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ABSTRACT

We are characterizing the corrosion behavior of the radioactive glass that was made with sludge from Tank 51 at the Defense Waste Processing Facility (DWPF) and a nonradioactive glass having the same composition, except for the absence of radionuclides. Static dissolution tests are being conducted in a tuff groundwater solution at glass surface area/solution volume ratios (S/V) of 2000 and 20,000 m^{-1}. These tests are being conducted to assess the relationship between the behavior of this glass in a 7-day Product Consistency Test and in long-term tests, to assess the effects of radionuclides on the glass corrosion behavior, and to measure the disposition of radionuclides that are released as the radioactive glass corrodes. The radioactive glass reacts slower than the nonradioactive glass through the longest test durations completed to date, which are 140 days for tests at 2000 m^{-1} and about 400 days for tests at 20,000 m^{-1}. This is probably because radiolysis results in lower solution pH values being maintained in tests with the radioactive glass. Rate-affecting alteration phases that had formed within one year in tests with other glasses having compositions similar to the Tank 51 glass have not yet formed in tests with either glass.

INTRODUCTION

We are conducting static dissolution tests with a glass made at the Savannah River Technology Center (SRTC) during a demonstration of the Defense Waste Processing Facility (DWPF) process control for remote vitrification [1]. The glass was made with sludge from Tank 51, SRL 202 frit, and added soda. This glass is similar to waste glasses being made in the current DWPF campaign. Parallel tests are being conducted with a nonradioactive glass made at ANL having the same composition as the radioactive glass, except without the radionuclides. The radioactive and nonradioactive glasses are referred to as 51R and 51S, respectively. The results of these tests provide information pertinent to assessing the long-term corrosion behavior of DWPF glasses, comparing the corrosion behaviors of radioactive and nonradioactive glasses, and characterizing the disposition of radionuclides as the glass corrodes.

An important effect in static tests with radioactive glasses is acidification of the leachate solution through the generation of nitric acid by the radiolysis of moist air [2]. Short-term tests with radioactive glasses and their nonradioactive homologues have generally shown the effects of radiolysis to be minor and the corrosion behaviors of radioactive and nonradioactive homologue glasses to be similar [3]. This is because the pH rise that accompanies the initial dissolution of the glass neutralizes most of the radiolytically produced acid. The slight differences in the reactivities that were observed in those tests can be attributed to minor differences in the compositions of the radioactive and nonradioactive glasses. However, the dissolution rate of a nonradioactive glass increased significantly after certain alteration phases formed in long-term static dissolution tests, while similar phases did not form in tests with a radioactive glass having a similar composition [4]. While similar alteration phases may not have formed in tests with the radioactive glass because of small differences in the glass compositions, the formation of those phases may also have been delayed or prevented by radiolytic effects. The compositions of the 51R and 51S glasses discussed in this paper are much more similar than the sets of radioactive and nonradioactive glasses used in previous tests, and the effects of glass composition and radiolysis on the corrosion behavior can be better distinguished.

Insight into the expected corrosion behavior of the 51R and 51S glasses was gained from tests that were conducted previously with actinide-doped SRL 202 frit-based glasses having similar compositions [5]. Radiolysis did not prevent the formation of rate-affecting alteration phases in

those tests. It was found that the increase in the glass dissolution rate resulting from phase formation did not lead to an increase in the amounts of actinides in the aqueous phase, although the amounts of Pu and Am fixed to the steel reaction vessel increased [6]. The actinide concentrations in the solutions were probably controlled by phases that were sparingly soluble, although the actinide-bearing solid phases were not identified. The effects of radiolysis are expected to be more pronounced in the 51R glass than in that actinide-doped glass.

