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DISPOSITION OF ACTINIDES RELEASED FROM HIGH-LEVEL WASTE GLASS

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INTRODUCTION

A series of static leach tests was conducted using glasses developed for vitrifying tank wastes at the Savannah River Site to monitor the disposition of actinide elements upon corrosion of the glasses. In these tests, glasses produced from SRL 131 and SRL 202 frits were corroded at 90°C in a tuff groundwater. Tests were conducted using crushed glass at different glass surface area-to-solution volume (S/V) ratios to assess the effect of the S/V on the solution chemistry, the corrosion of the glass, and the disposition of actinide elements. Observations regarding the effects of the S/V on the solution chemistry and the corrosion of the glass matrix have been reported previously [1,2]. This paper highlights the solution analyses performed to assess how the S/V used in a static leach test affects the disposition of actinide elements between fractions that are suspended or dissolved in the solution, and retained by the altered glass or other materials.

Knowledge of the behavior of radioactive waste components under various conditions will allow realistic source terms to be incorporated into performance assessment calculations. The solubilities of actinide elements are known to be sensitive to the solution pH, Eh, and complexant concentrations, as well as their oxidation states [3]. While the solubilities of many radionuclides are low in neutral and alkaline groundwaters, laboratory tests are required to support predictions of the effects of complexation, sorption, and colloid and secondary phase formation on the behavior of radioelements in a geologic environment. Actinides exist in borosilicate high-level waste glass as U(VI), Np(IV), Pu(IV), and Am(III) [4], where the oxidation states are controlled primarily by redox couples within the melt. The oxidation states of actinides in the leachate will be affected by other redox couples in the surrounding geology, and will be influenced by the atmosphere above the solution [5]. For example, the dominant oxidation states in tuff groundwater under slightly oxidizing conditions have been determined to be Np(V), Pu(V and VI), and Am(III) [6]. Uranium will be present primarily as U(VI) under these conditions.

All actinides readily undergo hydrolysis as the solution pH increases, with the ease of hydrolysis increasing with the atomic number. Most actinides are also complexed by a variety of ligands that are commonly present in groundwaters, including carbonate, fluoride, chloride, sulfate, and phosphate. Other ligands

that complex actinides may be generated by radiolysis of air, such as nitrate and nitrite. In general, complexation decreases in strength as $\text{CO}_3^{2-} > \text{PO}_4^{3-} > \text{F}^- > \text{SO}_4^{2-} > \text{HPO}_4^{2-} > \text{Cl}^-$, NO_3^- , and NO_2^- [7]. Because of its high complexing strength and natural occurrence in most groundwaters, carbonate is usually the dominant complexing ligand in natural groundwaters [5]. Anions released from the waste form as it corrodes will also be available to complex actinides. The anion contents of high-level waste glasses are seldom reported. Sulfur, nitrogen, chlorine, and fluorine are poorly soluble in borosilicate glasses and are usually assumed to be lost as off-gases during vitrification. Phosphorus is poorly soluble in silicate glasses and phosphorus-bearing phases have been found to precipitate in borosilicate waste glasses [8]. Dissolution of such phases may significantly affect the fluid chemistry and the solubility of radionuclides.

Colloidal materials that occur naturally in the groundwater or are generated during corrosion of engineered barrier materials will be available to sorb radionuclides [9,10]. This material may remain suspended in solution, or may eventually be sorbed by immobile phases, may be filtered from the solution by the surrounding geology, or may flocculate and settle out of solution. Sorption is generally stronger for actinides in lower oxidation states, so that Am(III) and Pu(IV) are expected to be more strongly sorbed to colloids and mineral phases than U(VI) or Np(V) [11].

Migration of radionuclides released from the glass will be affected by the many materials present in the geologic formations or engineered barriers surrounding the waste form, as well as by interactions involving the corrosion products of the waste glass itself. Various clays, zeolites, and other mineral phases are known to form as glass waste forms corrode [12,13], and interactions between released radionuclides and these secondary phases may either enhance or reduce their release from the immediate vicinity of the waste form, depending on the mobilities of the particular phases. Reliable long-term predictions of waste behavior must account for these interactions.

