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J. H. Westsik, Jr.
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March 1983

Prepared for the
Office of Nuclear Waste Isolation
under U.S. Department of Energy
Contract DE-AC06-76RLO 1830

Pacific Northwest Laboratory
Operated for the U.S. Department of Energy
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SUMMARY

Researchers at the Pacific Northwest Laboratory are studying the chemical durability of a simulated high-level waste glass when exposed to high-temperature geologic solutions. In the study reported here, simulated high-level waste glass-beads (76-68 glass) doped with technetium, uranium, neptunium, plutonium, curium and americium were leached in deionized water, Waste Isolation Pilot Plant salt brine "B", and 0.03M sodium bicarbonate solution at 150 and 250°C for 2, 4, 8, 16, and 32 days. The resulting solutions were analyzed for several nonradioactive glass components and for the radioactive dopants.

The major objective of this report is to present data not previously reported in open literature. We have also included data that was discussed and interpreted in a previous report (Westsik and Peters 1982).

The glass exhibited "incongruent" leaching behavior, i.e., the normalized releases (g-glass/m^2) based on the different elements spanned four orders of magnitude. Normalized releases based on boron, molybdenum, sodium, cesium, silicon, and technetium were the same within a factor of three. They were also the highest normalized mass losses. Normalized mass losses based on the actinides were at least one order of magnitude lower.

Most of the nonradioactive components of the glass were released more to the salt brine than to the other two solutions. However, silicon, boron, molybdenum, technetium, and the actinides had their lowest releases in the salt brine. Reaction-layer thickness on the glass surface and weight losses of the glass beads were also smallest in the brine solution. Actinide releases were highest in the sodium bicarbonate solution. Calcium, strontium and barium releases decreased with time and temperature; the releases of most other elements increased with time and temperature.

Solubility appears to be limiting the release of most elements. The leachate pH is controlled by chemical species within the original leachant and by species released as the glass leached. In addition, carbonate ion

complexes with some elements including uranium--effectively increasing their release. The more soluble elements including sodium, boron, molybdenum and technetium provide an indication of the actual rate of reaction between the glass and water.

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INTRODUCTION

The chemical durability of a simulated high-level waste (HLW) glass when exposed to high-temperature simulated geologic solutions is being studied at the Pacific Northwest Laboratory, operated by Battelle Memorial Institute for the U.S. Department of Energy. This report presents results of work conducted as part of the Waste Package Program (WPP) to evaluate the behavior of waste glass in simulated repository environments.

Specifically, this document reports the results of tests in which a simulated HLW glass (PNL 76-68 glass) doped with various radiotracers was exposed to: 1) a salt brine simulating that found near the Waste Isolation Pilot Plant (WIPP) site near Carlsbad, New Mexico; 2) a 0.03M sodium bicarbonate solution representing potential silicate rock repositories; and 3) deionized water. Tests were run at 150 and 250°C to provide kinetic information so that short-term test results may be extrapolated to longer times at lower temperatures. To achieve this goal, the tests discussed here were run under conditions similar to other WPP-sponsored tests being run at 25 and 75°C for longer times (Peters and Diamond 1981). Time and temperature dependences of leaching as determined by these tests have been reported by Westsik and Peters (1981). They emphasized the leaching behavior of boron, molybdenum, silicon and sodium.

The objective of the present document is to report the leach test results at 150 and 250°C for other components of the glass including actinides and technetium, that were not previously reported by Westsik and Peters. The experiments discussed in this report were conducted during 1979. Additional experiments on the high-temperature leaching of 76-68 glass, funded by the High-Level Waste Immobilization Program, were conducted in 1980; the data is listed in Appendix B. Both sets of data are being documented in this report in order to help complete the data base on the leaching behavior of 76-68 glass.

Because leaching is a complex reaction between the glass, the leachant, and the surrounding environment, our experimental philosophy has been to start with simple systems and then progress to more complicated systems as we gain an

understanding of the simpler ones (Bradley, McVay and Coles 1980). Therefore, work reported here addresses only the glass-solution interactions. More recent studies include the canister, backfill, and geologic media (Shade and Bradley 1980).

For ease of handling and chemical analysis, these tests used glass with nonradioactive isotopes substituted for the fission products. The same glass was also doped with technetium and these actinides: uranium, neptunium, plutonium, americium, and curium. The glass-solution interactions were studied with respect to 1) the amounts of materials released from the glass to the solutions, and 2) the changes in the glass as a result of the exposure.

EXPERIMENTAL

Undoped glass beads and glass beads doped with either ^{99}Tc, ^{244}Cm, ^{233}U and ^{243}Am, or ^{237}Np and ^{239}Pu were exposed to salt brine, 0.03M sodium bicarbonate solution and deionized water at 150 and 250°C for times ranging from 2 to 32 days. The nonradioactive simulated-HLW glass beads were used to facilitate chemical analysis of the leachates and analysis of the glass surfaces after leaching. Technetium and the actinides were included in other glass beads so that their specific leaching behavior could be studied. The test matrix is shown in Table 1. The composition including radiotracers of the glass, commonly known as 76-68, is shown in Table 2 (McElroy 1977). Preparation and characterization of the glass beads were described earlier by Bradley, Harvey, and Turcotte (1979), who found that the technetium segregated into a phase that coated small pores inside the glass beads, while the other dopants were evenly distributed throughout the glass matrix. Curium-doped beads had a geometric surface area of ~1.6 cm^2; all other beads had geometric surface areas of ~1.4 cm^2. The leachant compositions are shown in Table 3.

A single glass bead weighing about a third of a gram was placed in a gold capsule. Fifteen milliliters of solution were added to the gold capsules in the undoped-bead tests, and due to an error in pipetting, 22 mL were added to the doped-bead tests. The resulting specimen surface area to solution volume ratios (SA/V) therefore ranged from 0.06 to 0.09 cm^{-1} as indicated in Table 1. Each capsule was then purged with argon and sealed by tungsten-arc inert-gas welding. The capsules were placed in Hastelloy autoclaves that were then pressurized with argon to a pressure of 13.8 MPa (2000 psi) at the operating temperatures. After the tests, the capsules were cooled back to room temperature, the capsules were opened, and the solutions and beads were removed from the capsules. The solutions were not filtered but were acidified with 0.01 mL of concentrated nitric acid. In the doped-bead tests, the gold capsules were then rinsed with 30 ml of a 5M HNO_3+0.05M HF solution to remove any material that may have precipitated ("plated out") on the capsule walls. The nonradioactive solutions were analyzed using inductive-coupled

TABLE 1. Experimental Matrix Showing Number and Conditions of Tests

Dopant	Time, days	Solution: WIPP "B" Brine, Temperature, °C: 150	WIPP "B" Brine: 250	0.03M Sodium Bicarbonate: 150	0.03M Sodium Bicarbonate: 250	Deionized Water: 150	Deionized Water: 250
Undoped	2	1(a)	1	1	1	1	1
	4	1	1	1	1	1	1
	8	1	1	1	1	1	1
	16	1	1	1	1	1	1
	32	1	1	1	1	1	1
Cm^{244}	2	1(b)	1	1	1	1	1
	4	0	0	0	0	0	0
	8	1	0	1	0	1	0
	16	1	0	1	0	1	0
	32	1	1	1	1	1	1
Tc^{99}	2	1(c)	1	1	1	1	1
	4	2	1	2	1	2	1
	8	1	2	1	2	1	2
	16	1	1(d)	1	1(d)	1	1(d)
	32	1	2	1	2	1	2
^{233}U and ^{243}Am	2	1(c)	1	1	1	1	1
	4	1	1	1	1	1	1
	8	1	1	1	1	1	1
	16	1(d)	1(d)	1(d)	1(d)	1(d)	1(d)
	32	1	0	1	0	1	0
^{237}Np and ^{239}Pu	2	1(c)	1	1	1	1	1
	4	1	1	1	1	1	1
	8	1	1	1	1	1	1
	16	1	1(d)	1(d)	1(d)	1(d)	1(d)
	32	1	1	1	1	1	1

(a) SA/V = 0.09 for undoped beads.
(b) SA/V = 0.07 for curium-doped beads.
(c) SA/V = 0.06 except as noted for beads doped with Tc, U, Am, Np and Pu.
(d) SA/V = 0.09.

emission spectrometry and flame-emission spectrometry. Also, the leachate and rinse solutions obtained from the tests with the curium-doped beads and some technectium-doped beads were analyzed by ICP. Actinide and technetium concentrations were measured by radiochemical techniques described by Bradley, Harvey and Turcotte (1979).

TABLE 2. Nominal Composition of 76-68 Glass

Nonradioactive Constituents

Oxides	Weight Percent	Oxides	Weight Percent
SiO_2	39.8	Pr_6O_{11}	0.5
Na_2O	12.5	La_2O_3	0.5
Fe_2O_3	9.8	P_2O_5	0.5
B_2O_3	9.5	Cr_2O_3	0.4
ZnO	5.0	SrO	0.4
U_3O_8	4.6	Sm_2O_3	0.3
TiO_2	3.0	TeO_2	0.3
MoO_3	2.3	Y_2O_3	0.2
CaO	2.0	NiO	0.2
ZrO_2	1.8	Rh_2O_3	0.2
Nd_2O_3	1.7	Rb_2O	0.1
CeO_2	1.2	Eu_2O_3	<0.1
RuO_2	1.1	Gd_2O_3	<0.1
Cs_2O	1.0	Ag_2O	<0.1
BaO	0.6	CdO	<0.1
PdO	0.5		

Radioactive Tracers

Tracer	Weight Percent
^{99}Tc	0.46
^{233}U	2.5
^{237}Np	0.41
^{239}Pu	0.041
^{243}Am	0.086
^{244}Cm	0.012

TABLE 3. Leaching Solution Compositions

Leachant	Nominal Composition, mg/L	Analysis, mg/L[b]
WIPP "B" Salt, Brine[a]		
Na	115,000	NA[c]
K	15	34
Mg	10	10
Ca	900	723
Fe	2	1.3
Sr	15	28
Rb	1	NA
Cs	1	2.2
Cl^-	175,000	NA
$SO_4^=$	3,500	NA
$BO_3^=$	10	11.4
HCO_3^-	10	NA
Br^-	400	NA
I^-	10	NA
Ce		1.2
La		2.4
Nd		0.7
Si		3.2
U		1.5
Plus Traces of Other Elements		
Sodium Bicarbonate, 2.52 g/L		
Na	690	749
HCO_3^-	1,380	NA
Plus Traces of Other Elements		
Deionized Water, Trace Impurities		

(a) Dosch and Lynch (1978).
(b) Composition as measured by ICP and flame emission; does not include all elements.
(c) NA = not analyzed.

RESULTS AND DISCUSSION

The physical characteristics of the glass beads and leachate analyses after high-temperature leaching are discussed in this section.

SOLUTION ANALYSES

Elemental release data were obtained from 1) ICP analyses of the undoped-bead leachates, 2) ICP analyses of the curium-doped bead and some technetium-doped bead leachates, 3) radiochemical analyses of all doped-bead leachates, and 4) radiochemical analyses of the acid-rinse solution from the doped-bead leach containers. The results of this study are summarized below. Interested readers should refer to Appendix A for the complete data obtained (both solution concentrations and normalized releases are listed).

Calculations leading to the normalized releases are based on the equation

$$\text{normalized release } (\text{g-glass/m}^2) = \frac{a_n}{a_o} \cdot \frac{w}{s}$$

where

a_n = the amount of element or radionuclide released (mg or d/m)

a_o = the initial amount of element or radionuclide in the glass (mg or d/m)

w = the weight of the sample, g

s = the geometric surface area of the bead, m^2.

For the actinides and technetium, a_n includes both the quantity found in the leachate and the amount detected in the acid rinse of the gold capsules. The acid-rinse solutions contained at least 20% of the actinide leached from the glass beads, and in many of the tests, the rinse contained the majority of the actinide released. It is not known, however, how much of the actinide sorbed on the gold while the samples were at the test temperature and how much precipitated as the samples were cooled back to room temperature.

Nor is it known how much of the actinide sorbed on the glass bead itself. The discussions that follow are based upon the total amount released (i.e., leachate plus acid rinse), although normalized releases based only on the actinide concentration in the leachate (i.e., ignoring plateout) are also listed in Appendix A. For the technetium-doped beads, most (at least 93%) of the technetium released was found in the leachate.

