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R. D. Peters
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October 1981

Prepared for the
Office of Nuclear Waste Isolation
under its Contract with the
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Pacific Northwest Laboratory
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ACTINIDE LEACHING FROM WASTE GLASS: AIR-EQUILIBRATED VERSUS DEAERATED CONDITIONS

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SUMMARY

The principal objective of this work is to determine the effects of dissolved atmospheric oxygen on the leaching of plutonium, neptunium, and uranium from a simulated borosilicate waste glass. This is an important comparison, since many leach tests are performed in air-equilibrated solutions and actual repository waters may be very low in dissolved oxygen.

Leach tests were conducted in aearated and deaearated solutions using glass containing ^{239}Pu, ^{237}Np and ^{238}U. These tests were conducted at temperatures of 25° and 75°C and in deionized water, 0.03M $NaHCO_3$ and WIPP "B" salt brine for periods up to 341 days. In some cases, removal of 99% of the atmospheric oxygen had little or no effect on leaching of actinides. Under other conditions, significant dissolved oxygen dependencies were observed.

We found that deaerated leaching compares with aerated leaching as follows:

- Neptunium leaching was decreased by factors of 10 to 100 (depending on leach time) in the deaerated solutions at 75°C.
- Plutonium leaching decreased by factors of 3 to 5 due to deaeration, but only in the deionized water leachate at 25°C.
- Uranium leaching in salt brine and deionized water at 25°C was decreased by factors of 2 to 5 in deaerated solutions.

Time and temperature dependencies were also observed for the leaching of the actinides during the course of this work. After the first leach interval (2 days), the time dependent release curve for Pu was essentially flat or decreasing under all conditions, and maximum Pu solution concentration (at 25°C), as implied by release in aerated leachate, agrees with independent solubility data. The low ^{239}Pu releases observed in leach solutions are consistent with accumulation of ^{239}Pu (which is an alpha emitter) on the leached glass surface as determined by surface analysis using alpha energy spectroscopy. The amounts of uranium and neptunium leached increased with time under most conditions.

Temperature has a recognized strong effect of increasing the leach rate of waste glass matrix material. However, for Pu leaching, temperature has a small effect in deaerated leachates and negative effect in aerated leachates. Neptunium leaching generally increase with temperature under aerated conditions, but not in proportion to increases of matrix element leaching. In deaerated leachates, Np leaching decreases with temperature. Uranium leaching increases with temperature under aerated and deaerated conditions but not in proportion to matrix element increases. Arrhenius concepts, although applicable to matrix element release, do not readily explain actinide leaching.

Redox conditions probably affect the oxidation states of the actinides which, in turn, affect solubility. The precise redox conditions in these tests are not accurately known since radiolysis due to the alpha activity of ^{237}Np and ^{239}Pu may have generated oxidants in the leachate. Experiments with redox-buffered leachates are needed to fully understand the effects of reducing or oxidizing conditions on actinide release from waste glass.

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INTRODUCTION

The Waste/Rock Interaction Technology (WRIT) program has performed waste form-aqueous solution leach tests aimed at investigating the effects of temperature, leachate, and flow conditions for various radionuclide releases (Bradley, Harvey and Turcotte 1979; Coles 1981). These investigations are useful in geologic repository source term calculations and in understanding waste form behavior for predictive leach modeling purposes. Most of the WRIT program's previous leach test work has been performed using leachates exposed to air. Repository water is not necessarily air-equilibrated and in some cases is essentially free of oxygen. It is conceivable that oxygen levels effect the valence state of actinides, and the variety of resulting chemical species have different solubilities (Cleveland 1979). It is therefore necessary to know what effect, if any, the presence of oxygen has on the leaching of actinides. Detailed interpretation of actinide behavior due to deaeration of the leachate will depend on a better understanding of actinide chemistry in the glass-leachate system.

This report compares static leaching of U, Pu, and Np in air-equilibrated and deaerated leachates. Since two different temperatures and three different leachates were used, the effects of these parameters are also reported. Measurements of alpha-activity in the leachate and on leach container walls is the principal means of analysis. In some cases, the surface of leached glass was analyzed by alpha-spectroscopy. This provides depth distribution information for ^{239}Pu.

EXPERIMENTAL DETAILS

The static method of leaching was utilized, in which glass sample and leach solutions are left undisturbed until the leach test is terminated (Bradley, McVay and Coles 1980). Parallel experiments were executed at Pacific Northwest Laboratory (PNL) and Argonne National Laboratory (ANL), except that at PNL, air-equilibrated leachate was used, and at ANL more than 99% of the dissolved oxygen was removed with N_2 sparging.

PNL 76-68 is a sodium borosilicate glass containing 34 elements; its composition is given Table 1. It is used in these experiments in the shape of near hemispherical beads. Properties of beads are given in Table 2.

LEACHING CONDITIONS

Each leaching experiment consisted of three beads in a polypropylene bottle and 45 ml of leaching solution. The ratio of the bead-geometric surface area to solution volume was about 0.1 cm^{-1}. Three leachates, deionized water, 0.03$\underline{M}$ $NaHCO_3$, and WIPP "B" salt brine whose composition is given by Bradley, Harvey and Turcotte (1979), were prepared from deionized distilled water. The bicarbonate concentration used here is higher than most groundwaters. The WIPP "B" brine represents the composition of ground water that has percolated through a bedded salt-deposit. For practical purposes, this brine is only 80% saturated. The temperatures were held to 25° ± 2°C and 75° ± 2°C. The duration of the leach tests were 2, 8, 16, 32, 106, and 341 days except in air equilibrated tests where the 16-day interval was omitted and a multi-year leaching test is in progress. Each time-interval sample started with fresh beads and fresh solutions; neither beads nor solutions were recycled. At the end of each test, leach solutions were decanted and the walls of the containers were stripped with an $HF\text{-}HNO_3$ solution to remove any actinide deposits.