EXPERIMENTAL

Tests were performed using frits SRL 131 and SRL 202 supplied by the Savannah River Technology Center. The SRL 202 composition is the current reference composition for blended tank wastes at the Savannah River Site [14], while the SRL 131 composition was developed as a reference glass based on earlier estimates of tank waste compositions. Both glasses were doped at ANL with uranium-238, neptunium-237, plutonium-239, and americium-241 at levels of 2, 0.01, 0.01, and 0.0004 elemental wt %, respectively. The glass and leachant compositions have been presented elsewhere [1,2]. Tests were conducted following the Product Consistency Test (PCT) method B [15] by placing either 1 or 5 g of crushed glass and either 10 or 5 mL of a tuff ground water leachant in a 304L stainless steel reaction vessel and heating at 90°C. The leachant was prepared by prereacting groundwater from well J-13 of the Nevada

Test Site with tuff rock at 90°C and is referred to as EJ-13 water. These glass mass-leachant volume ratios yield S/V ratios of about 2000 and 20,000 m⁻¹.

After a predetermined test duration, the vessels were removed from the oven and opened. The leachate solutions and reacted glasses were removed for analysis. The leachates were removed while still hot to avoid thermal effects, such as precipitation of solids or flocculation of colloids. A portion of some solutions was filtered through a holey carbon grid used for transmission electron microscopy (TEM) to isolate suspended solids for later analysis. An aliquot was also removed for analysis of transuranics. The remaining solution was then filtered through a preheated 0.45 µm cartridge filter. A portion of this solution was further filtered through an Amicon model CF50A filter, which has an effective filter size of about 6 nm. Solutions were analyzed for pH (using a combination electrode), anions (by ion chromatography), uranium (by inductively coupled plasma-mass spectrometry), and transuranics (by alpha spectrometry). The measured pH values are expected to be affected by the temperature (they were measured at room temperature) and the high ionic strengths of the leachate solutions, but to provide a qualitative measure of the effects of the S/V ratio and reaction time on the solution chemistry.

The reacted glass was gently rinsed with deionized water then removed from the vessel and allowed to dry. The vessel was then filled with a nitric acid solution and placed in a 90°C oven overnight to dissolve transuranics which had become fixed to the stainless steel surface. An aliquot of this solution was analyzed using alpha spectroscopy.

RESULTS AND DISCUSSION

Tests conducted at different S/V generate very different solution chemistries which affect the glass corrosion rates [1,2]. The pH and anion concentrations are expected to affect the behavior of released actinides. The pH values and anion concentrations measured in the initial EJ-13 leachant solution and in leachate solutions from tests with SRL 131 and SRL 202 glasses at 2000, and 20,000 m⁻¹ are listed in Table I. High leachate pH values are reached after only a few days in tests with SRL 131 and SRL 202 due to dealkalization as the glasses corrode. Different pH values are attained in tests at different S/V due to the different solution volumes available to dilute corrosion products. The fluoride concentration in most tests was similar to the fluoride concentration in the EJ-13 leachant, about 3 ppm. Quantification of fluoride in some samples was complicated by the incomplete separation of fluoride and formate (CHO₂⁻) in the chromatography column. The fluoride levels in these tests were assumed to be 3 ppm and the formate levels were calculated by difference. Neither fluoride nor formate are expected to affect the solubilities of the actinides at the low levels measured in these tests. Oxalate (C₂O₄²⁻) was present in tests with SRL 202 glass, but not in most tests with SRL 131 glass.

Table I. Anion Concentrations in Tests with SRL 131 and SRL 202 Glass^a

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

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

^bFormate values estimated based on 3 ppm F⁻.

^cNot analyzed.

The chloride content increased slightly in tests at 2000 m⁻¹, but increased to more than 200 ppm in tests with both glasses at 20,000 m⁻¹. Nitrate concentration in all tests remained near the leachant level of 16 ppm. Both chloride and nitrate are weak complexants and these levels are not expected to significantly affect the actinide solubilities. The hydrogen phosphate levels are low in tests at 2000 m⁻¹ and high in tests at 20,000 m⁻¹: tests with SRL 202 glass attain concentrations exceeding 100 ppm, and tests with SRL 131 glass attain concentrations exceeding 400 ppm after two years. High sulfate concentrations were also measured in tests with SRL 131 glass at 20,000 m⁻¹ at all reaction times and with SRL 202 glass in tests conducted for two years. Both plutonium and americium form phosphates which have lower solubilities than the oxides. The release of hydrogen phosphate and sulfate as the glass

corrodes may reduce the solubilities of plutonium and americium in these leachates. Work is in progress to identify actinide-bearing phases. The organic and inorganic carbon contents of the leachates were also measured and the results are included in Table I as the total organic carbon (TOC) and total inorganic carbon (TIC) concentrations. The EJ-13 leachant contains about 3 ppm organic carbon and 22 ppm inorganic carbon. Corrosion of the glass increases both the organic and inorganic carbon levels. In general, tests with SRL 202 glass at 2000 m^{-1} attained slightly higher carbon levels than tests with SRL 131 glass at the same S/V ratio. This is consistent with the higher formate and oxalate concentrations found in tests with SRL 202 glass. However, these levels are not expected to significantly affect the solubilities of the transuranics. Also, tests at $20,000\text{ m}^{-1}$ show a decrease in the inorganic content after 364 days or longer. This may be due to the formation of various carbonate secondary phases (see below), which will decrease the amount of carbonate available to complex transuranics and may reduce their solubilities.