Figures 1 through 6 show normalized releases based on the behavior of the actinides and Tc. The figures show that the actinides are released more to the bicarbonate solution than to either the brine or deionized water leachants. In all leachants, the normalized releases of the actinides were always lower than the normalized releases of Si (Figures 7 through 12). Bradley, Harvey and Turcotte (1979) reported similar actinide behaviors in leach tests conducted at 22°C using a modified International Atomic Energy Agency (IAEA) long-term leach test procedure. It appears that the actinides leach at a lower rate than the glass matrix so that the actinides remain in a leached layer on the glass surface. In an early study, samples of an undoped 76-68 glass leached for 2 days in 99°C deionized water exhibited a rare-earth-rich layer near the glass surface (Houser, Tsong and White 1978). We assume that the actinides remain in a similar layer in our tests.

Technetium-based releases are always higher than the normalized, Si-based releases. Releases of Tc to the WIPP "B" brine were an order of magnitude lower than those to the other two leachants.

Figures 7 through 12 show normalized releases based upon some of the nonradioactive species in the glass. These curves are included only to show the general trends in behavior of the glass components. Actual data should be taken from the tables in Appendix A. Impurities in the acid solution used to rinse the gold capsules were in high-enough concentrations to mask any nonradioactive, "plate-out" species that may have redissolved in the rinse solution. Therefore, the curves shown in Figures 7 through 12 represent only results of the analyses of the leachate and do not include any plateout. By comparing the results of the analyses of the rinse blank, the rinse solutions themselves, and the leachates; it is possible to state qualitatively that the

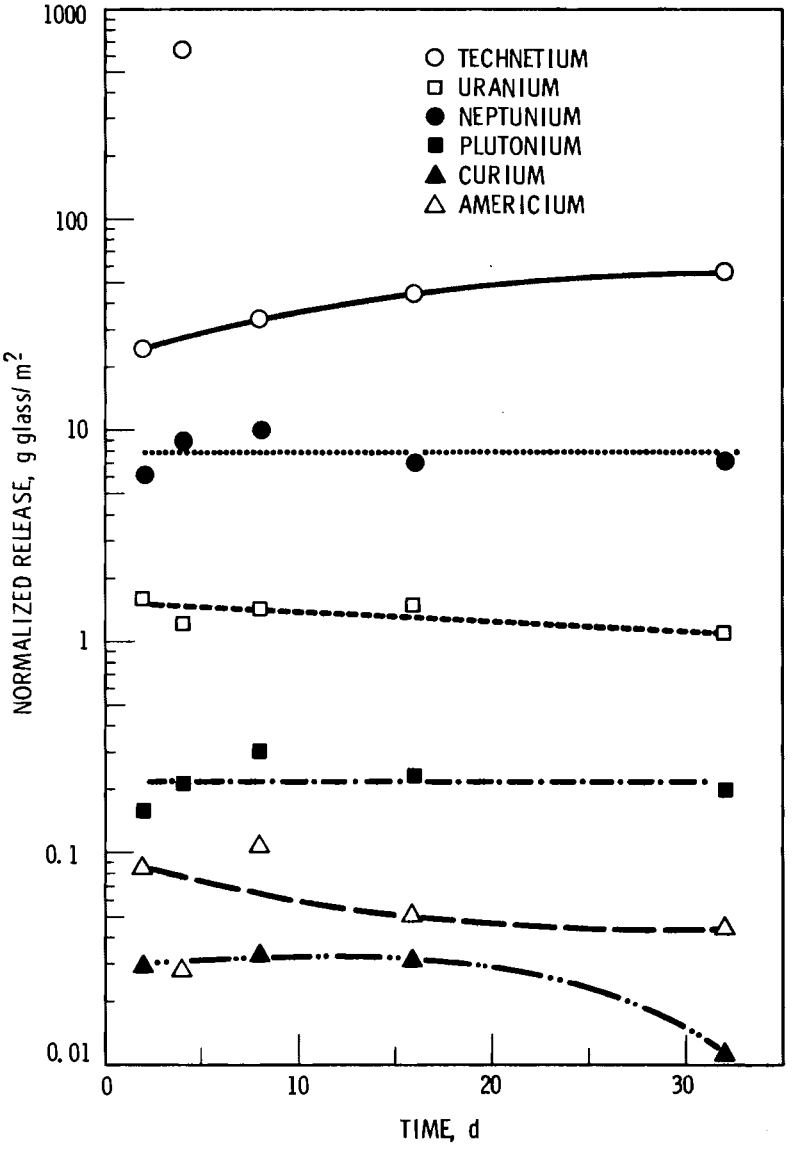

FIGURE 1. Actinide and Technetium Releases into 150°C WIPP "B" Brine

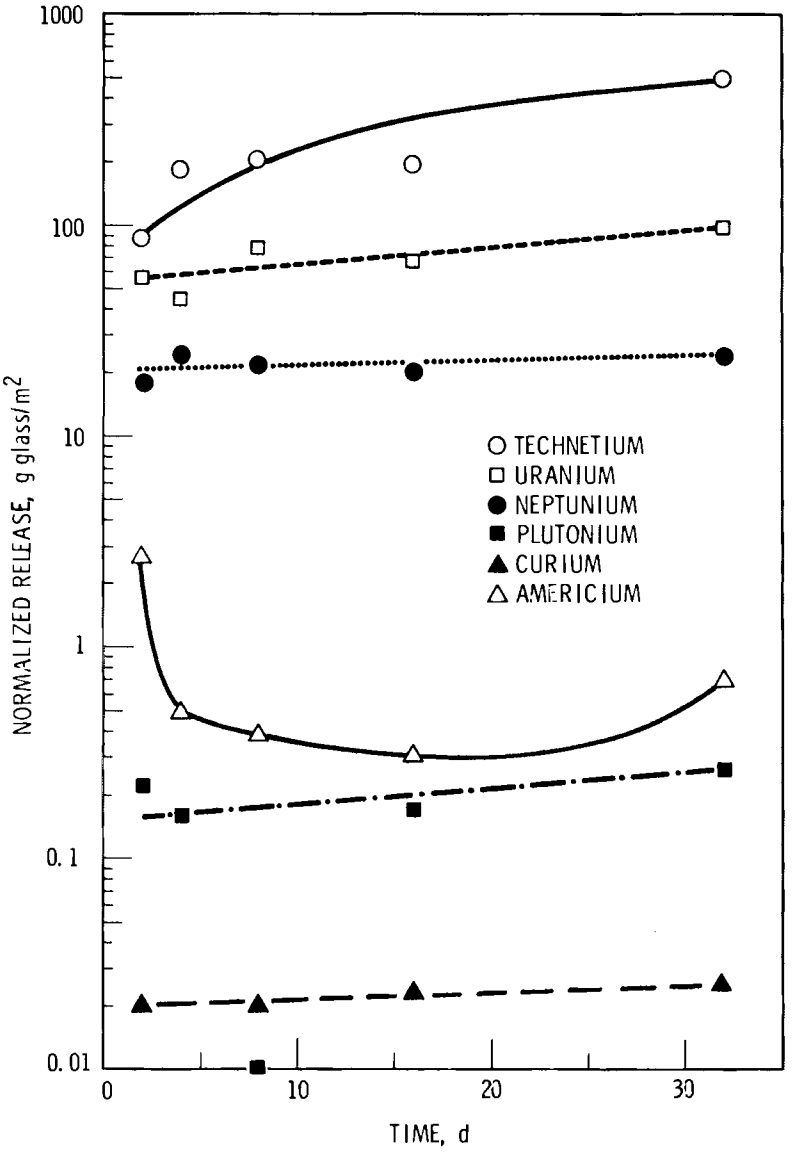

FIGURE 2. Actinide and Technetium Releases into 150°C Sodium Bicarbonate Solution