The solutions exposed to air contained about $2.6 \times 10^{-4}\underline{M}$ (8.3 ppm) oxygen at 25°C (brine, about 20% less) and $1.7 \times 10^{-4}\underline{M}$ (5.4 ppm) O_2 at 75°C. The equilibrium carbonate concentration (as H_2CO_3 and HCO_3^-) of water in contact with air is $1.6 \times 10^{-5}\underline{M}$ at 25°C (Garrells and Christ 1965) and $5 \times 10^{-6}\underline{M}$ at 75°C. The anoxic leach tests were performed in glove boxes containing nitrogen

TABLE 1. Composition of 76-68 Waste Glass

Glass Composition, wt%		Glass Composition, wt%	
40.0	SiO_2	0.53	Pr_6O_{11}
12.5	Na_2O	0.46	$^{237}NpO_2$
9.6	Fe_2O_3	0.46	P_2O_5
9.5	B_2O_3	0.40	Cr_2O_3
5.0	ZnO	0.37	SrO
4.2	$^{238}UO_2$	0.32	Sm_2O_3
3.0	TiO_2	0.26	TeO_2
2.2	MoO_3	0.21	Y_2O_3
2.0	CaO	0.20	NiO
1.7	ZrO_2	0.17	Rh_2O_3
1.65	Nd_2O_3	0.13	Rb_2O
1.19	CeO_2	0.070	Eu_2O_3
1.07	RuO_2	0.050	Gd_2O_3
1.03	Cs_2O	0.046	$^{239}PuO_2$
0.56	BaO	0.033	CdO
0.53	PdO	0.031	Ag_2O
0.53	La_2O_3		

atmospheres that were circulated through oxygen removal equipment. The oxygen content of the glove boxes was measured continuously by O_2 analyzers and was below 10^{-5} atmospheres (1 pascal). Each leach solution was initially degassed with specially deoxygenated nitrogen. Sparging of $NaHCO_3$ solutions was limited because a rise in pH indicated the removal of CO_2. The degassing treatment is estimated to reduce levels of O_2 by factors of 100 to 1000. Thus, prior to the start of leach tests, O_2 concentrations were on the order of $10^{-6}\underline{M}$. However, it is possible that H_2O_2 or O_2 was generated via radiolysis of the water by the alpha-activity. The levels of radiolysis products that accumulated are not

TABLE 2. Properties of 76-68 Doped Glass Beads (from Bradley, Harvey and Turcotte 1979)

Mass (g/bead)	0.353 ± 0.013
Density g/cm^3	3.028 ± 0.038
Diameter (cm)	0.681 ± 0.008
Eccentricity (%)	0.5
Height (cm)	0.437 ± 0.005
Amount of Isotope g/g glass	^{237}Np 4.08×10^{-3} ^{239}Pu 4.13×10^{-4} ^{238}U 3.7×10^{-2}

known, degassing was not performed beyond the initial treatment of leachates. Post-sparging carbonate concentrations in deionized water are calculated to be less than 10^{-8} $\underline{M}$ assuming the N_2 used is free of CO_2.

In deaerated leachate tests, leach containers were enclosed in mason jars and these were enclosed in sealed modified pressure cookers. A nitrogen atmosphere was kept over the mason jars inside the pressure cookers. The mason jars (and the polypropylene) allowed water vapor in the 75°C experiments to slowly escape. Water was added to the mason jars in experiments started after this problem was recognized, but the 341-day 75°C $NaHCO_3$ leaching went dry (results not reported), the 75°C 106-day and 341-day brine solutions exhibited large salt precipitates causing large error estimates, and the 75°C, 341-day water leachate sample lost 20 of 45 ml.

In air-saturated tests, leach containers were mounted in covered waterbaths. Ingress of water was noted for some of the WIPP "B" salt brine tests at 75°C, either due to permeation through the plastic bottle wall or through a poor lid seal. Results from the 75°C WIPP "B" salt brine tests are not available.

DATA ANALYSIS

Neptunium-237 and plutonium-239 leachate activities were determined by alpha counting. Gamma-ray analysis of 10 ml solutions were used for most of the deaerated leachate Np determinations. Uranium concentration was determined chemically by laser fluorescence. Elemental analysis of leachates was also performed using induction coupled plasma spectroscopy (ICP).

Alpha spectroscopy analysis of the glass surface was also performed. This technique reveals the depth distribution of alpha-emitters in solids, in this case the profile of ^{239}Pu (α-decay, 5.155 MeV) was determined. A silicon surface barrier detector was used to provide the analysis of the surface of beads leached in deaerated water at 75°C and deaerated 0.03M $NaHCO_3$ at 25°C. Alpha spectra were then transformed into depth distributions. Experimental details and the method of transforming energy spectra to profiles are given by Diamond and Friedman (1980).

RESULTS

An effective way to examine these leach results is to express the total amount of a constituent released from the glass normalized by its mass fraction. This provides a relative comparison of the leaching of different glass constituents. Normalized release is defined as:

$$R(g/m^2) = a/XA$$

where

R = normalized release of the specified element

a = amount of element removed from waste solid sample in g during a leach interval

X = initial specific concentration of element in the waste solid in g/g-glass

A = geometric surface area of waste form sample, m^2

The quantity "a" includes what remains suspended or dissolved in the leachate at the end of the test plus what has deposited on the walls of the leach container; the leachates were not filtered. Release data for the actinides are given in Tables 3, 4, and 5 along with pH measurements of the leachate. Released data for Mo, which is an indicator of glass matrix leaching, are given in Tables 3 and 4. Figures 1, 2, and 3 compare actinide release in a particular leachate for the two temperatures (25°C and 75°C) and for the two leach conditions of air saturated or N_2-sparged solutions. The distribution of leached material between leach solution and container walls is given in the Appendix. In general Np and U did not tend to plate out whereas the majority of leached Pu plated out in many tests. Leach solution concentrations are also given in the Appendix.

The saw-tooth appearance in the plots indicate there are fairly large uncertainties in the data. Possible reasons for this erratic release behavior will be mentioned under "Discussion." It is beyond the scope of this project

TABLE 3. Normalized Releases (g/m^2) and Solution pH from Leaching in Deionized Water

	Leach Time, days	Deaerated					Aerated				
		$^{(239)}$Pu	$^{(237)}$Np	U	Mo	pH	^{239}Pu	^{237}Np	U	Mo	pH
25°C	2	0.009	0.07	0.017	0.1	6.9	0.042	0.07	0.03	0.2	7.2
	8	0.014	0.11	0.006	0.1	6.6	0.033	0.2	0.04	0.2	7.5
	32	0.011	0.29	0.005	0.4	6.7	0.032	0.3	0.05	0.4	8.1
	106	0.010	0.53	0.04	0.4	9.1	0.084	0.3	1.5	1.3	---(a)
	341	0.013	0.80	0.24	1.2	9.2	0.024	1.3	0.12	1.2	---
75°C	2	0.05	0.08	0.05	3.5	7.3	0.009	1.0	0.06	1	7.2
	8	0.006	0.01	0.24	10.0	7.7	0.011	1.4	0.12	4	8.5
	32	0.012	0.04	0.51	27.0	8.6	0.015	1.4	0.22	9	8.6
	106	0.007	<0.01	0.93	46.0	9.6	0.018	2.0	0.90	28	---
	341	0.008	0.02	0.69	130	9.7	0.011	2.3	1.0	45	---

(a) Dashed lines (---) indicate measurements were not taken.