The amounts of neptunium, plutonium, and americium that were suspended in solution (that fraction which passed through a $0.45\text{ }\mu\text{m}$ filter but did not pass through a 6 nm filter), dissolved in solution (that fraction which passed through a 6 nm filter), or became fixed to the stainless steel reaction vessel (the fraction in the acid soak solution) were measured using alpha spectroscopy. The results of these analyses in terms of the amounts suspended in solution, dissolved in solution, and fixed to the steel are presented in Table II for several, representative tests under columns labeled "sus, dis, and ss." Actinides not found in these fractions are assumed to remain associated with the altered glass.

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

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

Table II. Mass of Actinides in Various Fractions and Actinide Retention Factors. Reaction Time in days; fractions that are suspended (sus), dissolved (dis), and fixed to the steel vessel (ss) in mg; normalized mass loss in g/m^2 ; retention factors are dimensionless

Time	Np, ng			Pu, ng			Am, ng			Retention Factors				
	sus	dis	ss	sus	dis	ss	sus	dis	ss	NL(B)	RF(U)	RF(Np)	RF(Pu)	RF(Am)
SRL 202 at 2000 m⁻¹														
14	420	96	51	34	0.75	8.6	0.87	0.017	0.24	0.26	2.4	0.91	12	19
70	110	290	15	60	12	30	1.3	0.23	1.2	0.46	6.6	2.2	9.2	14
280	540	160	0	140	3.1	170	2.1	0.074	3.6	0.70	6.4	2.0	4.5	9.7
560	0	550	0	130	34	110	3.2	0.79	2.6	0.82	4.8	3.0	6.1	9.9
980	87	170	0	74	19	150	1.8	0.39	3.0	1.0	6.7	12	8.3	15
SRL 202 at 20,000 m⁻¹														
14	3000	180	120	180	5.5	17	3.7	0.12	0.59	0.20	4.3	0.12	1.9	3.6
98	75	89	290	1.3	0.28	52	0.026	0.002	1.8	0.38	27	1.7	14	17
182	21	76	360	0	1.7	58	0.02	0.003	2.1	0.53	56	2.3	18	19
364	28	6.8	1100	0.11	0	88	0.01	0	2.6	5.2	1400	9.0	120	160
504	0.43	1.5	860	0	0.01	60	0	0	2.7	12	9200	28	400	360
728	0	69	660	0	0.37	203	0	0.004	6.9	18	8200	54	180	210
SRL 131 at 2000 m⁻¹														
14	221	125	580	5.4	0.65	4.5	0.07	0.004	0.097	1.2	6.7	6.7	120	570
70	64	230	0	13	4.7	46	0.20	0.036	0.93	1.7	300	11	120	120
280	41	35	0	2.4	11	92	0.022	0.009	1.0	5.2	61	59	110	380
560	18	17	0	0.25	0.22	109	0.013	0	1.7	9.7	280	550	180	460
980	46	0	0	0	0	122	0	0	1.9	14	1100	600	220	580
SRL 131 at 20,000 m⁻¹														
14	2.4	1.9	140	0.78	0.002	15	8.6	0	0.48	6.0	910	40	800	1000
98	1.3	0.95	330	0	0.002	140	0	0	3.9	9.2	2000	55	130	190
182	0	7.7	25	0	0.014	33	0	0	1.2	11	2000	710	670	730
364	0	2.7	383	0	0.012	24	0.05	0	0.78	20	5300	100	1700	2100
552	4.6	0	290	0.11	0	81	0.004	0	2.2	23	6800	160	570	840
728	13	0	330	1.4	0	100	0.006	0	2.8	36	5600	210	720	1000

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

Table III shows the highest levels of neptunium, plutonium, and americium measured in the dissolved fractions under each test condition. Also shown are the solubilities of these actinides measured in J-13 groundwater [6]. The concentrations attained in J-13 are several orders of magnitude greater than the amounts measured to be dissolved in these tests. This may be due to competing equilibria between actinides and colloidal phases and steel, or to different solubility controlling phases in tests with glass.

The relevance of these tests to glass performance is with regard to the amounts of each transuranic element that can be transported by groundwater flow. The dissolved fraction is assumed to be fully mobile, while the mobility of the suspended fraction will depend on the environment. Actinides sorbed to the vessel or remaining with the glass may be mobilized as the metal corrodes, as corrosion products slough off the glass, or as the solution chemistry changes. Subsequent interactions may also affect the mobilities of dissolved actinides.