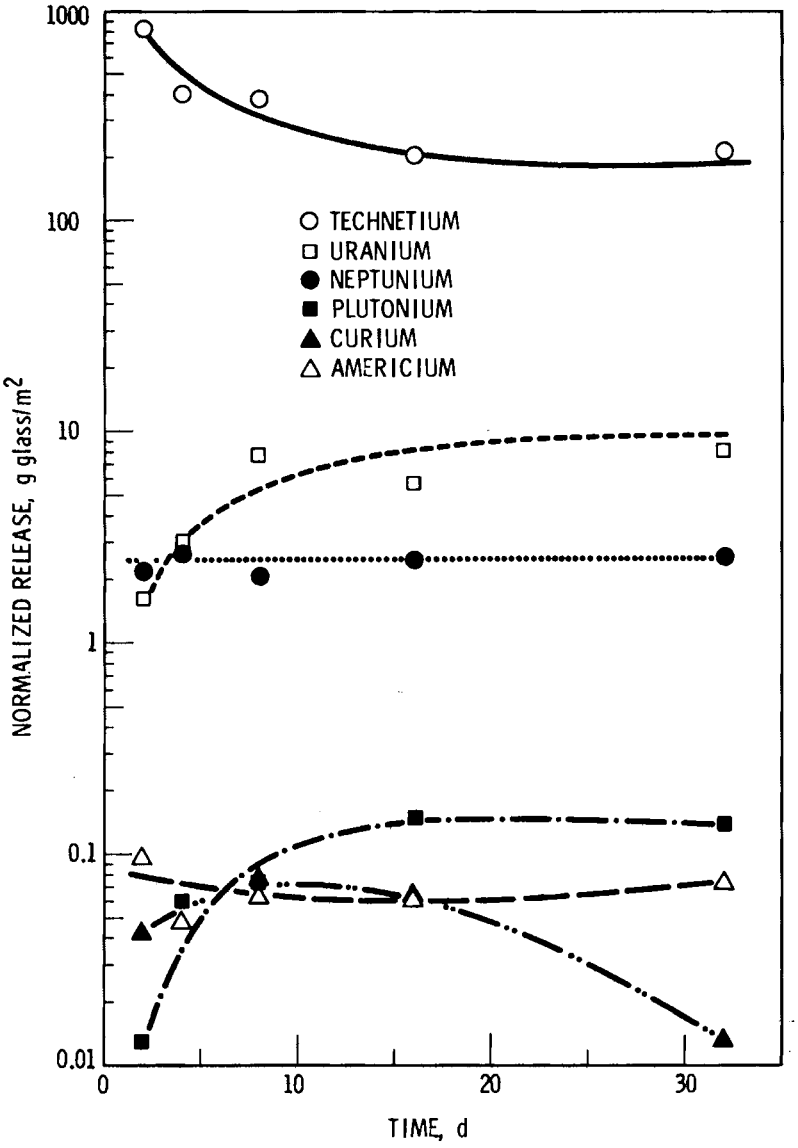

FIGURE 3. Actinide and Technetium Releases into 150°C Deionized Water

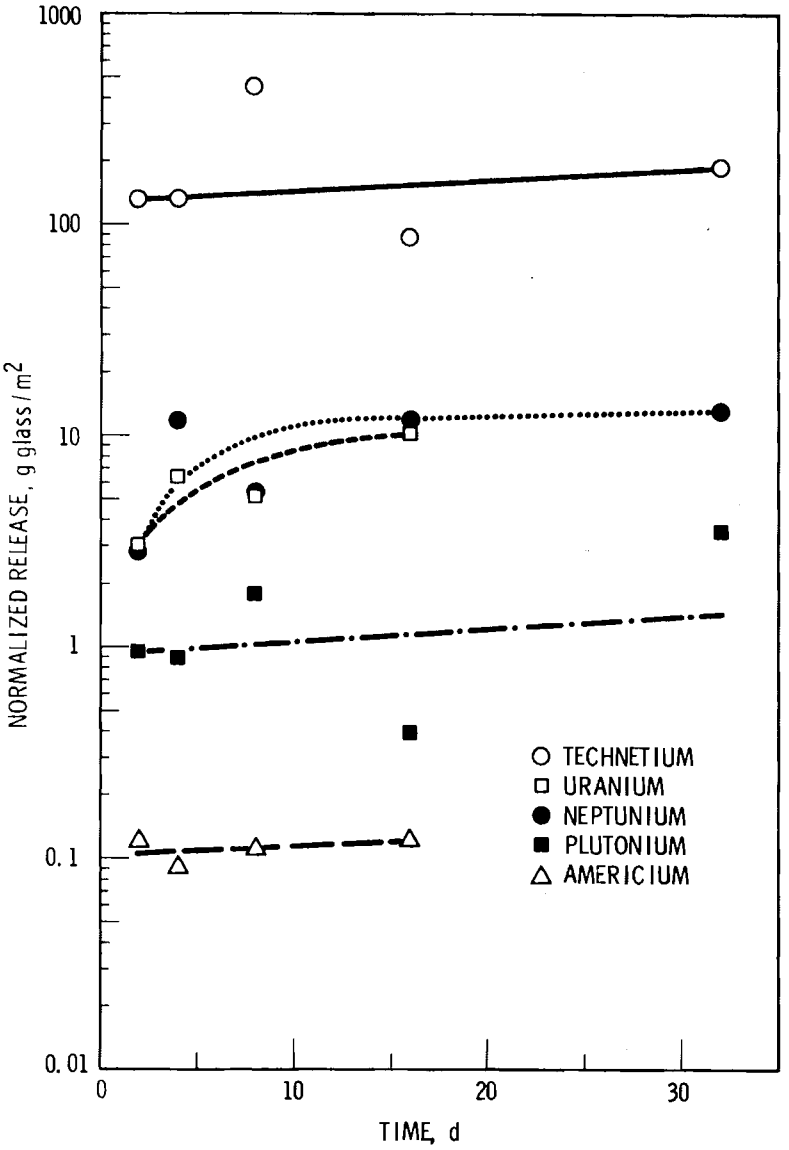

FIGURE 4. Actinide and Technetium Releases into 250°C WIPP "B" Brine

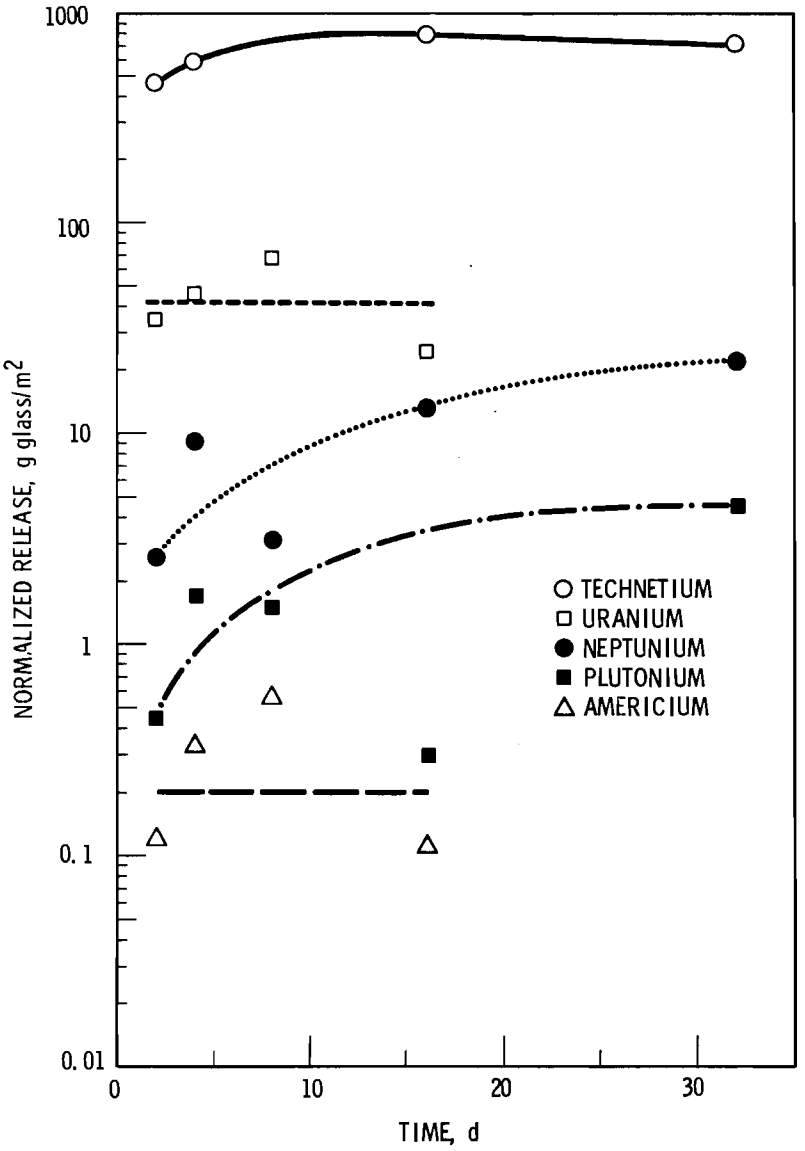

FIGURE 5. Actinide and Technetium Releases into 250°C 0.03M Sodium Bicarbonate Solution

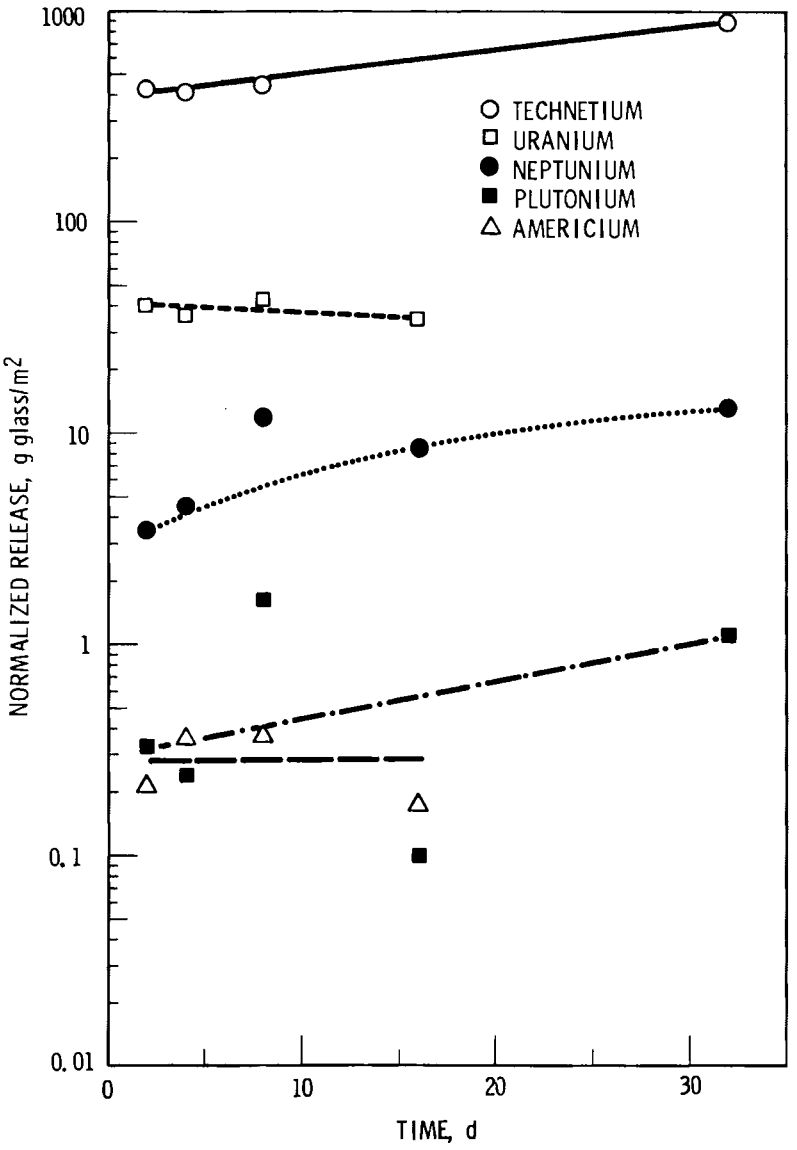

FIGURE 6. Actinide and Technetium Releases into 250°C Deionized Water

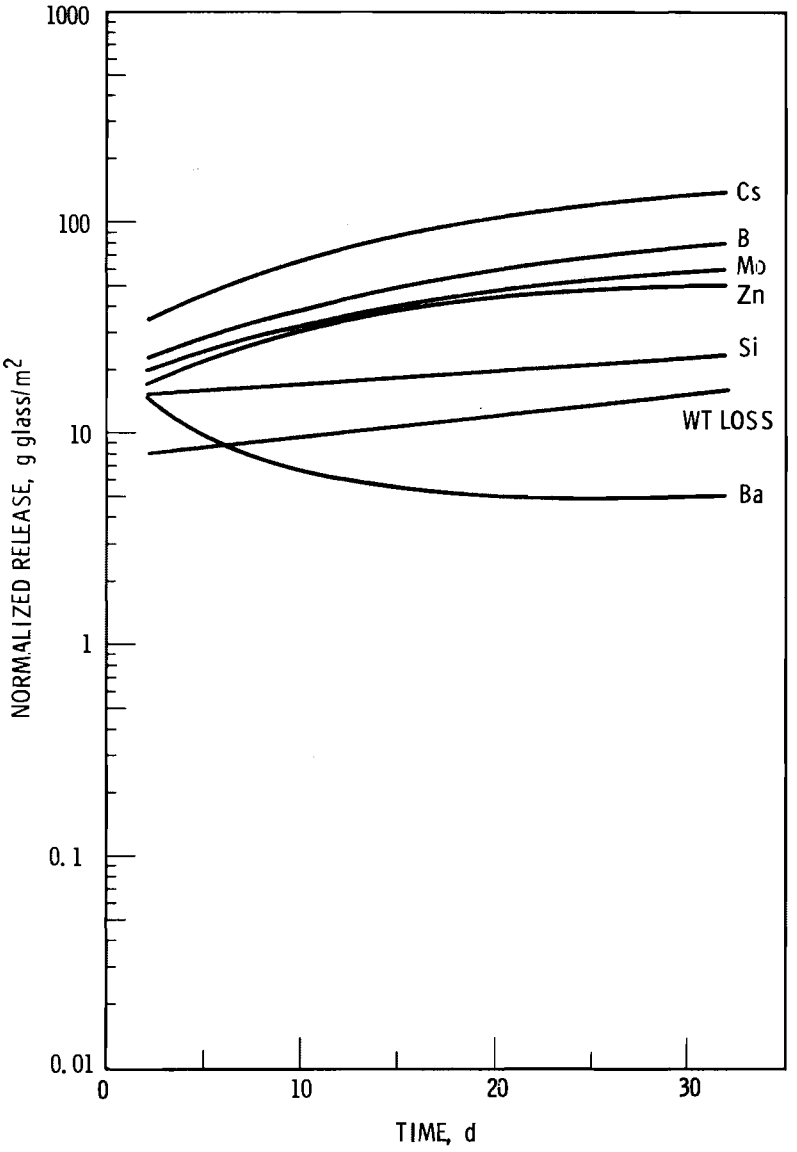

FIGURE 7. Releases of Nonradioactive Species into 150°C WIPP "B" Brine. (Refer to Appendix A for actual data points.)

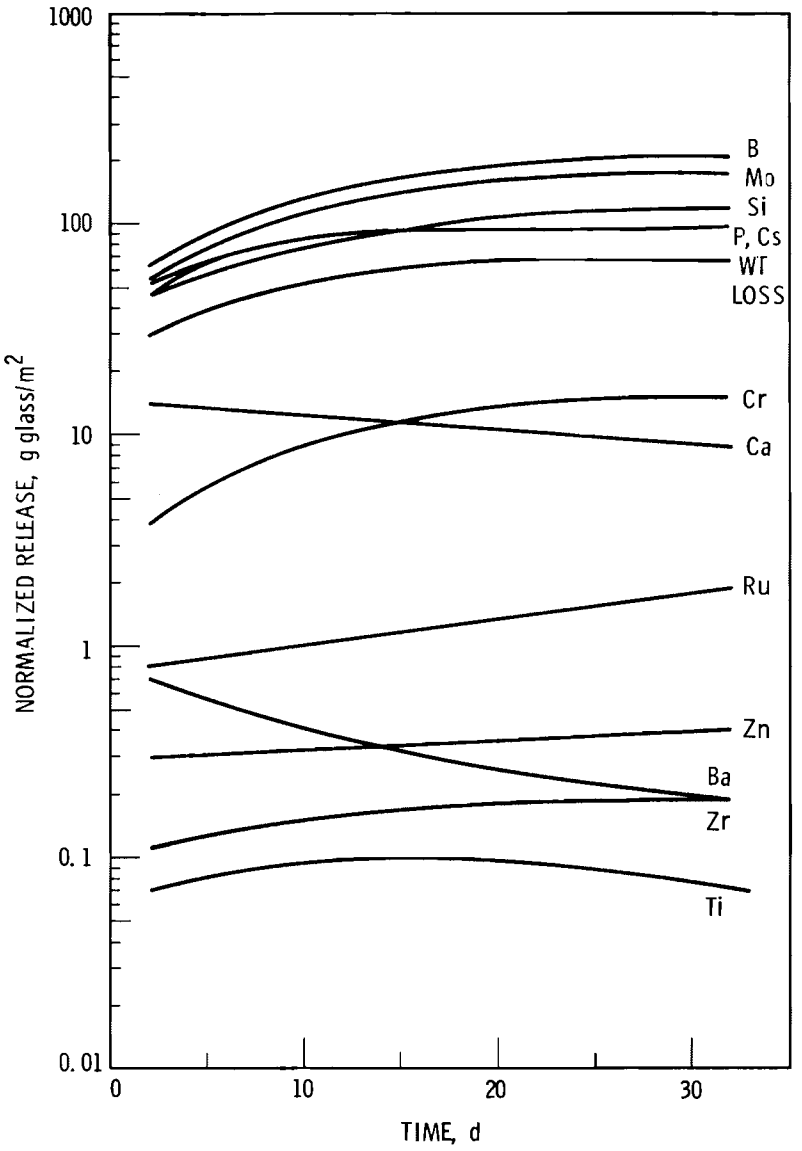

FIGURE 8. Releases of Nonradioactive Species into 150°C Sodium Bicarbonate Solution. (Refer to Appendix A for actual data points.)