TABLE 4. Normalized Releases (g/m^2) and Solution pH from Leaching in 0.03<u>M</u> $NaHCO_3$

	Leach Time, days	Deaerated					Aerated				
		^{239}Pu	^{237}Np	U	Mo	pH	^{239}Pu	^{237}Np	U	Mo	pH
25°C	2	0.04	0.1	0.005	0.2	9.7	0.078	0.2	0.16	0.1	9.1
	8	0.08	0.2	0.03	0.3	9.5	0.088	0.3	0.29	0.2	9.2
	32	0.08	0.6	0.90	2.3	9.6	0.095	0.8	0.80	0.7	9.1
	106	0.08	1.1	2.2	1.1	9.9	0.022	0.8	0.60	0.7	---(a)
	341	0.09	0.4	(b)	2.3	10.2	0.081	2.6	3.0	2.6	---
75°C	2	0.027	0.4	0.17	6	9.6	0.013	2.2	16	2.5	9.1
	8	0.004	0.6	18.0	18	9.8	0.022	5.4	7.1	6.6	8.9
	32	0.007	0.2	28	46	9.6	0.014	8.9	14	14	9.4
	106	0.005	0.2	33	60	10.8	0.018	16.0	29	35	---
	341	---	---	---	---	---	0.016	17.0	56	68	---

(a) Dashed lines (---) indicate measurements were not taken.
(b) Suspect measurement, not reported.

TABLE 5. Normalized Releases (g/m^2) and Solution pH from Leaching in WIPP "B" Brine

	Leach Time, days	Deaerated				Aerated			
		^{239}Pu	^{237}Np	U	pH	^{239}Pu	^{237}Np	U	pH
25°C	2	0.015	0.1	0.01	5.9	0.028	0.2	0.1	5.6
	8	0.032	0.2	0.14	6.7	0.013	0.2	0.3	6.4
	32	0.021	0.3	0.16	6.6	0.016	0.3	0.4	6.9
	106	0.031	0.5	0.36	8.6	0.007	0.3	0.6	---(a)
	341	0.015	0.5	0.02	8.7	0.009	0.7	1.4	---
75°C	2	0.015	0.5	0.03	6.8	---	---	---	
	8	0.030	0.5	0.46	6.8	---	---	---	
	32	0.018	1.0	0.35	7.1	---	---	---	
	106	0.017	1.5	0.86	8.4	---	---	---	
	341	0.020	1.2	0.30	7.7	---	---	---	

(a) Dashed lines (---) indicate measurements were not taken.

to repeat experiments and obtain satistical confidence limits. Differences in leaching less than a factor of 2 will therefore be considered insignificant.

An understanding of the dependence of 76-68 matrix leaching as a function of time, temperature and oxygen exposure is helpful in interpreting actinide release dependence on test parameters. The prominent matrix elements in 76-68 glass are Si, and B; however, in the deaerated test, only Mo in bicarbonate and deionized water was analyzed. These three constituents, as well as Na, are released from 76-68 glass at rates within a factor of two of each other and follow an Arrehnius dependence between 50° to 150°C (Westsik and Peters, 1980). Molybendum release is, therefore, used as an indicator to compare matrix leaching of 76-68 at the leach conditions in these tests. The comparison is limited to data taken in bicarbonate and deionized water solutions since Mo data are not available in deaerated brine.

Knowledge of actinide behavior on the surface of leached glasses is useful in the interpretation of release data obtained from solution analysis. The α-energy spectra from ^{239}Pu that is incorporated in 76-68 glass has been used