The solution chemistries of the various radionuclides will affect their distribution between dissolved, colloidal, precipitated, and adsorbed fractions during glass corrosion. The disposition of radionuclides as the glass corrodes in a disposal site is important because it will impact their dispersal. For example, radionuclides that are dissolved or associated with colloidal material may be transported by groundwater flow, while radionuclides that form precipitates or become fixed to other materials, such as steel containers, may become immobilized, depending on the stability of those materials. Several studies have shown the general order of actinide release from high-level waste glasses into solution in laboratory tests to be U, Np > Pu, Am, Cm [7]. The solubilities of these elements will be affected by the pH and Eh of the solution and by the presence of ligands. Therefore, it is important that laboratory tests be conducted with solutions that are relevant to the groundwater chemistry anticipated at the disposal site. The tuff groundwater solution used in the tests discussed in this paper was prepared from the reference groundwater for the Yucca Mountain Site. It contains about 120 mg/L HCO_3^- and has a pH of about 8, and so both the formation of carbonates and hydrolysis will affect the solution chemistries of some actinides. Other species in the groundwater and anions released from the glass, such as F^-, HPO_4^{2-}, and SO_4^{2-}, may also affect the speciation of some actinides.

EXPERIMENTAL

A sample of the 51R glass was obtained from SRTC and the 51S glass was produced at ANL from reagent-grade chemicals. The compositions of the two glasses are given in Table I. The difference in the concentration of each element in the glasses is within the combined analytical uncertainties for analyses of 51R and 51S glasses, which is less than 15% for most components. The biggest difference is in the U contents of the two glasses, which differ by about 11%. Static dissolution tests were conducted similar to the Product Consistency Test (PCT) [8]. Samples of each glass were prepared for testing by crushing and sieving to isolate the -100 +200 mesh fraction (74 -149 μm). The crushed glass was washed with deionized water to remove fines then dried in a 90°C oven. Tests were conducted at glass-to-water mass ratios of 1:10 and 1:1 to achieve glass surface area/solution volume ratios (S/V) of about 2000 and 20,000 m^{-1}, respectively. Most tests were conducted with tuff groundwater from well J-13 at the Nevada Test Site that had been pre-reacted with pulverized tuff at 90°C. The groundwater solution is referred to

Table I. Compositions of the 51S and 51R Glasses, in wt %

Element	51S	51R[a]	Element	51S	51R[a]
Al	2.55	2.56	Ni	0.23	0.23
B	1.99	2.00	P	0.26	0.26
Ca	0.99	0.99	Si	24.4	24.5
Cr	0.30	0.30	Th	0.02	0.02
Fe	9.17	9.21	U	0.86	0.97
K	1.30	1.31	Zn	0.11	0.11
La	0.47	0.47	^{241}Am	—[b]	7.4 x10^{-5}
Li	2.09	2.10	^{244}Cm	—	1.1x10^{-5}
Mg	1.24	1.24	^{237}Np	—	4.2 x10^{-4}
Mn	0.97	0.98	^{238}Pu	—	1.1 x10^{-4}
Na	6.93	6.96	^{239}Pu	—	2.2 x10^{-3}

[a]From Ref. [1].
[b]Not present in glass.

as EJ-13. It has a pH of about 8 and its composition, in mg/L, is Al : 1.4, B: 0.23, Ca: 3.2, Fe:0.13, K: 5.9, Li: 0.057, Mg: 0.035, Mn: 0.10, Na: 48, Si: 41.1, F: 1.8, Cl: 8.0, NO_3: 9.9, HPO_4^{2-}: <2.0, SO_4^{2-}: 20.1, and HCO_3 : 120. Tests with EJ-13 water are being conducted for reaction times from 7 days to longer than 560 days. Tests were also conducted for 7 days at 2000 m^{-1} in deionized water for comparison to similar tests conducted with 51R glass at SRTC. All tests were conducted in Type 304L stainless steel vessels.