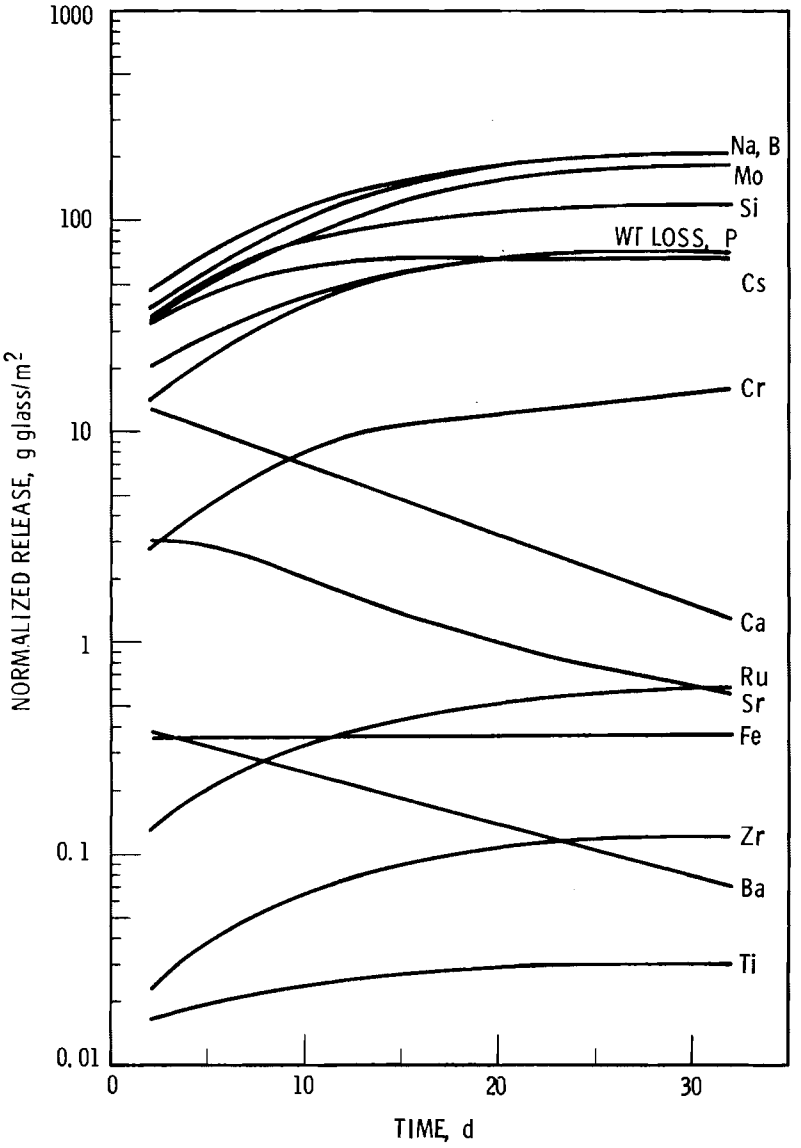

FIGURE 9. Releases of Nonradioactive Species into 150°C Deionized Water. (Refer to Appendix A for actual data points.)

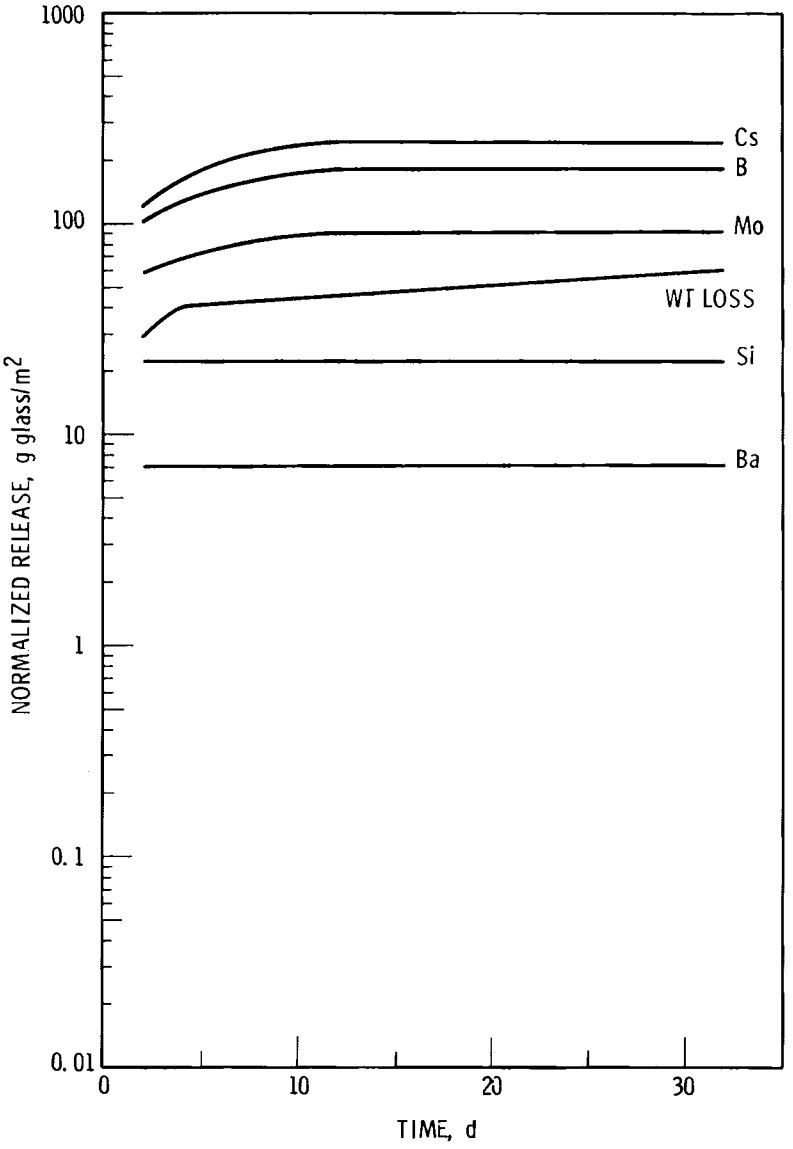

FIGURE 10. Releases of Nonradioactive Species into 250°C WIPP "B" Brine. (Refer to Appendix A for actual data points.)

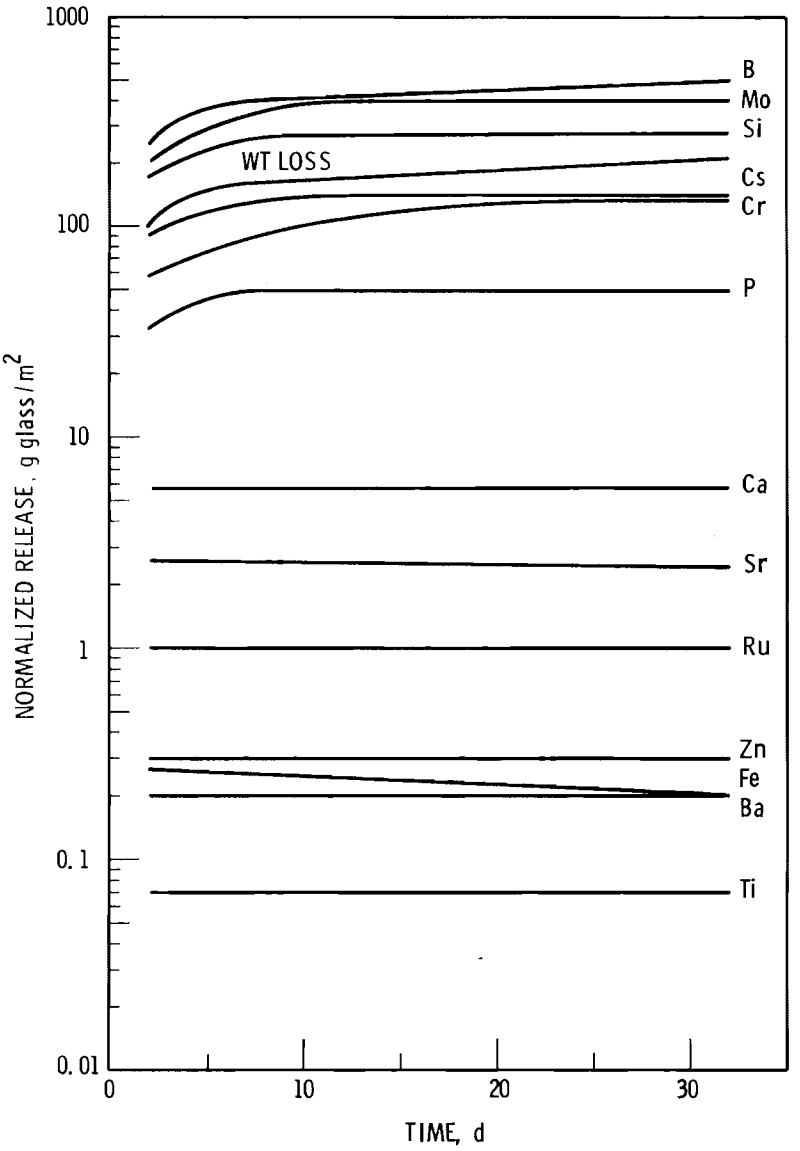

FIGURE 11. Releases of Nonradioactive Species into 250°C Sodium Bicarbonate Solution. (Refer to Appendix A for actual data points.)

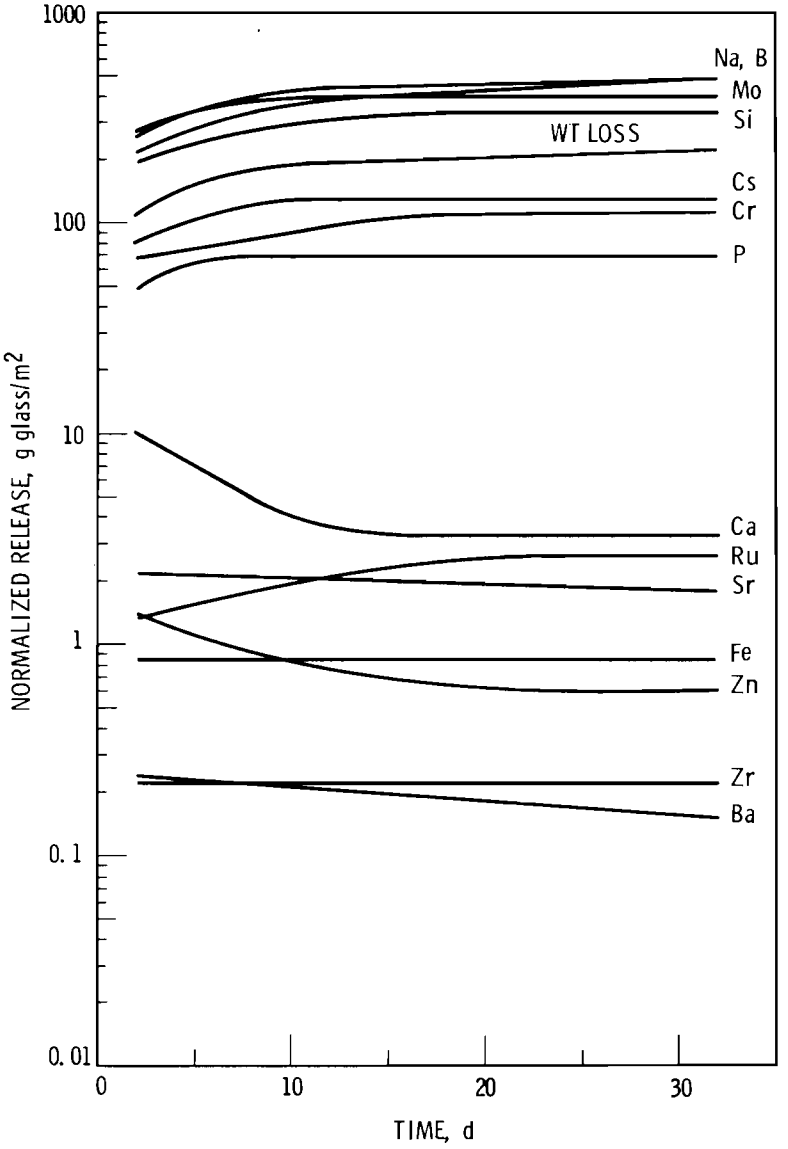

FIGURE 12. Releases of Nonradioactive Species into 250°C Deionized Water. (Refer to Appendix A for actual data points.)

rinse of the gold capsules did not remove significant quantities of most of the nonradioactive species. However, there may be sufficient quantities of Fe, Ti, Zn, Ba, Ca, and Sr in the rinse solutions to increase the calculated release of these elements above the releases calculated from the leachate analyses alone.