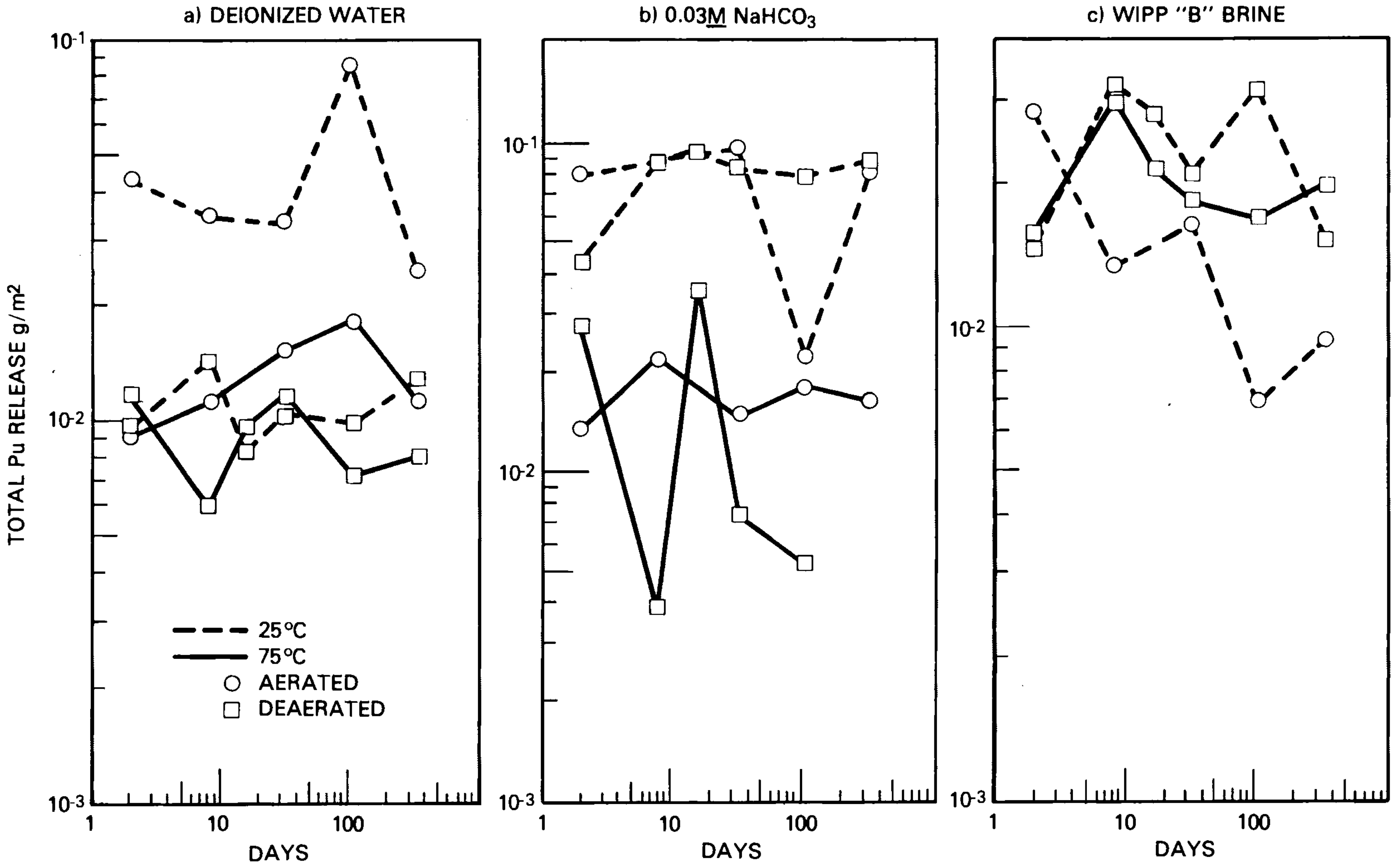

FIGURE 1. Normalized Release of ^{239}Pu from 76-68 Glass at 25° and 75°C in Static a) Deionized Water, b) 0.03M NaHCO$_3$ and c) WIPP "B" Salt Brine

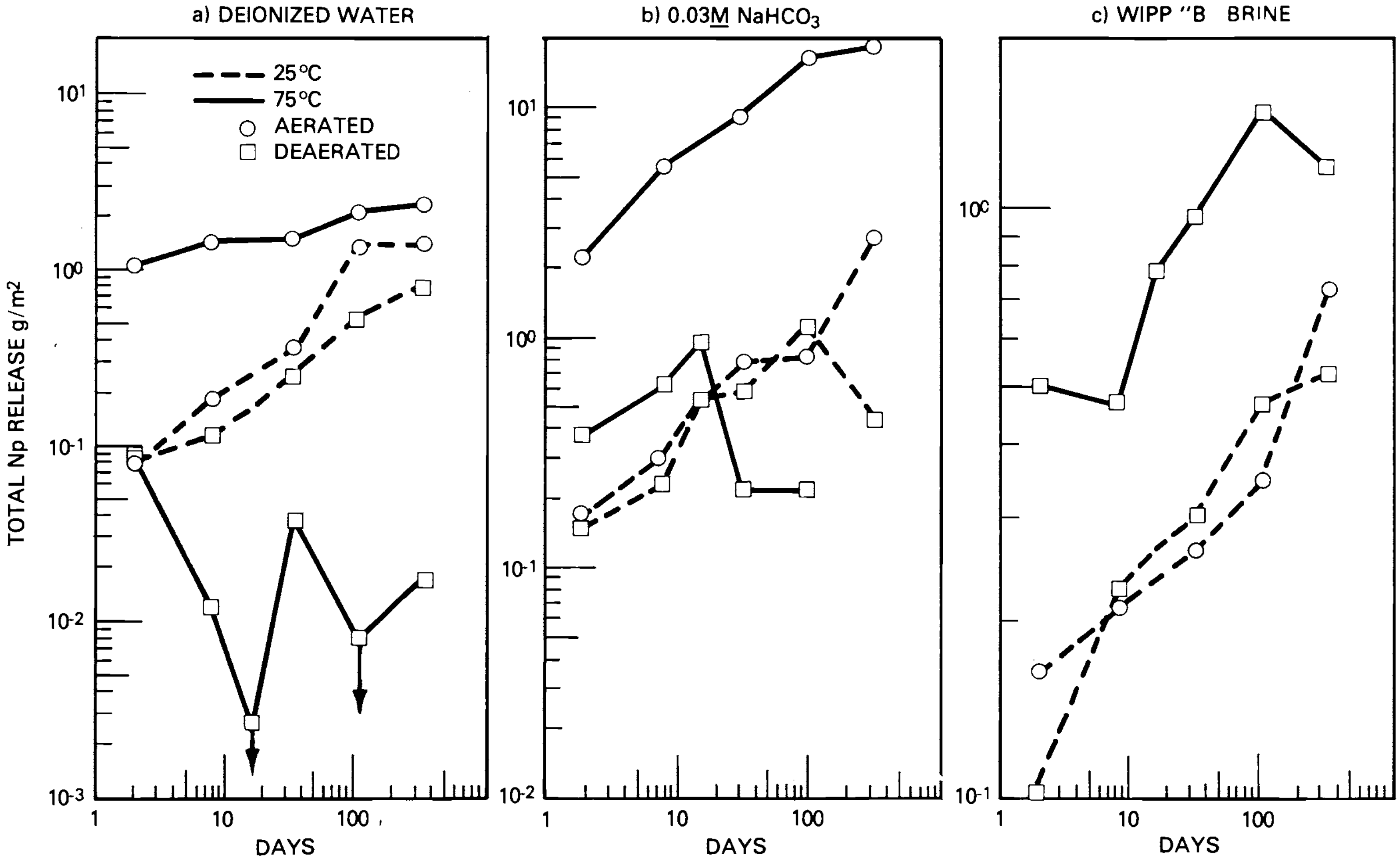

FIGURE 2. Normalized Release of ^{237}Np from 76-68 Glass at 25° and 75°C in Static a) Deionized Water, b) 0.03M $NaHCO_3$ and c) WIPP "B" Salt Brine. Arrows indicate upper limits to release