Both the leachate solutions and reacted solids were analyzed to characterize the corrosion behavior of each glass. The solutions were analyzed for pH with a combination electrode, anions with ion chromatography, and cations with inductively coupled plasma mass spectrometry (ICP-MS). The radionuclide concentrations in the solutions were measured with ICP-MS and also with alpha and gamma spectroscopy. Aliquots of unfiltered leachate and solution that passed through 1000, 450, 100, 10, and 6 nm filters were analyzed with alpha spectroscopy. An acid-strip solution of the vessel was also analyzed with alpha and gamma spectroscopy.

Some of the reacted glass samples were analyzed with scanning electron microscopy (SEM) and transmission electron microscopy (TEM). Both the SEM and TEM were equipped with energy dispersive X-ray spectrometers (EDS) for compositional analysis. Selected area electron diffraction (SAED) was also performed on some TEM samples.

RESULTS AND DISCUSSION

Tests in Deionized Water —The solution concentrations C(i) and normalized elemental mass losses NL(i) of B, Na, and Si in 7-day PCTs conducted in deionized water at 2000 m^{-1} at ANL and at SRTC are summarized in Table II. The normalized mass loss is calculated by dividing the difference of the measured solution concentration and the background concentration of component i by the S/V of the test and the mass fraction of i in the glass. The results of tests conducted at SRTC were only reported as concentrations, and we have calculated NL(i) values by assuming an S/V of 2000 m^{-1} for those tests.

We first compare the results of tests with 51S and 51R glass conducted at ANL. The measured pH values of tests with 51S glass are consistently higher than those of tests with the 51R glass. Likewise, the values of NL(B) and NL(Na) are higher in tests with 51S glass than in tests with 51R glass. In contrast, the values of NL(Si) are lower in tests with 51S glass than in tests with 51R glass. Dissolution of the 51R glass results in nearly stoichiometric releases of B, Na, and Si, while the 51S glass shows B and Na to be released preferentially to Si.

Next, we compare the results of tests with 51R glass conducted at ANL and SRTC. More 51R glass is measured to have dissolved in tests conducted at SRTC than in tests conducted at ANL based on pH, NL(B), and NL(Na). The values of NL(Si) agree within the overall experimental uncertainty, which is assumed to be about 15%. The differences in the results of tests conducted at SRTC and at ANL may be due to small differences in the compositions of the samples of 51R glass that were tested, differences in the amounts of remaining fines of the crushed glass tested, small differences in the oven temperatures (the PCT procedure specifies a temperature of 90±2°C), etc. For example, slightly different results were obtained from PCTs conducted at SRTC with glass taken from the top and bottom of the pour can [9]. However, the differences in the results are similar to the interlaboratory differences seen previously in tests with radioactive glass [10].

The results of 7-day tests conducted with EA and SRL 202 glasses are included in Table II for comparison. The PCT behavior of the EA glass is the benchmark for high-level waste glasses, and the SRL 202 glass is a reference "blend" glass for the DWPF having a composition similar to that of 51S and 51R that has been tested extensively at ANL [5]. The values for SRL 202 in Table II are from tests in Teflon vessels. The pH values of these tests may have been lowered slightly due to the influx of CO_2, although the tests were conducted in a water bath to minimize that effect. Similar values of NL(B), NL(Na), and NL(Si) are measured in tests with 51R, 51S, and SRL 202 glasses; all are significantly lower than the values measured in tests with the EA glass.

Table II. Results of PCT-A with 51S, 51R, EA, and SRL 202 Glasses

Glass	pH	NC(i), mg/L			NL(i), g/m^2		
		NC(B)	NC(Na)	NC(Si)	NL(B)	NL(Na)	NL(Si)
51S	10.4	14.9	47.3	86.2	0.3	0.3	0.2
51S	10.4	14.6	40.8	87.2	0.3	0.3	0.2
51S	10.3	12.9	40.2	85.7	0.3	0.3	0.2
51R	9.8	9.10	31.0	95.0	0.2	0.2	0.2
51R	9.9	8.80	28.0	94.0	0.2	0.2	0.2
51R	9.9	9.00	27.0	96.0	0.2	0.2	0.2
SRTC 51R[a]	10.3	14.0	44.0	102	0.4	0.3	0.2
SRL 202[b]	1.0	14.0	45.0	94.0	0.3	0.3	0.2
EA[a]	11.9	587	1662	893	8.5	6.7	2.0
EA[c]	11.7	629	1798	999	8.7	7.3	2.2

[a]Average of replicate tests conducted at SRTC [9].