The trends of normalized releases of the nonradioactive species to deionized water and to the sodium bicarbonate solution are similar. Releases increase with increasing temperature and increasing time of leaching. However, the alkaline earth (Ca, Sr, and Ba) concentrations decrease as the length of the test increases. As the glass undergoes hydrothermal alterations, these elements are probably incorporated into newly formed crystalline phases. Nickel, iron and zinc behaved erratically and their releases are not shown. They also may be involved in the formation of new phases. Iron in particular has been shown to recrystallize, forming an acmite phase, $NaFeSi_2O_6$, under hydrothermal conditions (Westsik and Turcotte 1978). Most of the nonradioactive species were released more to the WIPP "B" salt brine than to either of the other two solutions. Silicon, boron and molybdenum were the only nonradioactive elements with lower concentrations in the salt brine than in the deionized water or sodium bicarbonate solutions.

It is apparent from Figures 6 through 11 that for many cases the releases of radionuclides do not increase with time after a few days, and from Figures 7 through 12 that the same is true for many nonradioactive elements. In some cases, there is an obvious decrease in the release of an element. Hence, it appears that for many elements the releases are controlled by solubilities, as has been shown recently by Grambow (1982) for other experiments. From one leachant to another, the solubilities could be affected both by pH and the concentration of complexing ligands, such as CO_3^{2-}.

After 2 days at 250°C, the pH (measured at room temperature) of the solutions increased from the initial values as follows: deionized water, 7.0 to 9.7; bicarbonate, 8.6 to 9.3; WIPP "B" brine, 7.0 to 7.3. The rise of pH in deionized water can be attributed to the release of alkali from the glass and the buffering effect of silicic and boric acid also released from

the glass. The buffering effect of bicarbonate is responsible for the alkaline and relatively constant pH for that leachant. We would speculate that in the case of the WIPP "B" brine, the pH rise may be suppressed by the precipitation of magnesium or calcium compounds.

The pH values reported in the paragraph above are only pH values collected in these tests. Additional pH data are available from the work reported by Westsik and Peters (1981). In their work, glass cylinders of 76-68 glass were leached for 3 days in deionized water at 25, 50, 100, 150, 200, 250, 300, and 350°C. Additional experiments were conducted for 1, 12, 27,48, and 64 days in deionized water at 100 and 250°C. In all these tests the SA/V was maintained at 0.099 cm^{-1}. Experimental details and leachate analyses for these tests are presented in Appendix B. For all times and temperatures except 25 and 50°C for 3 days the pHs were in the range 9.4 to 10.0. The pHs at 25 and 50°C were measured to be 8.9 and 7.7, respectively. It is expected, therefore, that in the doped-bead tests in deionized water reported here, the final pH fell in this same range of 9.4 to 10.0.

The scatter in the actinide data is great enough to obscure subtle effects of pH; however, the increased release of uranium at 150°C in deionized water compared to WIPP "B" brine may be due to a higher solubility at the higher pH attained in the deionized water, due to the formation of anionic uranium species. The effect of carbonate complexation is evident in the still greater release of uranium in the bicarbonate solution at 150°C. The same trend appears at 250°C but is weaker. In the case of neptunium at 150°C, the lower pH of the brine compared to that of deionized water results in a greater release, perhaps corresponding to a greater solubility (e.g., involving only cationic or neutral species). Similar arguments regarding pH and CO_3^{2-} concentration can be offered regarding plutonium and the other actinides, but the scatter in the data is rather large.

The more soluble elements, such as sodium, boron, and molybdenum (presumably in anionic form), would appear in solution in proportion to the amount of glass reacted with water. With the exception of the data for

deionized water at 150°C, the normalized technetium release closely follows that of sodium or boron, and hence can be regarded as another soluble element. The scatter in the technetium release data may be attributable to the inhomogeneous distribution of technetium in the glass.

GLASS BEADS

Time, temperature, and solution composition all affected the physical appearance of the glass beads. Very little change was observed among the beads tested at 150°C. Those beads exposed to the salt brine showed some signs of pitting, and the beads leached in the sodium bicarbonate and deionized-water systems showed, at most, a slight loss of surface shine.

Among the beads tested at 250°C, it was much easier to differentiate the beads by their appearance. As at 150°C, the beads exposed to 250°C salt brine appeared to have pitted surfaces. The surface shine of the beads leached in the bicarbonate solution decreased as the length of the test increased. The color change of the beads leached in deionized water was very dramatic; after exposure for 2 days the beads had a metallic, gold sheen. With increasing test length, the sheen turned to a dull orange finish. Examination of the surface of the orange beads using Auger spectrometry revealed that these beads had a high iron concentration on the surface.

The undoped beads leached 32 days at 250°C were later sectioned and mounted for microscopic examination. Figure 13 shows an unleached bead and the beads leached in the three solutions. The reaction layer thicknesses were estimated to be 0.17 mm for the bicarbonate-leached bead, 0.21 mm for the deionized-water-leached bead, and 0.09 mm for the brine-leached bead.

Part of the reaction layer on the bead leached in the sodium bicarbonate solution was removed and examined with the scanning electron microscope with an energy dispersive x-ray (SEM-EDX) attachment. Figure 14 shows some of the crystalline phases found in this reaction layer and their associated x-ray fluorescence traces. The phase rich in silicon and iron may be an acmite and

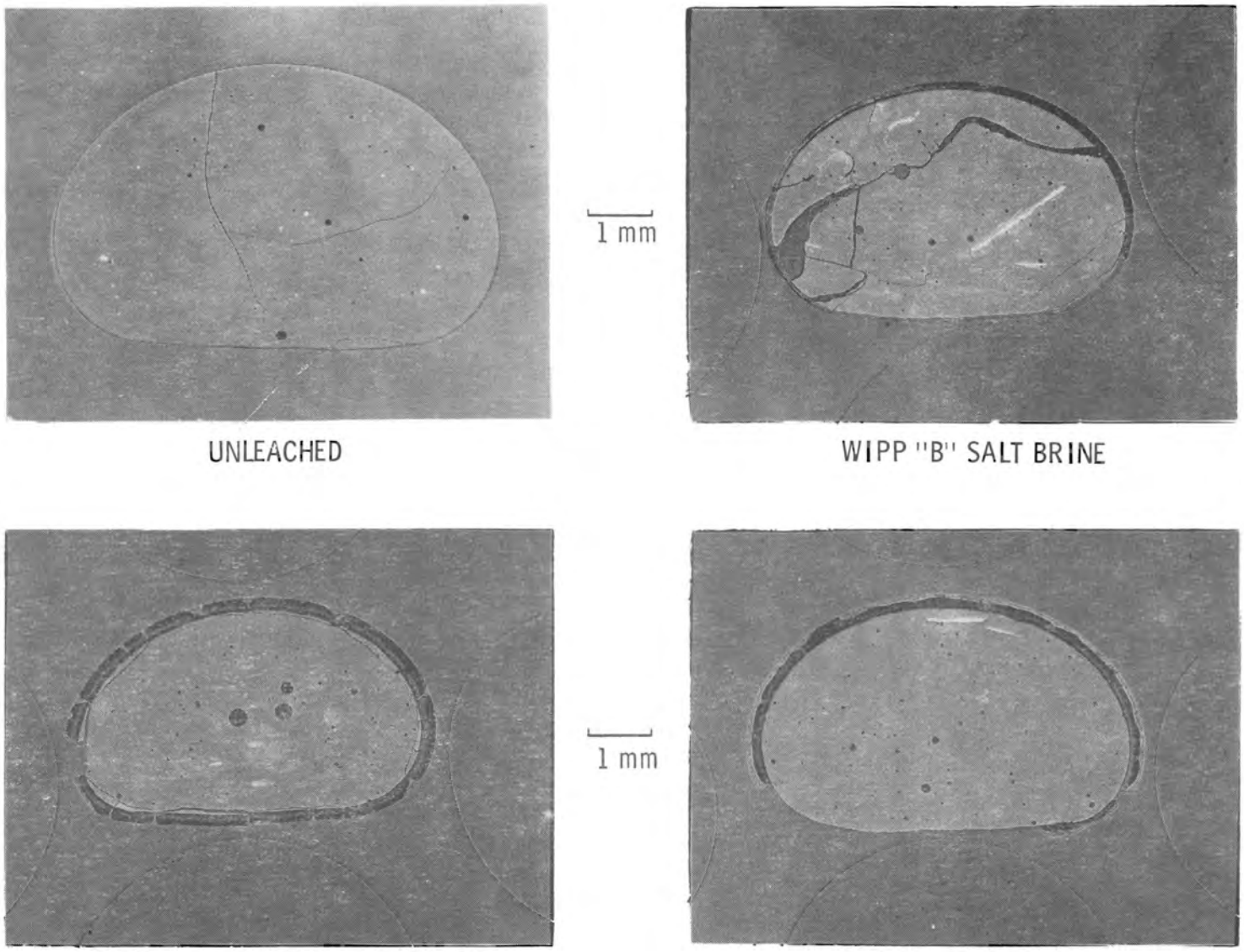

FIGURE 13. Cross Sections of Glass Beads Leached 32 Days at 250°C in the Indicated Solutions

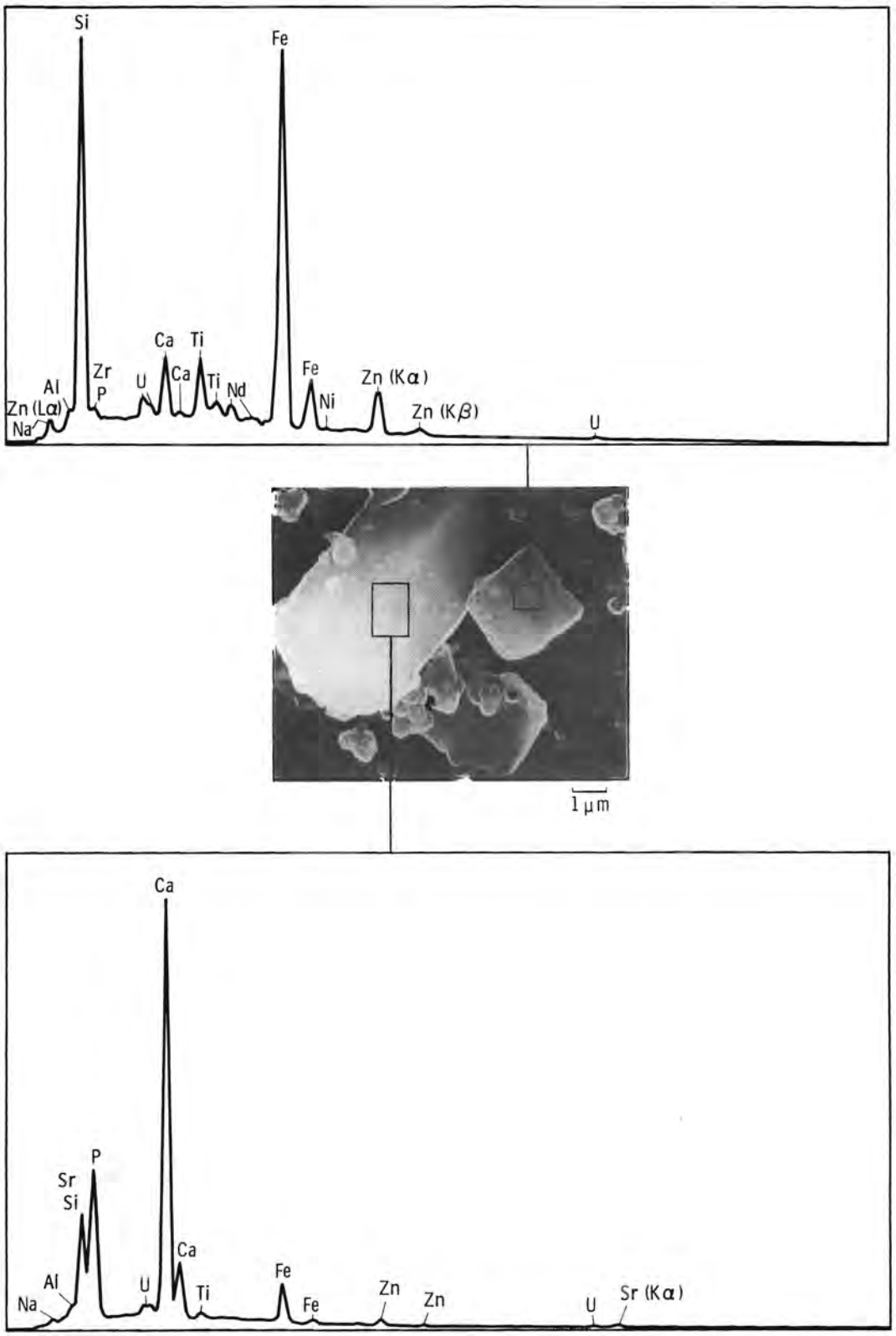

FIGURE 14. Photomicrograph of Crystalline Phases Observed in Reaction Layer of Glass Bead Leached 32 Days in 250°C Deionized Water

the calcium-rich phase may be an apatite similar to the acmite and apatite phases identified by McCarthy et al. (1978) in their studies of the hydrothermal reactions of 76-68 glass in deionized water and in an artificial basaltic ground water.

