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Waste Isolation Safety Assessment Program

Leaching of Actinides and Technetium From Simulated High-Level Waste Glass

D. J. Bradley
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August 1979

Prepared for the
Office of Nuclear Waste Isolation
under its Contract with the
U.S. Department of Energy

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

As part of continuing Department of Energy (DOE) sponsored studies in waste management, the Pacific Northwest Laboratory (PNL)[a] has been conducting the Waste Isolation Safety Assessment Program (WISAP) for the Office of Nuclear Waste Isolation (ONWI). The purpose of this program is to gather experimental data and develop models to assess the safety of nuclear waste isolation in geologic formations.

This report describes exerimental work undertaken by Task 2 of the Waste Isolation Safety Assessment Program with regard to the performance of nuclear waste forms in the geologic environment. Leach tests were conducted using a modified version of the International Atomic Energy Association (IAEA) procedure to study the behavior of glass waste-solution interactions. Release rates were determined for technetium, uranium, neptunium, plutonium, americium, curium, and silicon in the following solutions:

- WIPP "B" salt brine, NaCl (287 g/l)
- NaCl (1.76 g/l)
- $CaCl_2$ (1.66 g/l)
- $NaHCO_3$ (2.52 g/l)
- deionized water.

The leach rates for all elements decreased an order of magnitude from their initial values during the first 20 to 30 days leaching time. This is consistent with previously reported results. The element release rates were affected by an order of magnitude due to the leach solution composition. The sodium bicarbonate solution produced the highest elemental release rates, while the saturated salt brine and deionized water in general gave the lowest release.

Technetium has the highest initial release of all elements studied. The technetium release rates, however, decreased by over four orders of magnitude

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in 150 days of leaching time. In the prepared glass, technetium was phase separated, concentrating on internal pore surfaces. Further studies are required to establish the composition of this phase and to evaluate the general behavior of Tc in other glass compositions.

Neptunium, in all cases except $CaCl_2$ solution, shows the highest actinide release rate. In general, curium and uranium have the lowest release rates. The range of actinide release rates observed in the various solutions is summarized in the following table:

Solution Type	Actinide Release Rate Range ($g/cm^2/day$) At 1 Day	At 46 Days	At 150 Days
Salt brine	$4.3 \times 10^{-7} - 1.0 \times 10^{-5}$	$5.3 \times 10^{-8} - 9.7 \times 10^{-7}$	$2.2 \times 10^{-8} - 3.4 \times 10^{-7}$
$CaCl_2$	$4.4 \times 10^{-7} - 1.0 \times 10^{-5}$	$1.4 \times 10^{-7} - 8.5 \times 10^{-7}$	$1.6 \times 10^{-8} - 4.5 \times 10^{-7}$
NaCl	$5\text{-}3 \times 10^{-7} - 8.9 \times 10^{-6}$	$1.7 \times 10^{-7} - 8.9 \times 10^{-7}$	$4.3 \times 10^{-8} - 3.6 \times 10^{-7}$
$NaHCO_3$	$8.8 \times 10^{-7} - 9.6 \times 10^{-6}$	$5.4 \times 10^{-7} - 1.8 \times 10^{-6}$	$2.2 \times 10^{-7} - 9.9 \times 10^{-7}$
Deionized water	$2.2 \times 10^{-7} - 7.4 \times 10^{-6}$	$1.3 \times 10^{-7} - 8.7 \times 10^{-7}$	$4.8 \times 10^{-8} - 2.4 \times 10^{-7}$

This report constitutes the first progress report for the IAEA testing of doped waste glass as part of the Waste Isolation Safety Assessment Program. Future reports will provide more leach rate and surface analysis data. This information will be used to interpret future tests combining the waste form, canister, engineered barriers, and host rock into single tests.

CONTENTS

FIGURES

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CONCLUSIONS

The release rates of neptunium, uranium, plutonium, americium, curium, technetium, and silicon were determined from a simulated waste glass in five different leach solutions, ranging from a saturated salt brine to deionized water. In all cases, sodium bicarbonate solution produced the highest element release, as was expected. Release in general was lowest for the saturated salt brine and deionized water.

Neptunium displayed the highest release rates, while uranium and curium generally had the lowest release. The exceptions were a high release of americium to the calcium chloride solution, and uranium to the sodium bicarbonate solution.

The following more specific conclusions were made as a result of this study:

- The release rates of the actinide elements were lower than that for silicon in all cases except for neptunium, which had a release rate comparable to silicon.
- The leach rates decrease by an order of magnitude during the first 20 to 30 days of solution contact. This is consistent with previous studies.
- The range of leach rates for different solution types studied here is one order of magnitude.
- Technetium is phase-separated in the glass and concentrates on the pore surfaces within the glass matrix. Although it is in an apparent soluble form, its release will be controlled by dissolution of the silicon matrix.
- After over 400 days of leaching at 25°C, a glass surface reaction zone cannot be optically observed at 400X magnification, although there is a surface discoloration.
- The leach rate appears to have a small dependence on the frequency by which solutions are changed in the IAEA leaching procedure used in this study. The leach rates decrease as solutions are changed less frequently.