[b]Average of duplicate tests [5].

[c]Average of replicate tests conducted at ANL.

Tests in EJ-13 Water — We now compare the results of tests conducted with EJ-13 water. Tests with 51R and 51S glasses have been completed through 280 and 393 days, respectively; the tests with 51S have progressed to longer times because that glass was available for testing before the 51R glass. The average of the pH values attained in duplicate tests with the 51R and 51S glasses are plotted in Fig. 1. The pH values in tests at 20,000 m^{-1} are consistently higher than those in tests at 2000 m^{-1}. The pH values appear to reach nearly constant values after about 100 days of reaction for tests with both glasses at both S/V.

At a given S/V, the measured pH values for tests with 51R glass are consistently lower than those for corresponding tests with 51S glass. This reflects differences in the dissolution behaviors of the two glasses, the effects of radiolysis, and small compositional differences, including the effects of anions that may be present in the 51R glass but not in the 51S glass. While most waste glasses and sludges are not analyzed for the presence of anions, significant amounts of F$^-$, Cl$^-$, SO$_4^{2-}$, and HPO$_4^{2-}$ have been observed to be released as other sludge-based glass dissolved [4]. Since anion results for the present tests are not yet available, we estimated the effects radiolysis and the presence of anions on the pH. Geochemist's Workbench^{TM} software (GWB) was used to simulate the dissolution of glasses in which the anion concentrations were varied from 0.2 wt% to 2 wt%. These simulation indicated that the presence of these anions had little effect on the pH. Likewise, the small differences in the cation concentrations of the 51S and 51R glasses had a negligible effect. The effects of radiolysis were estimated with the Burns equation [11] using G values from the literature [12]. The results of those calculations indicate that radiolysis accounts for only a small percentage of the measured difference in the final pH. We note that these calculations show that radiolysis causes a negligible change in the amount of N$_2$ in the vessel, even after several years. While the concentrations of anions simulated to be in the leachates must be confirmed with their measured concentrations, it appears that radiolysis and the release of anions do not have a significant direct effect on the pH. Instead, it is likely that the small pH difference due to radiolysis affects the extents of ion exchange and hydrolysis that occurs. This is discussed below.

It is likely that differences in the reactivities of radioactive and simulated glasses that have been attributed to radiolysis or compositional effects in the past may have also been affected by differences in glass melts and sample preparation. This is especially important because of the extent to which nonradioactive homologue glasses have been used as surrogates for radioactive glasses. While further work is needed to understand these differences, we note that the reactivity of the radioactive 51R glass is less than that of the nonradioactive 51S glass, so that, at least for these glasses, the nonradioactive glass is a conservative surrogate.
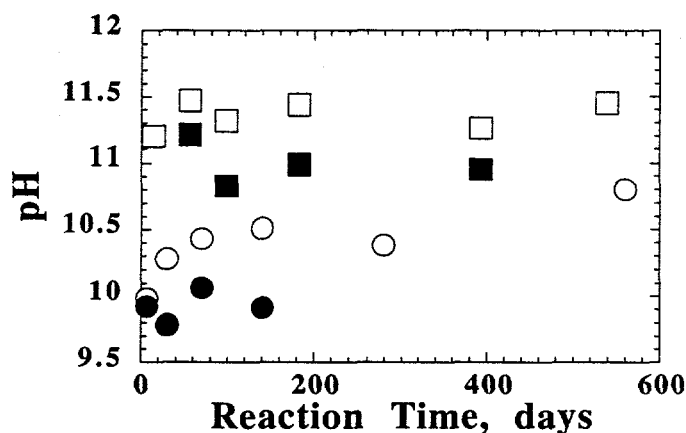
Figure 1. Measured pH vs. Reaction Time for Tests with 51R at 2000 m⁻¹ (●) and 20,000 m⁻¹ (■), and with 51S at 2000 m⁻¹ (○) and 20,000 m⁻¹ (▫).