CONCLUSIONS

A simulated high-level waste glass which contained tracer quantities of Tc, U, Np, Pu, Am, and Cm was leached in deionized water, a sodium bicarbonate solution and the WIPP "B" salt brine, all at 150 or 250°C. Normalized releases of the actinides tended to be higher in the sodium bicarbonate solution than in the other two leachants. Increasing the temperature from 150 to 250°C increased the releases of the actinides. Technetium releases were highest in the deionized water and bicarbonate leachants. Increasing the test temperature also increased the release of Tc. As a group the actinides all leached less than silicon but the technetium releases were always higher than silicon.

Many of the nonradioactive components of the glass were released more to the WIPP "B" brine than to either of the other two leachants. Silicon, boron, and molybdenum did not follow this trend but behaved more like Tc in that their releases were lowest in the salt brine. Except for the alkaline earths, the normalized releases based on the nonradioactive species increased with temperature and with increasing time of leaching.

Solubility constraints dictated by the leachate pH and species such as $CO_2^=$, which can complex with species leached from the glass, appear to control the release of many species from the glass.

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APPENDIX A

SOLUTION ANALYSES AND NORMALIZED MASS LOSS DATA

TABLE A.1. Normalized Releases to 150°C Deionized Water

	g/m^2														
	2 d			4 d			8 d			16 d			32 d		
Element	Undoped	Doped	Doped & Plateout	Undoped	Doped	Doped & Plateout	Undoped	Doped	Doped & Plateout	Undoped	Doped	Doped & Plateout	Undoped	Doped	Doped & Plateout
B	37	47		52	36		39	120		140	170		180	230	
Ba	0.46	0.16		0.19	0.59		0.53	0.11		0.086	0.25		0.087	0.055	
Ca	16	2.4		7.7	22		14	2.0		9.1	2.5		1.2	1.3	
Cd	(a)	0.47		1.1	1.0		1.1	1.4		1.4	1.4		1.8	2.8	
Ce				0.27											
Cr	2.7	2.9		4.0	3.6		3.4	9.9		9.3	13		15	17	
Cs	32			42			31			66			66		
Eu								0.23			0.23			0.23	
Fe		0.58		0.21	0.45		0.10	0.42		0.56	1.0		0.33	0.34	
Gd				3.7				1.4			1.0			1.4	
La				1.3	0.07			0.092			0.061			0.061	
Mo	29	40		45	34		31	100		110	150		150	210	
Na	39	56		55	67		58	140		140	180		170	240	
Nd				0.15										0.019	
Ni	1.7	0.96		3.7	1.6		0.80	2.1		3.2	10		2.6	2.6	
P	9.3	18		15	4.5		4.1	60		49	67		70	71	
Ru	0.13	0.12		0.35	0.17		0.16	0.42		0.39	0.50		0.54	0.67	
Si	29	38		40	56		48	73		86	110		100	140	
Sr	2.7	1.7		1.2	5.3		3.9	0.94		0.56	1.3		0.61	0.55	
Te	3.4			4.7			4.4			1.1			11		
Ti	0.018	0.016		0.018	0.027		0.012	0.031		0.012	0.031			0.031	
U	1.1			3			0.46	3.9		2.7	3.9		3.1	3.7	
Zn	1.6	0.10		0.77	0.31		0.22	0.63		1.3	4.2		1.5	1.1	
Zr	0.024	0.021		0.057	0.059		0.016	0.094		0.049	0.14		0.056	0.19	
Tc		720	810		400	400		380	380		200	200		200	210
U		0.70	1.6		1.2	3.0		5.0	7.7		3.0	5.7		5.3	8.2
Np		1.7	2.2		2.3	2.7		1.7	2.1		1.8	2.5		2.1	2.6
Pu		0.010	0.013		0.007	0.060		0.010	0.072		0.011	0.15		0.031	0.14
Am		0.007	0.095		0.013	0.047		0.024	0.064		0.016	0.062		0.035	0.072
Cm		0.007	0.042					0.022	0.071		0.017	0.061		0.005	0.013

(a) Blanks indicate not detected or a component of the leachate.

TABLE A.2. Normalized Releases to 150°C 0.03M Sodium Bicarbonate Solution

	g/m^2														
	2 d			4 d			8 d			16 d			32 d		
Element	Undoped	Doped	Doped Plateout	Undoped	Doped	Doped Plateout	Undoped	Doped	Doped Plateout	Undoped	Doped	Doped Plateout	Undoped	Doped	Doped Plateout
B	57	68		75	91		110	130		150	190		190	230	
Ba	1.1	3.9E-1		8.5E-1	4.8E-1		3.1E-1	3.9E-1		4.3E-1	2.7E-1		1.9E-1	1.9E-1	
Ca	19	8.8		22	16		12	14		10	11		8.4	9.2	
Cd	(a)	1.8			2.1			2.3			2.7			8.7	
Ce	3.4E-1	1.4		1.9E-1	1.1			2.0			2.3			1.5	
Cr	2.6	4.9		4.2	6.3		5.3	5.9		7.6	18		11	18	
Cs	47			60			79			94			96		
Eu	3.6E-1	1.4		3.5E-1	1.3			1.8			2.3			2.6	
Fe	1.4E-1	1.6E-1		2.7E-1	3.0E-1		1.2E-1	1.8E-1		1.7E-1	4.3E-1		7.1E-2	2.9E-1	
Gd	7.3	27		6.2	22			37			43			31	
La		1.0			9.3E-1			1.5			1.7		.	1.4	
Mo	49	61		62	81		84	110		120	170		150	200	
Na															
Nd	1.3E-1	6.3E-1		6.0E-2	5.0E-1			9.1E-1			1.1			7.3E-1	
Ni	9.3E-1	1.6		6.0E-1	2.3			1.9			2.1			2.1	
P	39	63		52	86		81	78		86	150		76	120	
Ru	3.1E-1	1.2		3.0E-1	1.2		1.2E-1	1.7			2.2			1.9	
Si	44	46		52	73		69	69		81	110		98	140	
Sr	11	4.9		6.7	5.5		4.8	6.9		6.3	4.1		3.7	3.1	
Te	4.1	4.6E-1		3.5			5.0	1.6		5.6	1.3E-1		6.2		
Ti	4.1E-2	1.0E-1		6.0E-2	9.8E-2		3.1E-2	1.4E-1		3.0E-2	1.7E-1		1.8E-2	1.2E-1	
U	31	46		35	40		36	69		56	81		39	63	
Zn	4.7E-1	1.3E-1		6.2E-1	3.0E-1		2.0E-1	2.9E-1		2.4E-1	4.6E-1		2.4E-1	5.8E-1	
Zr	4.1E-2	1.9E-1		5.0E-2	1.9E-1		1.6E-2	2.7E-1		1.6E-2	3.5E-1		4.2E-2	3.1E-1	
Tc		82	85		180	180		200	200		190	190		480	480
U		52	55		43	43		76	76		64	66		94	97
Np		16	18		24	24		21	22		19	20		23	24
Pu		5.9E-2	2.2E-1		1.0E-1	1.6E-1		8.9E-3	9.8E-3		6.2E-2	1.7E-1		6.9E-2	2.6E-1
Am		3.9E-1	2.6		3.5E-1	4.8E-1		2.8E-1	3.8E-1		2.0E-1	3.0E-1		5.4E-1	6.8E-1
Cm		4.6E-3	2.0E-2					2.9E-3	2.0E-2		1.7E-3	2.3E-2		1.2E-3	2.5E-2

(a) Blanks indicate not detected or a component of the leachate.

TABLE A.3. Normalized Releases to 150°C WIPP "B" Brine

	g/m²														
	2 d			4 d			8 d			16 d			32 d		
Element	Undoped	Doped	Doped & Plateout	Undoped	Doped	Doped & Plateout	Undoped	Doped	Doped & Plateout	Undoped	Doped	Doped & Plateout	Undoped	Doped	Doped & Plateout
B	18	27		18	39		26	42		41	58		80	76	
Ba	16	14		17	6.6		7.7	5.8		5.3	5.8		5.7	4.5	
Ca	(a)														
Cd															
Ce															
Cr															
Cs	40			33			58			89			140		
Eu															
Fe	4.0	2.0		12	2.1		1.4	2.3			1.2		5.7		
Gd															
La															
Mo	15	24		15	39		22	37		31	51		54	65	
Na															
Nd															
Ni															
P															
Ru															
Si	18	12		18	17		18	14		19	15		22	25	
Sr															
Te															
Ti															
U															
Zn	15	19		18	26		16	28		29	31		57	45	
Zr															
Tc		24	24		640	640		36	36		44	44		56	56
U		1.0	1.6		4.1E-1	1.2		5.5E-1	1.4		0.49	1.5		2.9E-1	1.1
Np		5.9	6.2		8.3	8.9		9.4	9.9		6.3	7.1		6.4	7.2
Pu		9.8E-2	1.6E-1		4.6E-1	2.1E-1		1.1E-1	3.0E-1		3.8E-2	2.3E-1		5.0E-2	2.0E-1
Am		3.3E-2	8.6E-2		1.3E-2	2.8E-2		9.1E-2	1.1E-1		2.7E-2	4.9E-2		1.1E-2	4.4E-2
Cm		1.6E-2	2.9E-2					1.6E-2	3.3E-2		1.2E-2	3.1E-2		2.5E-3	1.1E-2

(a) Blanks indicate not detected or a component of the leachate.

TABLE A.4. Normalized Releases to 250°C Deionized Water

	g/m²														
	2 d			4 d			8 d			16 d			32 d		
Element	Undoped	Doped	Doped Plateout	Undoped	Doped	Doped Plateout	Undoped	Doped	Doped Plateout	Undoped	Doped	Doped Plateout	Undoped	Doped	Doped Plateout
B	230	280		310			320	420		410			460	500	
Ba	0.17	0.31		0.22			0.22	0.22		0.15			0.13	0.16	
Ca	16	4.0		7.2			4.9	5.0		2.8			2.6	4.3	
Cd	1.4	3.7		3.3			3.9	5.7		4.6			4.6	5.9	
Ce	(a)	0.51						0.42					0.10	0.55	
Cr	63	71		64			95	72		110			130	100	
Cs	83			97			130			120			130		
Eu		0.68						1.0					0.36	1.1	
Fe	0.45	1.4		0.59			1.1	0.77		1.2			0.46	0.86	
Gd		10		1.3				9.4		1.6			3.1	12	
La		0.43						0.42						0.40	
Mo	180	250		240			270	400		330			360	450	
Na	220	310		310			320	470		380			430	520	
Nd		0.25						0.21						0.29	
Ni	2.2	2.0		2.8			2.9	2.7		3.3			3.3	2.6	
P	45	51		62			61	74		72			67	71	
Ru	1.2	1.7		1.3			1.5	1.9		2.4			2.6	2.6	
Si	160	230		220			230	320		260			310	360	
Sr	1.6	2.3		2.4			1.9	2.7		1.7			1.7	1.8	
Te	15			19			23			26			28	8.3	
Ti	0.035	0.14		0.018			0.061	0.072		0.061			0.031	0.076	
U	9.1	19		11			12	20		14			14	20	
Zn	1.8	1.0		1.9			0.86	0.89		0.61			0.38	0.86	
Zr	0.10	0.31		0.13			0.15	0.37		0.19			0.22	0.43	
Tc		410	420		400	400		440	440		2600	2600		810	870
U		33	39		27	36		35	43		27	34			
Np		2.0	3.5		3.7	4.5		5.6	12		2.0	8.5		2.2	13
Pu		0.090	0.32		0.085	0.24		0.045	1.6		0.027	0.10		0.026	1.1
Am		0.13	0.21		0.27	0.36		0.25	0.38		0.086	0.17			
Cm		0.018	0.071											0.005	0.015

(a) Blanks indicate not detected or a component of the leachate.