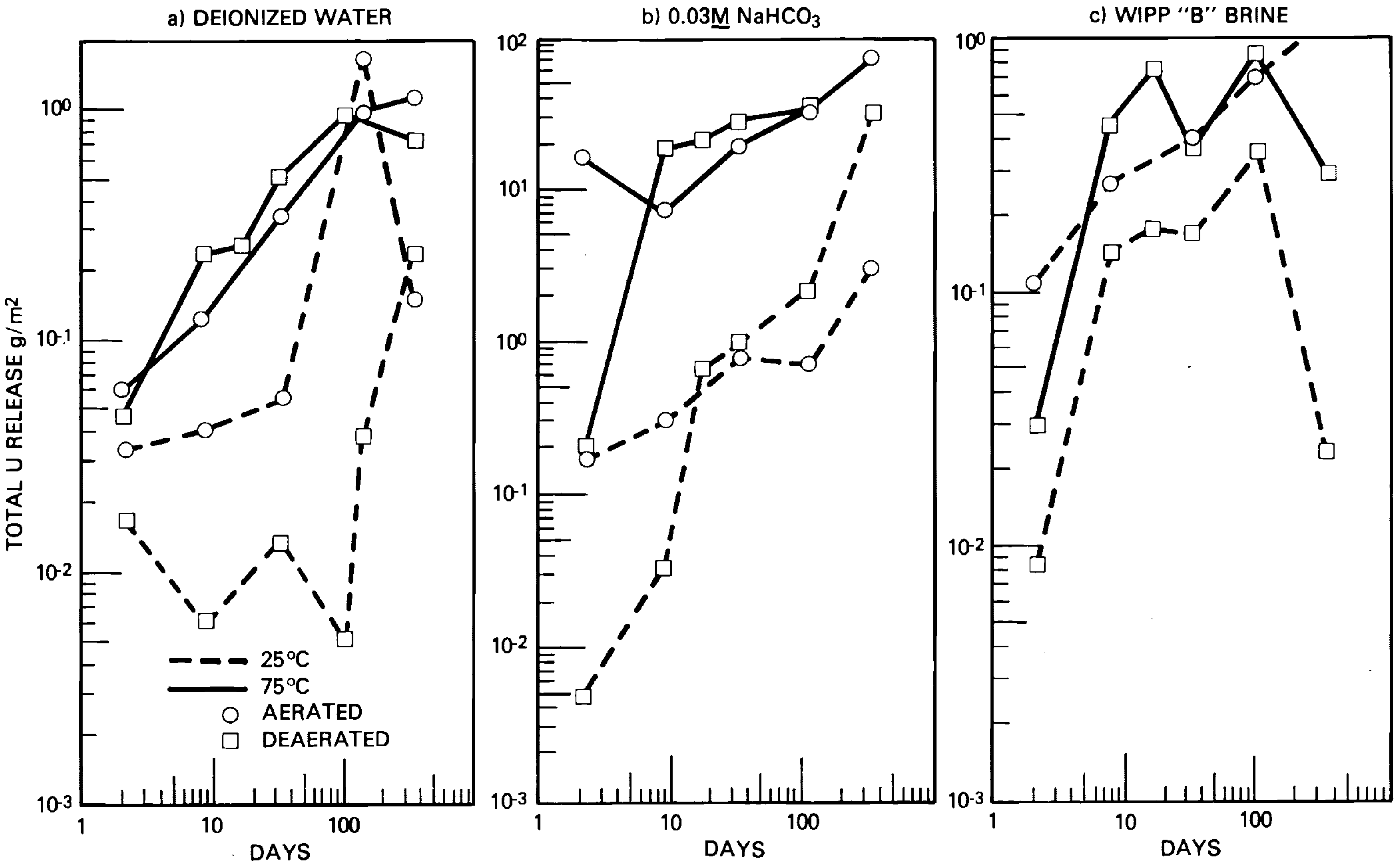

FIGURE 3. Normalized Release of Uranium from 76-68 Glass at 25° and 75°C in Static a) Deionized Water, b) 0.03M NaHCO$_3$ and c) WIPP "B" Salt Brine

here to investigate the near surface (within 10 to 15 μm) distribution of this species as a function of leaching progress. Alpha spectra were obtained from beads leached under the anoxic conditions at 75°C in deionized water and at 25°C in 0.03M $NaHCO_3$. Leaching in bicarbonate at 25°C did not significantly alter the surface Pu profile, probably due to the sluggishness of matrix leaching at 25°C. Only profiles from glass leached in 75°C water will be discussed. Profiles obtained from α-energy spectra are given in Figure 4. For the conversion to depth it is assumed that the leached surface is of uniform composition and density and bulk values for 76-68 glass are taken for these properties. Actually, the surface composition and probably the density has been altered by the leaching process and the extent of these alterations is not precisely known. Further, the ^{237}Np alpha activity, whose energy is 0.37 MeV less than ^{239}Pu (5.16 MeV) is treated as if it were ^{239}Pu alpha-activity. Therefore, the depth profiles obtained by the α-spectra must be regarded as qualitative.