INTRODUCTION

Information concerning the release of radionuclides from candidate waste forms for geologic disposal is needed to evaluate the safety of geologic repository storage of nuclear wastes. These data are used both in release consequence modeling and as a source term for radionuclide migration experiments.

This report examines the leach testing of a simulated waste glass doped with the amounts of technetium, uranium, neptunium, plutonium, americium, and curium expected to be present in high-level waste. These elements constitute most of the potential long-term biological hazard associated with nuclear wastes. The glass was doped with these elments, some on a single dopant basis, so that its behavior could be studied in detail without the radiochemical interferences and radiation fields that would be encountered with a fully radioactive waste. Future tests will include a limited amount of work with these elements associated with fully radioactive wastes so that their behavior can be verified.

In addition to solution anlyses, some glass surface and microstructural examination was undertaken with the objective of further defining mechanisms of leaching. At the time of this report, the glass surface studies and solution analyses for glass constituents (other than Si) have not been completed. A definitive interpretation will therefore be made later. The intention of this report is primarily to make data available for the previously unknown dependence of actinide leach rates on ground water composition.

PREPARATION OF DOPED WASTE GLASS

In order to study isotopes of potential long-term hazard without the high radiation fields of actual high-level waste, sample waste forms were made using these isotopes alone. Four glasses, based on the same composition, were made incorporating the following isotopes:

- ^{99}Tc
- ^{244}Cm
- ^{233}U + ^{243}Am
- ^{237}Np + ^{239}Pu.

These isotopes were doped into the waste glass to the extent normally present in high-level waste. These amounts were determined by using the ORIGEN code (Bell 1973), which calculates the production of fission products and actinides in reactor fuels. The assumptions for this reference waste were:

- burnup = 33,000 MWd/MTU
- reactor type = reference PWR, 3.3% enriched
- age of waste = 2 years old
- reprocessing loss = 0.5% U + Pu
- specific power = 26.4 MW
- waste loading = 33% (fission products + actinidies + reprocessing chemicals).

The doped glasses, except for the ^{233}U + ^{243}Am doped glass, contained the expected amount of uranium as ^{238}U.

As a compromise between the reality of cracked glass monoliths and the powdered glass frequently studied in accelerated leach testing, glass beads were chosen as the sample waste form for study. A resistance heated unit was installed in a glove box for preparation of the doped glasses. The process involves heating a comparatively large volume of glass to the melt temperature, continuously forming droplets at the end of a nozzle, which fall and are collected on a heated metal plate as solid beads.

The procedure for making doped glass beads using the resistance heated unit has the following steps:

1. Frit and calcine (33% calcine for 76-68 glass) are placed in a platinum crucible and melted (3 hrs at 1050°C), then quenched and broken out of the crucible.

2. The weighed dopant is dissolved in 6N HNO_3, using a minimum of liquid. Concentrated HNO_3 with 5% HF is used to dissolve NpO_2 and PuO_2 yielding a clear solution. Aliquots of the solutions are retained for future analysis.

3. The premelted glass is crushed to a coarse powder, weighed, and thoroughly "wetted" with the dopant solution.

4. The wetted glass is vacuum dried and preheated to 500°C to reduce nitrate concentration that can cause glass frothing.

5. The doped glass is remelted in stainless steel crucibles for 2 hrs at 1050°C, water quenched, and broken out of the crucible.

6. The glass is placed in a quarter bead-making tube, the temperature adjusted from 1050° to 1100°C, and the beads collected on steel pans maintained at 400°C.

7. The beads are annealed for 2 hr at 500°C and cooled to ambient temperature at 100°C/hr.

The resulting glass composition for glass 76-68 is shown in Table 1, along with frit and calcine compositions. Table 1 is specific for the glass doped with ^{237}Np and ^{239}Pu.

Approximately 1.5 kg of doped glass beads were prepared in four separate runs. Table 2 summarizes the bead characteristics, including the concentrations of the dopant isotopes. The confidence limits shown are at one standard deviation. Thus, the beads had constant properties to within a few percent. This illustrates that a major value of this approach is reproducibility of sample preparation.