TABLE A.5. Normalized Releases to 250°C 0.03M Sodium Bicarbonate Solution

	g/m²														
	2 d			4 d			8 d			16 d			32 d		
Element	Undoped	Doped	Doped & Plateout	Undoped	Doped	Doped & Plateout	Undoped	Doped	Doped & Plateout	Undoped	Doped	Doped & Plateout	Undoped	Doped	Doped & Plateout
B	220	270		340			370	440		390			440	520	
Ba	0.19	0.11		0.31			0.086	0.41		0.27			0.13	0.17	
Ca	7.1	3.5		9.0			3.7	9.2		3.9			5.5	4.1	
Cd	(a)	5.0		1.1			1.1	5.1		1.1			0.73	6.0	
Ce		0.77						1.2						0.76	
Cr	57	57		62			91	95		110			110	160	
Cs	90			110			170			140			140		
Eu		1.1						1.6						1.4	
Fe	0.14	0.40		0.13			0.13	0.36		0.11			0.24	0.16	
Gd		17						25						17	
La		0.70						0.95						0.60	
Mo	170	240		260			300	410		320			340	460	
Na															
Nd		0.40						0.62						0.41	
Ni		1.8		0.46				2.8		0.61				2.7	
P	21	45		43			64	64		47			39	57	
Ru		1.6		0.31			0.62	2.5		0.39			0.36	2.7	
Si	140	200		190			230	310		120			220	330	
Sr	2.1	1.6		3.0			2.0	3.8		2.7			2.9	1.9	
Te	6.1			13			15			15			14	8.4	
Ti		0.070		0.012			0.037	0.12						0.076	
U	11	24		17			18	36		15			5.5	23	
Zn	0.20	0.47		0.90			0.27	0.28		0.21			0.10	0.52	
Zr	0.016	0.27		0.080			0.15	0.46		0.056			0.073	0.49	
Tc		460	460		580	580		1400	1500		750	770		670	700
U		32	35		43	46		65	67		19	24			
Np		1.6	2.6		3.3	9.2		2.0	3.1		2.1	13		1.4	22
Pu		0.049	0.44		0.058	1.7		0.49	1.5		0.10	0.29		0.10	4.5
Am		0.086	0.12		0.25	0.33		0.47	0.55		0.050	0.11			
Cm		0.003	0.023											0.002	0.027

(a) Blanks indicate not detected or a component of the leachate.

TABLE A.6. Normalized Releases to 250°C WIPP "B" Brine

	g Glass/m^2														
	2 d			4 d			8 d			16 d			32 d		
Element	Undoped	Doped	Doped Plateout	Undoped	Doped	Doped Plateout	Undoped	Doped	Doped Plateout	Undoped	Doped	Doped Plateout	Undoped	Doped	Doped Plateout
B	89	110		99			120	180		170			180	190	
Ba	11	5.4		17			3.5	7.8		15			8.6	4.1	
Ca	(a)														
Cd	140			140			140			240			250		
Ce				5.6			32			4.3					
Cr	26			34			37			70			58		
Cs	120			190			220			310			170		
Eu				5.3			3.5			3.6			3.7		
Fe															
Gd				95			40			94			68	93	
La				39			30			50			31		
Mo	37	80		61			70	120		84			92		
Na															
Nd				2.0			1.3			2.0			2.0		
Ni							6.7			13			6.8		
P															
Ru				5.3			2.7			5.5			5.5		
Si	31	17		20			22	21		23			20	26	
Sr															
Te				20			20			31			20		
Ti				0.24			0.24			0.24			0.24		
U				8.2						5.5			37		
Zn	37	66		21			22	26		65			58	10	
Zr				0.97						0.58			0.99		
Tc		130	130		130	130		440	440		83	85		180	180
U		0.55	3.0		0.88	6.3		0.39	5.1		0.25	10			
Np		0.71	2.8		0.90	12		0.41	5.4		0.61	12		0.53	13
Pu		0.33	0.96		0.066	0.88		0.093	1.8		0.18	0.39		0.35	3.5
Am		0.10	0.12		0.021	0.090		0.021	0.11		0.026	0.12			
Cm		0.017	0.037											0.006	0.077

(a) Blanks indicate not detected or a component of the leachate.

TABLE A.7. Elemental Concentrations (mg/L) in 150°C Deionized Water Leachate[a]

	Time									
	2 d		4 d		8 d		16 d		32 d	
Element	Undoped	Doped[b]	Undoped	Doped[c]	Undoped	Doped[b]	Undoped	Doped[b]	Undoped	Doped[b]
B	9.99	9.84	14.4	6.92	10.8	25.5	39	35.5	50	48.7
Ba	0.02	0.01	0.01	0.01	0.03			0.01		
Ca	2.16	0.26	1.04	1.95	1.82	0.20	1.2	0.26	0.16	0.13
Cd	(d)									0.01
Ce			0.02							
Cr	0.71	0.06	0.11	0.06	0.09	0.20	0.24	0.26	0.40	0.35
Cs	2.89		3.76		2.81		5.92		6.01	
Eu										
Fe	0.24	0.29	0.14	0.20	0.06	0.21	0.36	0.50	0.21	0.16
Gd			0.01							
La			0.06							
Mo	4.01	4.54	6.39	3.22	4.59	11.7	16	16.2	21	22.8
Na	33.2	36.8	47.0	39.9	49.6	91.5	119	122	149	164
Nd			0.02							
Ni	0.03	0.01	0.05	0.02	0.01	0.02	0.48	0.12		0.03
P	0.18	0.27	0.30	0.06	0.08	0.91	0.94	1.03	1.4	1.1
Ru	0.01	0.01	0.03	0.01	0.01	0.02	0.03	0.03	0.04	0.04
Si	51.9	51.3	68.9	65.9	84.1	100	150	151	175	197
Sr	0.08	0.04	0.04	0.11	0.11	0.02	0.02	0.03	0.02	0.01
Te	0.07		0.09		0.08		0.21		0.22	
Ti										
U	0.04		1.05		0.17	1.1	1	1.1	1.1	1
Zn	0.59	0.03	0.29	0.08	0.08	0.18	0.48	1.21	0.55	0.33
Zr			0.01	0.01		0.01	0.01	0.01	0.01	0.02
Tc		23		12		11		5.7		6.0
U		1.1E-1		1.8E-1		7.6E-1		6.9E-1		8.1E-1
Np		0.04		0.06		0.04		0.07		0.05
Pu		2.6E-5		1.8E-5		2.6E-5		4.4E-5		7.9E-5
Am		3.9E-5		7.3E-5		1.3E-4		1.3E-4		2.0E-4
Cm		6.0E-6		---		1.9E-5		1.5E-5		4.6E-6

(a) Concentrations of acidified, unfiltered samples measured at room temperature.
(b) Nonradioactive components measured in leachate from curium-doped beads.
(c) Nonradioactive components measured in leachate from technetium-doped beads.
(d) Blanks indicate not detected or a component of the leachate.

TABLE A.8. Elemental Concentrations (mg/L) in 150°C 0.03M $NaHCO_3$ Solution Leachate[a]

	Time									
	2 d		4 d		8 d		16 d		32 d	
Element	Undoped	Doped[b]	Undoped	Doped[c]	Undoped	Doped[b]	Undoped	Doped[b]	Undoped	Doped[b]
B	15.8	14.5	2.06	17	28.8	27.1	42	40.9	51	49.5
Ba	0.05	0.01	0.04	0.02	0.01	0.01	0.02	0.01	0.01	0.01
Ca	2.53	0.92	2.99	1.44	1.61	1.45	1.35	1.19	1.1	0.96
Cd	[d]					0.01		0.01		0.02
Ce	0.03	0.1	0.02	0.7		0.14		0.16		0.11
Cr	0.07	0.1	0.11	0.11	0.14	0.12	0.20	0.37	0.28	0.36
Cs	4.22		5.30		7.07		8.54		8.64	
Eu		0.01		0.01		0.01		0.01		0.01
Fe	0.09	0.08	0.17	0.14	0.08	0.09	0.11	0.21	0.05	0.14
Gd	0.03	0.08	0.02	0.06		0.10		0.12		0.09
La		0.03		0.03		0.05		0.06		0.05
Mo	6.8	6.7	8.7	7.88	12.0	12.4	17.7	18.8	21.6	22.6
Na	690	703	728	712	745	748	770	827	808	892
Nd	0.02	0.07	0.01	0.05		0.09		0.11		0.08
Ni	0.01	0.02	0.01	0.02		0.02		0.02		0.02
P	0.75	0.98	1	1.14	1.6	1.20	1.7	2.34	1.5	1.83
Ru	0.02	0.07	0.02	0.06	0.01	0.10		0.13		0.11
Si	77	63.2	92	85.8	120	93	141	155	170	188
Sr	0.32	0.11	0.2	0.11	0.14	0.16	0.19	0.09	0.11	0.07
Te	0.08	0.01	0.07		0.10	0.02	0.11		0.12	
Ti	0.01	0.01	0.01	0.01	0.01	0.02	0.01	0.02		0.02
U	11	13.1	13	10	13	19.2	20	23	14	17.8
Zn	0.17	0.04	0.23	0.08	0.64	0.08	0.09	0.13	0.09	0.17
Zr	0.01	0.02	0.01	0.02		0.03		0.03	0.01	0.03
Tc		2.4		5.3		5.9		5.5		14
U		8.3		6.5		1.2		1.5		1.5
Np		0.43		0.62		0.54		0.74		0.60
Pu		1.5E-4		2.7E-4		2.4E-5		2.4E-4		1.8E-4
Am		2.1E-3		1.9E-3		1.5E-3		1.6E-3		2.9E-3
Cm		4.0E-6		---		2.6E-6		1.5E-6		1.0E-6

(a) Concentrations of acidified, unfiltered samples measured at room temperature.
(b) Nonradioactive components measured in leachate from curium-doped beads.
(c) Nonradioactive components measured in leachate from technetium-doped beads.
(d) Blanks indicate not detected or a component of the leachate.

TABLE A.9. Elemental Concentrations (mg/L) in 150°C WIPP "B" Brine Leachate[a]

	Time									
	2 d		4 d		8 d		16 d		32 d	
Element	Undoped	Doped[b]	Undoped	Doped[c]	Undoped	Doped[b]	Undoped	Doped[b]	Undoped	Doped[b]
B	7.1	7.65	7.1	9.65	9.1	11.1	14	14.3	25	18.2
Ba	0.8	0.55	0.82	0.25	0.4	0.25		0.25	0.3	0.20
Ca	723	745	750	735	680	790	690	720	670	710
Cd	0.1									
Ce	(d)						1.3			
Cr										
Cs	5.95		5.2		8.49		10.7		14.9	
Eu									0.05	
Fe	3.79	2.3	9.2	2.2	2.2	2.5	1.1	1.9	5.0	0.65
Gd					0.6					
La					3		3.5			
Mo	2.2	2.8	2.1	3.7	3.1	4.2	4.8	5.6	7.6	7.25
Na		86000		95000		110000		98000		89000
Nd									0.7	
Ni			0.4							
P										
Ru										
Si	34	19	34	23	35	22	38	24	42	36
Sr	28	28	30	28	27	30	28	29	29	27
Te										
Ti										
U	9		7		13		14		11	
Zn	5.7	5.4	6.8	6.6	6	8.3	11	9.1	21	13
Zr							0.1			
Tc		0.69		19		1.0		1.3		1.6
U		1.6E-1		6.4E-2		8.8E-2		1.1E-1		4.4E-2
Np		0.15		0.22		0.24		0.24		0.17
Pu		2.6E-4		1.2E-4		2.8E-4		1.5E-4		1.3E-4
Am		1.8E-3		7.1E-5		5.0E-4		2.2E-4		5.8E-5
Cm		1.4E-5		---		1.4E-5		1.0E-5		2.1E-6

(a) Concentrations of acidified, unfiltered samples measured at room temperature.
(b) Nonradioactive components measured in leachate from curium-doped beads.
(c) Nonradioactive components measured in leachate from technetium-doped beads.
(d) Blanks indicate not detected or a component of the leachate.