The values of NL(B), NL(Na), and NL(Si) for tests conducted with 51S and 51R glasses at 2000 m^{-1} are plotted in Fig. 2a. Uncertainty bars are only shown for the Na results for clarity. The uncertainties in NL(B), NL(Na), and NL(Si) (and other components) are all dominated by the assumed ±10% analytical uncertainty. The values of NL(Na) are significantly higher in tests with 51S glass than in tests with 51R glass, while the values of NL(B) are the same in tests with the two glasses, within experimental uncertainty, as are the values of NL(Si). The lower values of NL(Na) in the tests with the 51R glass are consistent with the lower pH values in those tests. Thus, it appears that less ion exchange to release Na (and also Li and Cs) occurs in tests with 51R glass than in tests with 51S, but the extent of hydrolysis to release B and Si are about the same for both glasses. The lower extent of ion exchange in tests with 51R glass is consistent with the lower final pH in these tests, compared to tests with 51S glass.

The results of tests conducted at 20,000 m^{-1} are shown in Fig. 2b. The average of duplicate test results are shown and the difference between the duplicates is usually within the ±10% analytical uncertainty. As in the tests conducted at 2000 m^{-1}, the values of NL(Na) are higher in tests with 51S glass than in tests with 51R glass over the first 100 days, although the difference is smaller at longer test durations. The values of NL(B) increase with time in tests with 51S glass at a rate of about 0.002 g/(m^2•d), while the values of NL(Na) remain nearly constant.

The values of NL(i) are consistently higher in tests conducted at 2000 m^{-1} than in tests conducted at 20,000 m^{-1} for both glasses. This is because the Si concentrations are higher in tests at 20,000 m^{-1} than in tests at 2000 m^{-1}, and the dissolution rates decrease with increasing concentrations of orthosilicic acid. For example, the Si concentrations measured in tests with 51S glass at 2000 m^{-1} increased from 75 mg/L after 7 days to about 150 mg/L after 282 days, and from about 250 mg/L after 14 days to about 470 mg/L after 393 days in tests with 51S glass at 20,000 m^{-1}. Although a smaller proportion of the Si concentration is orthosilicic acid at the higher pH values attained in tests at 20,000 m^{-1}, speciation calculations show that more orthosilicic acid is present in the 20,000 m^{-1} tests.

The speciation calculation raises another interesting point. Notice in Fig. 2b that the values of NL(Si) for the tests with 51R and 51S glasses are about the same, while the pH values for the tests with 51R glass are lower than for tests with 51S glass. This means that less Si is present in solution as H_4SiO_4 in tests with 51S glass than with 51R glass. The chemical affinity for corrosion of the 51S glass is, therefore, higher than that for corrosion of the 51R glass, and the dissolution rate should be higher. However, the values of NL(B) become higher in tests with 51R glass than in tests with 51S glass after about 182 days, contrary to what is expected.