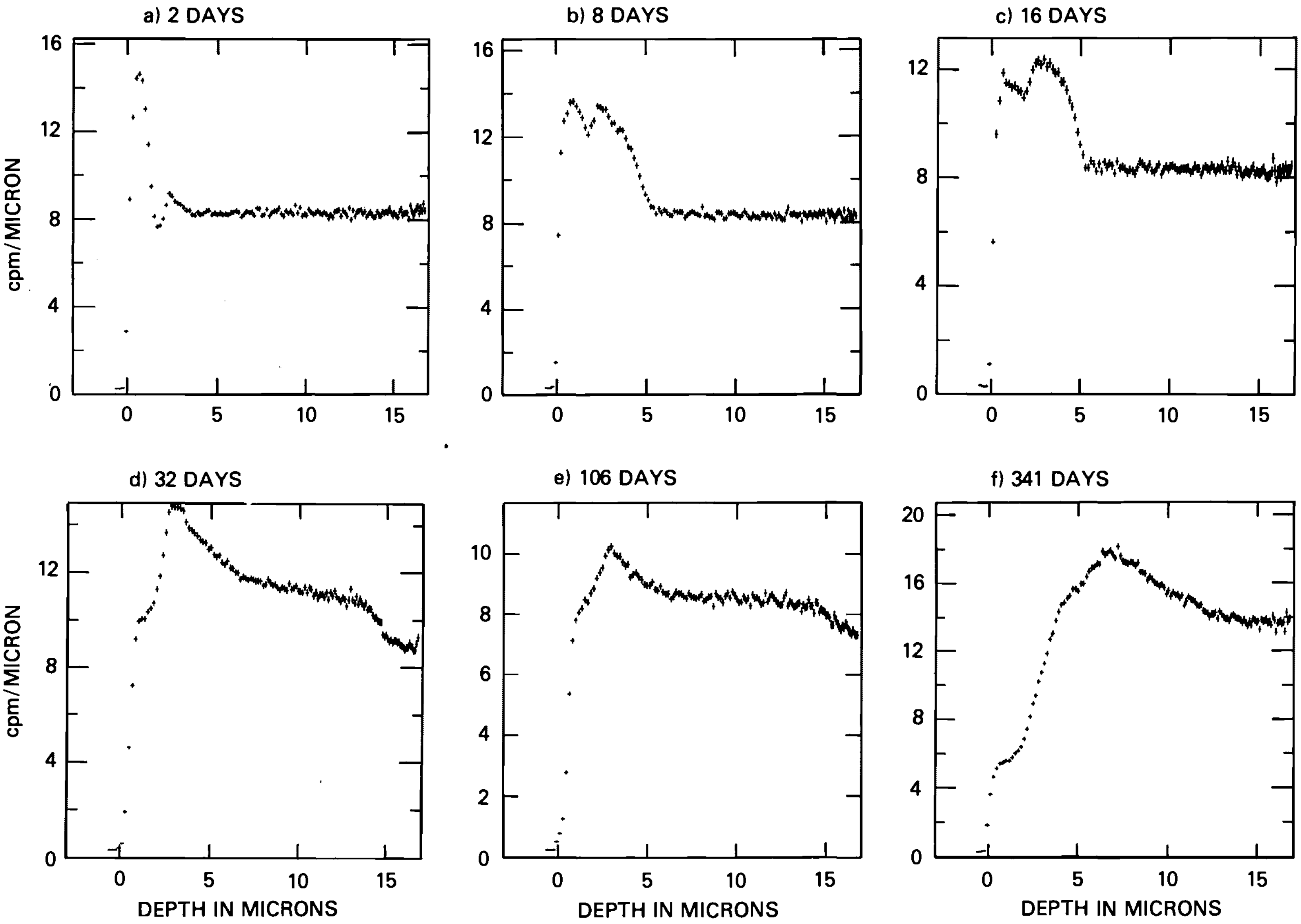

FIGURE 4. Alpha Spectroscopy Profiles of ^{239}Pu on the Surface of 76-68 Glass Leached for Various Times in 75°C Deaerated Water

DISCUSSION

The effects of deaeration on the leaching of actinides are summarized in Table 6. Uranium and Pu seem to respond similarly to deaeration, both showing decreases in release at 25°C and essentially no consistent effects at 75°C. Neptunium, on the other hand, appears to be affected only at 75°C, and by much larger factors than for U and Pu at 25°C. Depending on the leaching time, Np at 75°C decreases up to 100-fold due to deaeration of the solution.

The changes in actinide leaching due to the presence or absence of O_2 are not explained by changes in the corrosion rate of glass. At 25°C, Mo release (which is an indicator of glass breakdown) is not affected by deaeration. At 75°C, Mo release increases due to the removal of O_2 whereas Np decreases. It has not been demonstrated that Mo follows Si and B leaching in low oxygen solutions, therefore, Mo may not be as effective a matrix indicator in deaerated leach test results.

To some extent, deaeration appears to affect the temperature dependance of actinide release. In aearated deionized water, Pu release (Figure 1a) is lower by a factor of 5 at 75°C than at 25°C. But in the deaerated case, temperature had no effect on leaching. In deionized water and bicarbonate solution, Np release (Figure 2a) increases with temperature under aearated conditions but decreases with temperature in deaerated water.

The magnitude of release of the various actinides can be readily compared using Tables 3-5. Overall, releases of Pu are 10-1000 times less than corresponding Np and U releases. The order of Np and U depends upon aearation/deaeration. In aearated solution, normalized releases of U and Np are about the same (with U being higher in bicarbonate leachate). In deaerated leachate, the Np released is suppressed compared with U release.

Interpretation of these trends in actinide leaching behavior is difficult since the aqueous chemistry of actinides in the waste glass/leachate system is poorly understood. Potential ligands for complexation of actinides in this study are OH^-, Cl^-, $HSiO_3^-$, HCO_3^-, and HBO_3^-. In addition, a mixed carbonate-hydroxide complex has been reported for Pu (Jensen 1980).

TABLE 6. Factors Showing the Decrease in Actinide Leaching in Deaerated Solutions Relative to Leaching in Aerated Solutions[a]

	25°C			75°C	
	Deionized Water	0.03M $NaHCO_3$	WIPP "B" Salt Brine	Deionized Water	0.03M $NaHCO_3$
Pu	Decrease by 3 to 5	N	Decrease by 2 to 5	N	N
Np	N	N	N	Decrease by 50 to 100	Decrease by 10 to 100
U	Decrease by 2 to 3	N	Decrease by 1.5 to 2	N	N

(a) The symbol N denotes no significant or consistent effect observed. Ranges of the decreases apply for different times.

Competition with actinides for these anions by other cations in the system (such as Na^+, Ca^{++}, Cs^+ and others available from the breakdown of 76-68 glass) is also occurring. Dissolved oxygen can affect the oxidation state of the actinides, but the exact species involved for such aqueous reactions are not known for these experiments. The remainder of this discussion will be concerned with a comparison of actinide release with glass matrix breakdown (via Mo release), solubility considerations, temperature and leachate composition effects.

Of the actinides studies here, the plutonium release data deviates most sharply from the Mo release data. Plutonium release does not increase consistently with time after the second leach interval and also decreases with increasing temperature. Coles (1981) also observed negative temperature dependance for Pu in flowing leach tests. This behavior suggests Pu solubility limits are achieved early in the leach process. The maximum Pu release in 25°C aerated water corresponds to a concentration in leachate solution of about $10^{-9}\underline{M}$ at a pH of 8. In calculating this concentration it is assumed that all of the material suspended in water is dissolved. The maximum Pu concentration is $10^{-10}\underline{M}$ at a pH of 9 in 25°C deaerated water. In the bicarbonate solution, the maximum for both aearated and deaerated conditions is about 1.5 x 10^{-9} $\underline{M}$ at pH 9 and 10, respectively. Rai, Serne and Moore (1980) measured PuO_2 and $Pu(OH)_4$ solubilities from a pH of 3.8 to 7.9. Extrapolation of this data to pH's of 8 or 9 gives solubilities which are comparable to these maximum concentrations.

At 25°C the Np release in water and bicarbonate is more comparable with Mo release than is the Pu release. Leaching of Np continually increases with time. Evidently Np solubility is not reached at 25°C during these tests. At 75°C, the Mo releases far exceed Np releases especially in the deaerated leachates where they differ by 2 or 3 orders of magnitude. Although more Np is leached at 75°C than at 25°C, the amount of Np released does not increase with temperature in proportion with Mo. Solubility or sorption seems to limit Np release at 75°C much more than at 25°C.