TABLE A.10. Elemental Concentrations (mg/L) in 250°C Deionized Water Leachate[a]

	Time									
	2 d		4 d		8 d		16 d		32 d	
Element	Undoped	Doped[b]	Undoped	Doped[c]	Undoped	Doped[b]	Undoped	Doped[b]	Undoped	Doped[b]
B	63	60.8	84		88	79.6	111		126	108
Ba	0.01	0.01	0.01		0.01	0.01	0.01		0.01	0.01
Ca	2.12	0.43	0.96		0.65	0.45	0.38		0.34	0.45
Cd	(d)	0.01	0.01		0.01	0.01	0.01		0.01	0.01
Ce		0.04				0.03			0.01	0.04
Cr	1.65	1.43	1.70		2.50	1.27	2.89		3.37	2.09
Cs	7.6		8.8		11.6		10.3		11.7	
Eu										0.01
Fe	0.29	0.71	0.37		0.68	0.33	0.73		0.29	0.43
Gd		0.03	0.01			0.02	0.01		0.01	0.04
La		0.01				0.01				0.01
Mo	24.8	28.1	33.9		37	37.5	46		52	50.3
Na	190	202	270		276	272	340		372	349
Nd		0.03				0.02				0.03
Ni	0.03	0.02	0.04		0.04	0.03	0.05		0.05	0.03
P	0.88	0.80	1.2		1.19	0.98	1.4		1.29	1.08
Ru	0.09	0.10	0.10		0.11	0.10	0.18		0.20	0.15
Si	284	307	376		392	386	454		524	473
Sr	0.05	0.05	0.07		0.06	0.06	0.05		0.05	0.04
Te	0.03		0.38		0.44		0.52		0.53	0.13
Ti	0.01	0.02			0.01	0.01	0.01		0.01	0.01
U	3.3	5.4	4		4.4	4.9	5		5	5.6
Zn	0.66	0.30	0.70		0.32	0.23	0.23		0.14	0.25
Zr	0.01	0.03	0.02		0.02	0.03	0.02		0.03	0.04
Tc		10		13		12		110		24
U		5.2		5.3		4.2		6.3		
Np		0.05		0.15		0.10		0.08		0.06
Pu		2.4E-4		1.2E-4		2.2E-4		1.7E-4		6.7E-5
Am		6.9E-4		1.4E-3		1.5E-3		6.9E-4		
Cm		1.6E-5								4.2E-6

(a) Concentrations of acidified, unfiltered samples measured at room temperature.
(b) Nonradioactive components measured in leachate from curium-doped beads.
(c) Nonradioactive components measured in leachate from technetium-doped beads.
(d) Blanks indicate not detected or a component of the leachate.

TABLE A.11. Elemental Concentrations (mg/L) in 250°C 0.03M $NaHCO_3$ Solution Leachate[a]

	Time									
	2 d		4 d		8 d		16 d		32 d	
Element	Undoped	Doped[b]	Undoped	Doped[c]	Undoped	Doped[b]	Undoped	Doped[b]	Undoped	Doped[b]
B	60	58.3	91		99.3	83.8	106		118	113
Ba	0.01		0.02			0.01	0.01		0.01	0.01
Ca	0.94	0.36	1.19		0.5	0.85	0.52		0.72	0.42
Cd	(d)	0.01				0.01				0.01
Ce		0.06				0.07				0.05
Cr	1.5	1.16	1.61		2.42	1.69	2.73		2.9	3.28
Cs	8.05		9.75		15.4		13.1		12.2	
Eu		0.01				0.01				0.01
Fe	0.09	0.20	0.08		0.08	0.16	0.07		0.15	0.08
Gd		0.05				0.06				0.05
La		0.02				0.03				0.02
Mo	24.6	26.6	37.3		41.9	38.8	44.7		48.8	51.9
Na	840	904	954		936	1010	928		1028	1110
Nd		0.04				0.06				0.04
Ni		0.02	0.01			0.03	0.01			0.03
P	0.42	0.69	0.85		0.96	0.85	0.90		0.75	0.89
Ru		0.09	0.02		0.05	0.13	0.03		0.03	0.16
Si	242	268	330		408	350	213		388	456
Sr	0.06	0.04	0.09		0.06	0.08	0.08		0.08	0.04
Te	0.12		0.25		0.29		0.29		0.27	0.13
Ti		0.01			0.01	0.01				0.01
U	3.8	6.74	6		6.7	8.67	5.5		2	6.44
Zn	0.07	0.13	0.33		0.10	0.07	0.08		0.04	0.15
Zr		0.03	0.01		0.02	0.04	0.01		0.01	0.05
Tc		12		17		4.2		32		20
U		5.0		10		6.6		4.3		
Np		0.04		0.05		0.09		0.08		0.04
Pu		1.2E-4		1.3E-3		1.5E-4		4.0E-4		2.6E-4
Am		4.7E-4		2.7E-3		1.3E-3		3.9E-4		
Cm		2.9E-6								1.9E-6

(a) Concentrations of acidified, unfiltered samples, measured at room temperature.
(b) Nonradioactive components measured in leachate from curium-doped beads.
(c) Nonradioactive components measured in leachate from technetium-doped beads.
(d) Blanks indicate not detected or a component of the leachate.

TABLE A.12. Elemental Concentrations (mg/L) in 250°C WIPP "B" Brine Leachate[a]

	Time									
	2 d		4 d		8 d		16 d		32 d	
Element	Undoped	Doped[b]	Undoped	Doped[c]	Undoped	Doped[b]	Undoped	Doped[b]	Undoped	Doped[b]
B	24	26.6	27		32	35.3	46		49	42
Ba	0.5	0.2	0.8		0.25	0.25	0.7		0.4	0.15
Ca	697	735	650		660	690	640		590	605
Cd	0.4		0.4		0.4		0.7		0.7	
Ce	0.8		1.7		1.5		1.6		1.1	
Cr	0.7	0.08	0.9		1.0		1.8		1.5	
Cs	13.2		19.8		21.8		27.4		17.9	
Eu	[d]		0.08		0.07		0.07		0.07	
Fe	0.5	0.9	0.4		0.5		0.8		0.9	
Gd	0.3		0.8		0.6		0.8		0.7	
La	2		4		3.6		4.5		3.7	
Mo	5.2	8.8	8.6		10	11.6	12		13	10.4
Na		99,000				97,000				103,000
Nd	0.5		1		0.9		1		1	
Ni					0.1		0.2		0.1	
P										
Ru			0.8		0.6		0.8		0.8	
Si	56	27	34		38	28	40		35	38
Sr	30	29	17		29	30	30		30	30
Te			0.4				0.6		0.4	
Ti			0.1				0.1		0.1	
U	8		18		15		17		26	
Zn	14	19	7.8		8.1	6.6	24		21	3.0
Zr			0.2				0.15		0.2	
Tc		3.2		13		3.8		3.1		5.3
U		8.5E-2		6.0E-2		1.4E-1		5.6E-2		
Np		0.02		0.01		0.02		0.02		0.01
Pu		8.8E-4		2.4E-4		1.7E-4		7.1E-4		9.0E-4
Am		5.6E-4		1.2E-4		1.1E-4		2.1E-4		
Cm		1.5E-5								$5.E^{-6}$

(a) Concentrations of acidified unfiltered samples, measured at room temperature.
(b) Nonradioactive components measured in leachate from curium-doped beads.
(c) Nonradioactive components measured in leachate from technetium-doped beads.
(d) Blanks indicate not detected or a component of the leachate.

APPENDIX B

SUPPLEMENTAL LEACH TESTS AND RESULTS

APPENDIX B

SUPPLEMENTAL LEACH TESTS AND RESULTS

At the same time that the Waste Package Program was supporting the research presented in the body of this report, the High-Level Waste Immobilization Program (HLWIP) at PNL was also studying the hydrothermal reactions of 76-68 glass. The experiments were designed so that the results obtained from tests funded by one program supplemented and complemented the results obtained through the other program. The work reported by Westsik and Peters (1981) included data from both programs. To complete the data base of the studies on the hydrothermal reactions of 76-68 glass, a brief experimental description and the raw leachate analyses from the tests sponsored by the HLWIP are provided in this appendix.

EXPERIMENTAL DESCRIPTION

Right circular cylinders of 76-68 glass were leached for 3 days in 190 mL of deionized water at 25, 50, 100, 1560, 200, 250, 300 and 350°C. Additional tests were conducted for 1, 12, 27, 48, and 64 days in deionized water at 100 and 250°C. The glass specimens weighed about 18.5 g and had a geometric surface area of 18.8 cm^2. The sample surface area to leachate volume ratio was then 0.099 cm^{-1}. Table B.1 lists the composition of the glass as determined by ICP analyses of glass samples fused in sodium peroxide and in potassium hydroxide.

The leach vessels were Inconel and Hastelloy autoclaves fitted with valves, tubing and a cooler to permit taking samples of the leachate while the system was at the operating temperature. All tests were run at 2000 psi using argon as a cover gas except for the tests at 350°C, which were run at 2750 psi.

At the conclusion of a leaching period, approximately 10 mL of leachate were bled off to flush the sampling system. A 100-mL sample was then taken. Dilutions of 10X and 100X were immediately made. The autoclave system was then allowed to cool to room temperature. During cooling, the glass specimens were supported above the remaining leachate. Finally, the specimen was

TABLE B.1. Analyzed Composition of 76-68 Glass Used in HLWIP Hydrothermal Studies

Oxide	Weight Percent	Oxide	Weight Percent
SiO_2	38.5	RuO_2	0.8
Na_2O	13.4	Cr_2O_3	0.7
Fe_2O_3	11.6	Al_2O_3	0.6
B_2O_3	9.6	P_2O_5	0.5
U_3O_8	4.7	La_2O_3	0.5
ZnO	4.6	BaO	0.4
TiO_2	2.8	SrO	0.3
CaO	2.0	TeO_2	0.3
MoO_3	1.8	NiO	0.3
ZrO_2	1.5	MgO	0.3
Nd_2O_3	1.4	Gd_2O_3	0.1
Ce_2O_3	1.0	MnO_2	0.1
Cs_2O	0.8		

removed and cross sectioned to determine the reaction layer thickness. The remaining leachate was sampled, the pH was measured, and the leachate and dilutions were analyzed by ICP. The results are listed in Table B.2.

TABLE B.2. Specimen Data and Leachate Analyses for HLWIP Hydrothermal Studies

Temperature (°C)	25	50	100	150	200	250	300	350	350	100	100	100	100	100	100	250	250	250	250	250
Time (days)	3	3	3	3	3	3	3	3	3	1	12	27	27	48	64	1	12	27	48	64
Initial Weight (g)	18.3551	18.5952	18.5692	18.4584	18.5426	18.6097	18.5328	18.4966	18.4503	18.6086	18.3927	18.5525	18.5525	18.7191	18.7313	18.6133	18.5421	18.5035	18.2960	18.7307
Weight Lost (g)	0.0004	0.0006	0.0098	0.0607	0.1405	0.2478	0.7009	3.6675	2.3757	0.0073	0.0290	0.0501	0.0501	0.0720	0.0146	0.1908	0.3566	0.3917	0.4619	0.4959
Final pH	8.9	7.7	9.4	9.4	9.6	10.3	9.6	9.1	9.8	9.4	9.6	9.6	9.6	9.7	9.2	9.7	9.9	10.0	10.0	9.9
Reacted Glass Thickness (nm)				0.024	0.067	0.10	0.19	2.8	3.0							0.07	0.12	0.096	0.12	0.13
Leachate Concentrations (mg/L)																				
Si	0.081	0.387	17.8	89.1	203	351	638	440	926	11.5	42.6	76.1	76.1	94.8	29.1	262	455	511	557	587
Na		0.40	9.33	60.2	134	236	679	2680	2630	7.95	31.7	64.8	64.8	87.2	16.8	183	346	375	429	485
Fe					1.72		2.02	1.32												
B	0.011	0.044	3.39	21.3	47.7	95.7	383	2300	2520	1.81	8.96	20.6	20.6	28.2	3.74	54.2	110	126	144	157
U				0.82	2.1	1.9	2.8	3.9						5.2		3.6	2.8	2.7	1.9	3.3
Zn	0.072	0.078		1.53			0.30	0.24						0.42						
Ti							0.026	0.03		0.003					0.004	0.009	0.01			
Mo	0.040	0.161	1.58	8.78	18.9	32.4	142	1050	1040	1.2	4.19	8.47								
Zr				0.004		0.015	0.06	0.45	0.42	0.003			8.47	11.5	2.09	23.3	45.1	50.9	61.2	65.2
Ce					0.026		0.11	0.40	0.53		0.03					0.019	0.036	0.042		0.044
Cs	0.025	0.013	0.87	5.13	10.4	17.4	29	292	342	0.69	2.6	4.32								
Ru				0.042		0.20		0.93	1.0		0.018			0.03						
Cr				0.143	0.93	0.10		1.67				0.026	4.32	3.9	2.6	13.5	15.5	15.7	18.2	13.8
P														0.06		0.06	0.05			0.13
Ba	0.014	0.009	0.021	0.006					0.008	0.009			0.026		0.020	0.044	0.062	0.058	0.064	0.078
Sr	0.006	0.007	0.15	0.015		0.032				0.06	0.006	0.016		0.85	0.27	0.63	0.90	1.1	1.2	1.56
Te				0.16	0.38	0.48	0.99				0.065									
															0.004	0.004	0.007		0.006	0.006
													0.016		0.007	0.013				
																0.13			0.127	0.46

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