Clay Layer— Examination of reacted glass samples with TEM shows the formation of a thin clay layer at the glass surface. Images of 51S and 51R samples reacted at 20,000 m^{-1} for 98 and 182 days, respectively, are shown in Fig. 3. The layers formed on samples of each glass are
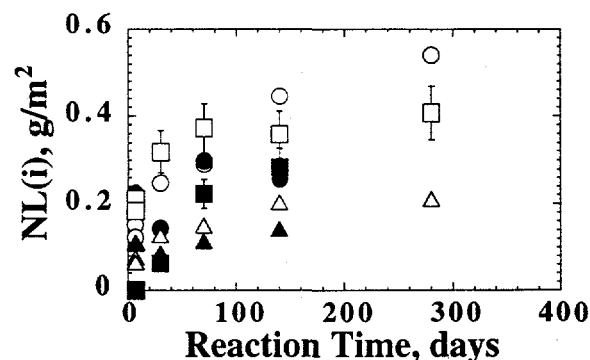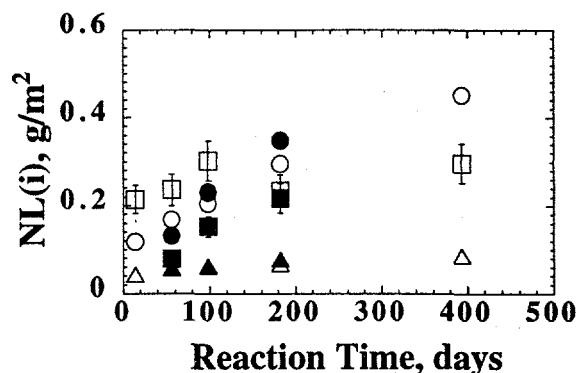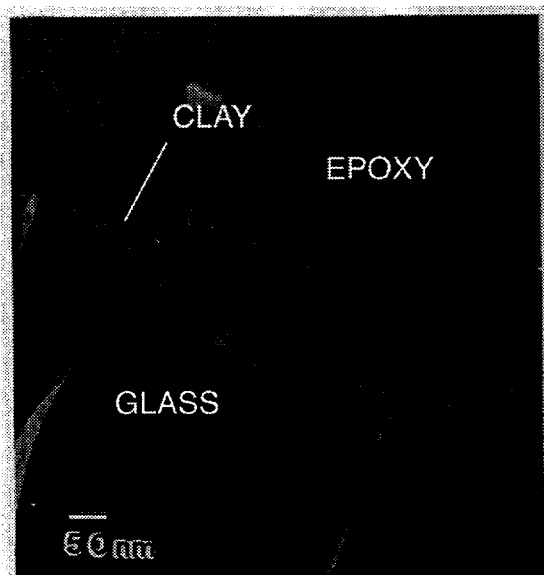
Fig. 2. Normalized Mass Loss vs. Reaction Time for PCTs in EJ-13 at (a) 2000 m⁻¹ and (b) 20,000 m⁻¹: NL(B) (● and O), NL(Na) (■ and □), and NL(Si) (▲ and Δ); filled symbols for tests with 51R glass; open symbols for tests with 51S glass.

Fig. 3. TEM Photomicrographs of (a) 51S Glass Reacted at 20,000 m⁻¹ for 98 Days, and (b) 51R Glass Reacted at 20,000 m⁻¹ for 182 Days

composed of wisps of clay crystallites, and are less than about 200 nm thick. Although the layers formed in tests completed to date are too thin to be characterized with EDS or electron diffraction, they appear to be identical to the layers of smectite clay that formed during the corrosion of other DWPF glasses [4, 12]. Thicker layers are expected to form in tests conducted for longer durations; these layers will be examined to determine if radionuclides accumulate in the layer.

Radionuclide Disposition —We have measured the amounts of several actinide elements that were dissolved, suspended in the solution, or fixed to the steel reaction vessel. We anticipate measuring the amounts of radionuclides in the clay layer and in alteration phases when samples reacted for longer durations that have thick alteration layers become available. Aliquots of some leachates were sequentially filtered through 1000, 100, 10, and 6 nm filters to measure the size distribution of suspended material. The remaining leachate was filtered with 450 nm filters; that