Uranium releases agree with Mo release much better in bicarbonate than in deionized water for both temperatures and atmospheres. The bicarbonate leachate pH remains higher than that in deionized water leachate after 106 days (9.2 versus 8.3), and this may influence the solubility of uranium. It is also possible that a soluble carbonate complex of U is formed (Jensen 1980).

Some of the data of Figures 1 through 3 are erratic, having a saw tooth-type release dependance with respect to time. Several uncontrolled variables may have caused this. There could be variations in glass specimens from test to test. Glass leached at these times and temperatures are affected only in the near surface region (first 10 to 15μm) and these surfaces may be different due to differences in exposure to air or humidity. Also portions of the leached layer (or "gel" layer) which contained signficant amounts of actinides may have spalled off from the glass surface causing some spikes in the leach data curves. It is not likely that analytical counting techniques contributed much error. Repeated analysis are usually reproducible to within 10%.

The key feature of the alpha-spectra profiles (Figure 4) is the increase in Pu activity near the solution/glass interface relative to unleached glass. This effect is noticeable even after two days as seen in Figure 4 and is characterized at this time by a narrow peak. As time progresses the peak broadens appreciably which probably corresponds to an increasing leached glass zone due to attack by water. The magnitudes of the peaks (the measured cpm/μm) do not change consistently but vary between 10 and 18 cpm/μm.

The accumulation of Pu on leached 76-68, as shown in Figure 4 is consistent with the leach data shown in Figure 1. The amount of Pu in the leachate never increases from the first leach interval, whereas the leaching of glass continues with time as discussed earlier in this report. There is an increasing level of Pu at the glass/water interface as indicated by increasing areas under the profiles in Figure 4. Conceivably Pu is released from the vitreous matrix due to attack by water but never leaves the leached glass surface. Either Pu is being sorbed in the gel layer or is solubility constrained, thus forming a crystalline precipitate. For the longer time intervals of 106 and 341 days, the peak seems to occur at progressively deeper locations--conceivably nonradioactive material deposits on the surface over the Pu.

The depths of leach alteration in deionized water, as implied by the α-profiles in Figure 4, are larger than what would be expected, based on alteration depths obtained from electron spectroscopy for chemical analysis (ESCA) as described by McVay and Pederson (1980). After 14 days leaching at 90°C, the alteration depth as implied by the ESCA study is 2.5 μm; whereas in Figure 4 the alteration depth appears to be about 3 μm after 8 days at 75°C. In estimating alternative depths from Figure 4, a correction of 2.2 μm must be subtracted from the depth scale. This correction is a result of interference of ^{237}Np activity.

CONCLUSIONS

Static leaching of actinide doped waste glass has been performed in the presence and near absence of atmospheric oxygen. In many cases initial removal of dissolved air to less than 10^{-6} $\underline{M}$ had a relatively small effect on leaching, but in some tests large reductions occurred. Leaching of Pu and U was reduced by factors less than 10 and Np by factors of 10 to 100. Plutonium and uranium were affected significantly at 25°C but not at 75°C and Np was affected at 75°C but not at 25°C. Leaching of matrix material (as indicated by Mo release) is unaffected by deaeration at 25°C and, in contrast to Np, is increased at 75°C. In most cases, the leaching of actinides from waste glass in aerated solutions will give conservative results compared to leaching in deaerated solutions.

General observations on the nature of actinide leaching were also made. Plutonium release is not generally dependent on the length of the leach period (beyond 2 days) and decreases with temperature under both aerated and deaerated conditions. Alpha-energy surface profiling show Pu building up at the surface of leached glass. The release of Np and U increases with time and temperature in most cases but not in proportion with glass matrix leaching. Glass matrix elements are leached faster than the actinides so actinide removal is not determined by matrix removal in static tests. The Arrhenius equation does not describe the temperature dependence of leaching of actinides from waste glass. It appears that solubility dominates the leaching of Pu and to a lesser extent controls Np and U release.

Detailed interpretation of the effects of the atmosphere and other parameters in this study will depend on a better understanding of aqueous leachate chemistry. The precise redox conditions after the start of leaching are not known in this work, since unbuffered solutions were used and also radiolysis (from α-emitters in the glass) occurred which may have introduced oxidants into the deaerated leachates. Leach tests performed with redox-controlled solutions are needed for a more precise determination of the effects of redox conditions on actinide release.

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APPENDIX

CONVERSION OF NORMALIZED RELEASE TO SOLUTION CONCENTRATION AND FRACTION RELEASED

APPENDIX

CONVERSION OF NORMALIZED RELEASE TO SOLUTION CONCENTRATION AND FRACTION RELEASED

Tables A.1 through A.3 give the fraction of actinide leached from glass which was found in the leachate solution. This quantity includes any material suspended in solution, dissolved or particulate. The remaining fraction deposited on contaner walls, as determined by analyzing the acid-wash solution used on emptied leach containers.

Tables A.4 through A.6 give the concentration of elements or isotopes in leach solutions. These concentrations are based on the fraction of leached material found in the leachate as given in Tables A.1 through A.3. Concentrations in A.4-6 are apparent since they include any suspended particulates.

TABLE A.1. Fraction of Total Actinide Leached from Glass in Deionized Water which Remained in Solution. The remaining fraction plated out on leach container walls

	Leach Time, Days	Deaerated			Aearated		
		^{239}Pu	^{237}Np	U	^{239}Pu	^{237}Np	U
25°C	2	0.33	0.92	0.75	0.91	1.0	0.86
	8	0.85	1.0	0.18	0.93	1.0	0.84
	32	0.28	1.0	0.74	0.77	0.95	0.86
	106	0.07	1.0	0.80	0.65	0.98	1.0
	341	0.27	1.0	0.97	0.43	1.0	0.90
75°C	2	0.27	0.43	1.0	0.26	0.99	0.25
	8	0.30	0.80	0.99	0.41	0.99	1.0
	32	0.20	0.20	0.97	0.67	0.95	1.0
	106	0.26	0.25	1.0	0.44	0.99	1.0
	341	0.39	0.48	0.95	0.05	0.98	1.0

TABLE A.2. Fraction of Total Actinide Leached from Glass in 0.03M Sodium Bicarbonate which Remained in Solution. The remaining fraction plated out on leach container walls