The amounts of actinides that remain associated with the glass during static leach tests can be estimated by comparing the amounts found to be suspended, dissolved, or fixed to the vessel to the extent of glass corrosion in each test. A "retention factor" has been used to describe ability of the corroded glass to mitigate the release of actinides to the solution [16]. The retention factor is defined as the ratio of the normalized boron mass loss (which defines the extent of corrosion) to the normalized actinide mass loss. The sum of the amounts in the suspended, dissolved, and fixed fractions was used to compute the normalized mass losses of the actinides. The normalized boron mass losses are given for the tests in Table II, along with the calculated retention factors for each actinide, $RF(\text{An})$, where An is U, Np, Pu, or Am. Values of $RF(\text{An})$ near

Table III. Highest Concentrations of Actinides Measured in Dissolved Fractions

Test	U (M)	Np (M)	Pu (M)	Am (M)
SRL 202 at 2000 m^{-1}	2×10^{-5}	2×10^{-7}	1×10^{-8}	3×10^{-10}
SRL 202 at $20,000 \text{ m}^{-1}$	3×10^{-6}	2×10^{-7}	5×10^{-9}	1×10^{-10}
SRL 131 at 2000 m^{-1}	2×10^{-5}	1×10^{-7}	5×10^{-9}	1×10^{-11}
SRL 131 at $20,000 \text{ m}^{-1}$	1×10^{-5}	7×10^{-9}	1×10^{-11}	0
Solubility in J-13 (25°C) ^a		2×10^{-3}	4×10^{-5}	1×10^{-6}

^aFrom ref. [6]

one indicate that the actinide is released into solution to a similar extent as boron, while large values of RF(An) indicate that the actinide is retained by the glass as it corrodes. Retention may result from several phenomena, including secondary phase formation, sorption, flocculation of colloidal material, and simply the limited solubilities of the actinides.

In general, these tests show (1) the retention factors for uranium and neptunium are less than those of plutonium and americium, (2) the retention factors increase with the test duration, (3) the retention factors increase with the S/V ratio of the test, and (4) the retention factors are higher in tests with SRL 131 glass than in tests with SRL 202 glass. The first finding can be explained as a result of the higher solubilities of uranium and neptunium, and the second is consistent with the nonstoichiometric corrosion behavior of these glasses [2]. The third finding can be explained as an effect of the solution chemistry and colloid formation. Analysis of the leachate solution before and after filtration through 6 nm filters has indicated that material containing aluminum, iron, and probably silicon becomes suspended in the leachates as the glasses corrode [2]. This material remains suspended in the leachates of tests conducted at 2000 m^{-1} , but settles out of solution after 28 days in tests at $20,000 \text{ m}^{-1}$. Actinides that become associated with suspended material during the initial stages of corrosion are removed from solution as the material flocculates and settles out of solution. Although this material settles out of the solution during static tests, it is likely to be dispersed under even very low flows. The higher retention by SRL 131 glass is due in part to its poorer durability. The NL(B) values for tests with SRL 131 glass are about an order of magnitude higher than those for tests with SRL 202 glass under the same test conditions. Colloids containing actinides form in, and then settle out of, solution faster in tests with SRL 131 than in tests with SRL 202 glass.

Colloids that have been identified to be suspended in the leachates of these tests include smectite and kaolinite clays, nagelschmidite, calcite, dolomite, iron oxide, magnesium oxide, titanium oxide, wecksite, uranophane, boltwoodite, and other uranium-titanium-, iron-, and aluminum-rich phases [17]. Smectite clays are the most ubiquitous and abundant phases found suspended in the solution. Smectite clays are known to be an abundant alteration phase of the hydrated surfaces of these glasses [1]. All of the other phases have also been found to be either associated with alteration layers formed on individual glass grains or existing as separate phases mixed with the reacted glass. These phases may nucleate either in the solution or on the reacted glass surface, and may either enter or settle out of the solution, depending on the solution chemistry and the size of the particles.

The suspended phases provide a means of attaining mobile fractions of actinide elements at levels far above their solubilities either through their being sorbed onto or incorporated into the phases. While several uranium-bearing phases have been identified, the low concentrations of transuranic elements used in testing make it difficult to identify the secondary phases which sequester them

and/or affect their solubilities. The present results are consistent with previous work that has shown plutonium and americium oxides to be associated with smectite clay colloids [18]. Work is in progress to identify phases formed during glass corrosion in the present tests.

CONCLUSIONS

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

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