste forms.

CONCLUSIONS

Several PCTs were conducted at different S/V ratios, temperatures, and durations to measure the effects of these variables on the dissolution rate and the reproducibility of a 7-day test. The dissolution rates measured in a 7-day tests were also compared with the rates measured in tests conducted for longer durations. The following conclusions were reached from this study. A 7-day PCT at a water volume/glass mass ratio of 10 mL/g (S/V ratio of about 2000 m^{-1}) can be used to distinguish the reactivity of LD6-5412 glass at 20 and 40°C . The precision of a 7-day PCT decreases with the test temperature and is limited by analytical uncertainties. The average dissolution rates measured at 20 and 40°C are 0.0018 and $0.0046 \text{ g}/(\text{m}^2\cdot\text{d})$ based on the release of boron, and 0.0016 and $0.0042 \text{ g}/(\text{m}^2\cdot\text{d})$ based on the release of silicon. The effects of temperature and pH on the glass dissolution rate in PCTs are much smaller than the effects measured under dilute test conditions because of the competing effect of the orthosilicic acid buildup (i.e., the effect of the affinity term) in the PCTs. The dissolution rate of LD6-5412 glass measured in a 7-day PCT does not provide an upper bound for long-

term corrosion at 70 or 90°C after zeolite alteration phases form. It is uncertain whether alteration phases will form or affect the dissolution rate at lower temperatures.

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