solution was also analyzed for pH, anions, and other cations. The amounts of Np, Pu, and Cm measured to have passed through the different filter sizes, and the amounts measured in the solution from an acid strip of the vessel are summarized in Table III. Although there is significant scatter in the data, the amounts of all nuclides in all filtrates generally increase with time in tests conducted at 2000 m^{-1}. Most of the Pu is removed from the solution by a 10 nm pore size filter. Quantification of Np with α-spectroscopy is complicated by severe overlap of the Np peak with the tail of the Pu peak. We estimate the uncertainties of the values presented for Np to be on the order of 50%, and the uncertainties in the values of Pu or Cm to be on the order of 25%. The higher uncertainty of the Np results is evidenced by the conflicting results obtained after filtering with 10 and 6 nm filters. Since the measured Np concentrations after filtration with a 10 nm filter are significantly higher than the concentrations measured after the solution was filtered with either 450 or 100 nm filters, these are less reliable than the results of the 6 nm filtration step.

Most of the Np and Pu that is present in the leachate exists in a form that can be removed by filtering with a 10 nm filter; only a small amount of Np and Pu is dissolved. Tests with the SRL 202 glass showed that colloidal material was present in the leachates of tests run for short times, but that the colloids flocculated and fell out of solution after longer test durations [5]. The decrease in the amounts of Np and Pu in the 1000 and 450 nm filtrates between 70 and 140 days may indicate that flocculation is occurring in these tests, as well. Longer test durations should reveal whether or not the suspended material settles out of solution. The amounts of each nuclide measured to be present in the solution (i.e., either suspended or dissolved) and fixed to the steel vessel are similar.

Table III. Masses of Actinides Measured after Filtration (Np and Pu in ng; Cm in pg)

Time, days	pH	Nuclide	Filter Pore Size					Acid Strip
			1000 nm	450 nm	100 nm	10 nm	6 nm	
Tests at 2000 m^{-1}								
7	9.8	Np-237	—[a]	20	—	—	—	54
		Pu-239	—	3.8	—	—	—	2.6
		Cm-244	—	0.01	—	—	—	0.00
	9.8	Np-237	—	90	—	—	—	11
		Pu-239	—	8.5	—	—	—	5.7
		Cm-244	—	0.05	—	—	—	0.05
70	10.1	Np-237	600	700	600	1100	60	490
		Pu-239	59	44	22	1.0	1.9	33
		Cm-244	0	0.28	0	0	0.29	0.10
140	10.4	Np-237	1600	300	800	1000	200	2100
		Pu-239	29	22	49	1.0	4.5	59
		Cm-244	0	0	0	0	0	0.03
Tests at 20,000 m^{-1}								
14	11.2	Np-237	—	0	—	—	—	50
		Pu-239	—	72	—	—	—	3.2
		Cm-244	—	0	—	—	—	0.03
56	10.9	Np-237	—	0	—	—	—	60
		Pu-239	—	0	—	—	—	4.9
		Cm-244	—	0.06	—	—	—	0.06
98	11.0	Np-237	6200	20	—	—	—	40
		Pu-239	72	0.21	—	—	—	11
		Cm-244	0	0.01	—	—	—	0.32

[a] "—" indicates that a particular filtration or analysis was not performed.

CONCLUSIONS

Static dissolution tests are in progress to characterize the long-term corrosion behavior of a glass made with sludge from Tank 51 at the DWPF (51R) and a nonradioactive homologue glass (51S). The compositions of the glasses are essentially identical, except for the radionuclides, so that differences in the corrosion behavior of the two glasses are due to the effects of the radionuclides. Tests completed to date indicate that corrosion of the 51R glass results in slightly lower solution pH values and slightly lower B and Na concentrations than does corrosion of the 51S glass. The difference is probably due to radiolysis effects in the tests with 51R glass, although this remains to be confirmed. Most of the Np and Pu released from the glass is present in the solution as suspended material that can be removed by filtration with a 6 nm filter. Only small amounts of Np, Pu, and Cm are dissolved in the solution. Thin surface layers that are similar in appearance to clays formed during the corrosion of other DWPF glasses have formed during the corrosion of both glasses; these layers were too thin (about 200 nm) to be analyzed to determine if they retained actinides.

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