	Leach Time, Days	Deaerated			Aearated		
		^{239}Pu	^{237}Np	U	^{239}Pu	^{237}Np	U
25°C	2	0.99	1.0	0.60	0.97	1.0	1.0
	8	0.99	1.0	0.97	0.98	1.0	1.0
	32	0.77	1.0	1.0	0.87	0.99	0.99
	106	0.80	1.0	1.0	0.54	0.99	0.99
	341	0.55	0.81	1.0	0.69	0.99	1.0
75°C	2	0.20	0.90	0.91	0.27	0.99	1.0
	8	0.28	1.0	1.0	0.60	0.99	1.0
	32	0.52	0.94	1.0	0.70	0.99	1.0
	106	0.27	1.0	1.0	0.44	0.98	0.98
	341				0.21	0.99	0.99

TABLE A.3. Fraction of Total Actinide Leached from Glass in WIPP Brine which Remained in Solution. The remaining fraction plated out on leach container walls

	Leach Time, Days	Deaerated			Aearated		
		^{239}Pu	^{237}Np	U	^{239}Pu	^{237}Np	U
25°C	2	0.94	1.0	0.75	0.99	1.0	0.97
	8	0.92	0.94	0.90	0.94	1.0	0.98
	32	0.96	1.0	1.0	0.85	1.0	0.98
	106	1.0	1.0	1.0	0.32	1.0	0.98
	341	0.97	1.0	0.96	0.27	0.99	0.99
75°C	2	0.81	0.90	0.61			
	8	0.61	0.86	0.90			
	32	0.46	0.41	0.23			
	106	0.57	0.66	0.71			
	341	0.14	0.16	0.01			

TABLE A.4. Concentrations of Isotopes and Elements[(a)] in Deionized Water Leachate, μM[(b)]

	Leach Time, Days	Deaerated				Aearated			
		^{239}Pu	^{237}Np	U	Mo	^{239}Pu	^{237}Np	U	Mo
25°C	2	5×10^{-5}	2×10^{-2}	2×10^{-2}	2×10^{-1}	7×10^{-4}	2×10^{-2}	5×10^{-2}	3×10^{-1}
	8	2×10^{-4}	2×10^{-2}	2×10^{-3}	2×10^{-1}	5×10^{-4}	3×10^{-2}	6×10^{-2}	3×10^{-1}
	32	5×10^{-5}	5×10^{-2}	7×10^{-3}	1	4×10^{-4}	5×10^{-2}	8×10^{-2}	1
	106	1×10^{-5}	9×10^{-2}	6×10^{-2}	1	1×10^{-3}	5×10^{-2}	3	2
	341	6×10^{-5}	1×10^{-1}	4×10^{-1}	2	2×10^{-4}	2.4×10^{-1}	2×10^{-1}	2
75°C	2	5×10^{-5}	1×10^{-2}	9×10^{-2}	5	4×10^{-3}	2×10^{-1}	2×10^{-2}	2
	8	3×10^{-5}	2×10^{-3}	4×10^{-1}	15	8×10^{-5}	2×10^{-1}	2×10^{-1}	6
	32	4×10^{-5}	7×10^{-3}	9×10^{-1}	41	2×10^{-4}	2×10^{-1}	4×10^{-1}	14
	106	3×10^{-5}	2×10^{-3}	2	70	1×10^{-4}	3×10^{-1}	2	42
	341	5×10^{-5}	3×10^{-3}	1	200	1×10^{-5}	4×10^{-1}	2	68

(a) Concentrations are apparent, species containing these isotopes and elements are not necessarily dissolved.

(b) μM = 10^{-6}M.

TABLE A.5. Concentrations of Isotopes and Elements(a) in 0.03M $NaHCO_3$ leachate, μM(b)

	Leach Time, Days	Deaerated				Aearated			
		^{239}Pu	^{237}Np	U	Mo	^{239}Pu	^{237}Np	U	Mo
25°C	2	7×10^{-4}	2×10^{-2}	5×10^{-3}	3×10^{-1}	1×10^{-3}	3×10^{-2}	3×10^{-1}	2×10^{-1}
	8	1×10^{-3}	3×10^{-2}	5×10^{-2}	5×10^{-1}	2×10^{-3}	5×10^{-2}	5×10^{-1}	3×10^{-1}
	32	1×10^{-3}	1×10^{-1}	1.6	3	1×10^{-3}	1×10^{-1}	1	1
	106	1×10^{-3}	2×10^{-1}	4	2	2×10^{-4}	1×10^{-1}	1	1
	341	8×10^{-4}	6×10^{-2}		3	1×10^{-3}	5×10^{-1}	5	3.9
75°C	2	9×10^{-5}	6×10^{-2}	3×10^{-1}	9	6×10^{-5}	3	29	4
	8	2×10^{-5}	1×10^{-1}	32	27	2×10^{-4}	1	13	10
	32	6×10^{-5}	3×10^{-2}	50	70	2×10^{-4}	2	25	21
	106	2×10^{-5}	3×10^{-2}	60	90	1×10^{-4}	3	52	53
	341					6×10^{-5}	3	100	102

(a) Concentrations are apparent, species containing these isotopes and elements are not necessarily dissolved.

(b) μM = 10^{-6}M.

TABLE A.6. Concentrations of Isotopes and Elements(a) in WIPP "B" Brine leachate, µM(b)

	Leach Time, Days	Deaerated			Aearated		
		^{239}Pu	^{237}Np	U	^{239}Pu	^{237}Np	U
	2	2.4×10^{-4}	2×10^{-2}	1×10^{-3}	5×10^{-4}	3×10^{-2}	2×10^{-1}
	8	5×10^{-4}	3×10^{-2}	2×10^{-1}	2×10^{-4}	3×10^{-2}	5×10^{-1}
25°C	32	3×10^{-4}	5×10^{-2}	3×10^{-1}	2×10^{-4}	5×10^{-2}	7×10^{-1}
	106	5×10^{-4}	9×10^{-2}	6×10^{-1}	4×10^{-5}	5×10^{-2}	1
	341	2×10^{-4}	9×10^{-2}	3×10^{-2}	4×10^{-5}	1×10^{-1}	3
	2	2×10^{-4}	8×10^{-2}	3×10^{-2}			
	8	3×10^{-4}	8×10^{-2}	7×10^{-1}			
75°C	32	1×10^{-4}	7×10^{-2}	1×10^{-1}			
	106	2×10^{-4}	2×10^{-1}	1			
	341	3×10^{-4}	3×10^{-2}	5.4×10^{-3}			

(a) Concentrations are apparent, species containing these isotopes and elements are not necessarily dissolved.

(b) µM = 10^{-6}M.

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