TABLE 1. Composition of Frit, Wastes and Doped Waste Glass Used for Actinide Release Studies

Frit Composition,[a] wt%		Waste Calcine Composition,[b] wt%		Glass Composition,[c] wt%	
59.7	SiO_2	29.2	Fe_2O_3	40.0	SiO_2
14.2	B_2O_3	15.0	Na_2O	12.5	Na_2O
11.2	Na_2O	12.6	$^{238}U_3O_8$	9.6	Fe_2O_3
7.45	ZnO	6.8	MoO[d]	9.5	B_2O_3
4.45	TiO_2	5.3	ZrO_2	5.0	ZnO
3.0	CaO	5.0	Nd_2O[e]	4.2	$^{238}UO_2$
		3.6	CeO_2	3.0	TiO_2
		3.2	RuO_2	2.2	MoO_3
		3.1	Cs_2O	2.0	CaO
		1.7	BaO	1.7	ZrO_2
		1.6	PdO	1.65	Nd_2O_3
		1.6	La_2O_3	1.19	CeO_2
		1.6	Pr_6O_{11}	1.07	RuO_2
		1.41	$^{237}NpO_2$	1.03	Cs_2O
		1.4	P_2O_5	0.56	BaO
		1.2	Cr_2O_3	0.53	PdO
		1.1	SrO	0.53	La_2O_3
		0.98	Sm_2O_3	0.53	Pr_6O_{11}
		0.78	TeO_2	0.46	$^{237}NpO_2$
		0.64	Y_2O_3	0.46	P_2O_5
		0.60	NiO	0.40	Cr_2O_3
		0.52	Rh_2O_3	0.37	SrO
		0.38	Rb_2O	0.32	Sm_2O_3
		0.21	Eu_2O_3	0.26	TeO_2
		0.15	Gd_2O_3	0.21	Y_2O_3
		0.140	$^{239}PuO_2$	0.20	NiO
		0.10	CdO	0.17	Rh_2O_3
		0.090	Ag_2O	0.13	Rb_2O
				0.070	Eu_2O_3
				0.050	Gd_2O_3
				0.046	$^{239}PuO_2$
				0.033	CdO
				0.031	Ag_2O

(a) 76-101 Frit
(b) PW-8a-3 Calcine } PNL Waste
(c) 76-68 Glass } Form Identifiers
(d) Chemical Stand-In for Mo + Tc
(e) Chemical Stand-In for Nd, Am and Cm

TABLE 2. Doped Glass Bead Characteristics

	Dopants			
	^{244}Cm	^{99}Tc	$^{237}Np/^{239}Pu$	$^{233}U/^{243}Am$
Beads Produced	400	450	2400	650
Mass (g/bead)	.459 ± .024	.362 ± .006	.353 ± .013	0.381 ± .005
Density (g/cm^3)	3.040 ± .004	3.042 ± .025	3.028 ± .038	2.974 ± .015
Diameter (cm)	0.759 ± .010	0.683 ± .008	0.681 ± .008	0.696 ± .005
Eccentricity (%)	0.7	0.3	0.5	0.3
Height (cm)	0.455 ± .005	0.437 ± .008	0.437 ± .005	0.442 ± .003
Amount of Isotope g/g glass	1.17×10^{-4}	4.6×10^{-3}	^{237}Np 4.08×10^{-3}	^{243}Am 8.62×10^{-4}
			^{239}Pu 4.13×10^{-4}	^{233}U 2.46×10^{-2}

Average "second" phase concentrations - for all beads

1-3% M_3O_4 - spinel, M + Fe, Cr, Ni, Zn

1% MO_2 - fluorite, M = U, Ce

0-3% porosity

MICROSTRUCTURAL EXAMINATION

Random samples of the beads were examined by x-ray diffraction, metallography, and α-radiography. X-ray diffraction traces of the various glass preparations are shown in Figure 1. Weak reflections from a spinel-type oxide (MO_2; M = U, Ce) are observed in the 1-3 wt% range. The actual concentration varies somewhat from sample to sample because these "difficult to solve" phases tend to form clusters. Small concentrations of Pd metal and RuO_2 are also present in the glass but ≤1 wt%. All of these phases are common to waste glass compositions. At this relatively low concentraton level and small crystal size (mainly 1≤ m), other studies have shown they do not significantly affect any of the properties of the glass, at least with regard to dispersibility.

Optical micrographs and autoradiographs are shown for the doped glasses in Figures 2 through 5.

The porosity of all of the beads, except for the ^{99}Tc beads, was less than 3%, which is lower than that measured in a full-scale canister of glass produced by the in-can melting (ICM) process (Larsen and Bonner, eds. 1976). In any case, the porosity will not affect the leaching studies now under way. Even after two years at a leach rate of 1×10^{-5} g/cm^2/day, the depth of glass penetrated would only be ∿20 μm.

The autoradiographs for ^{237}Np + ^{239}Pu, ^{233}U + ^{243}Am, and ^{244}Cm doped glasses show that these isotopes are very evenly distributed through the glass. For the case of the ^{99}Tc doped glass, the activity is concentrated on the surfaces of internal pores, giving rise to unusual leach behavior, as shown later. The leaching data presented should be regarded as preliminary, since the general phase behavior of Tc in other waste glasses is unknown.

Large spinel phase inclusions, which form by corrosion of the steel melting crucible (and also occur in full-scale ICM glasses), are seen to be highly depleted in actinides, as expected. The porosity in the glass tends to give a false impression of some variation in track density, and some dark spots

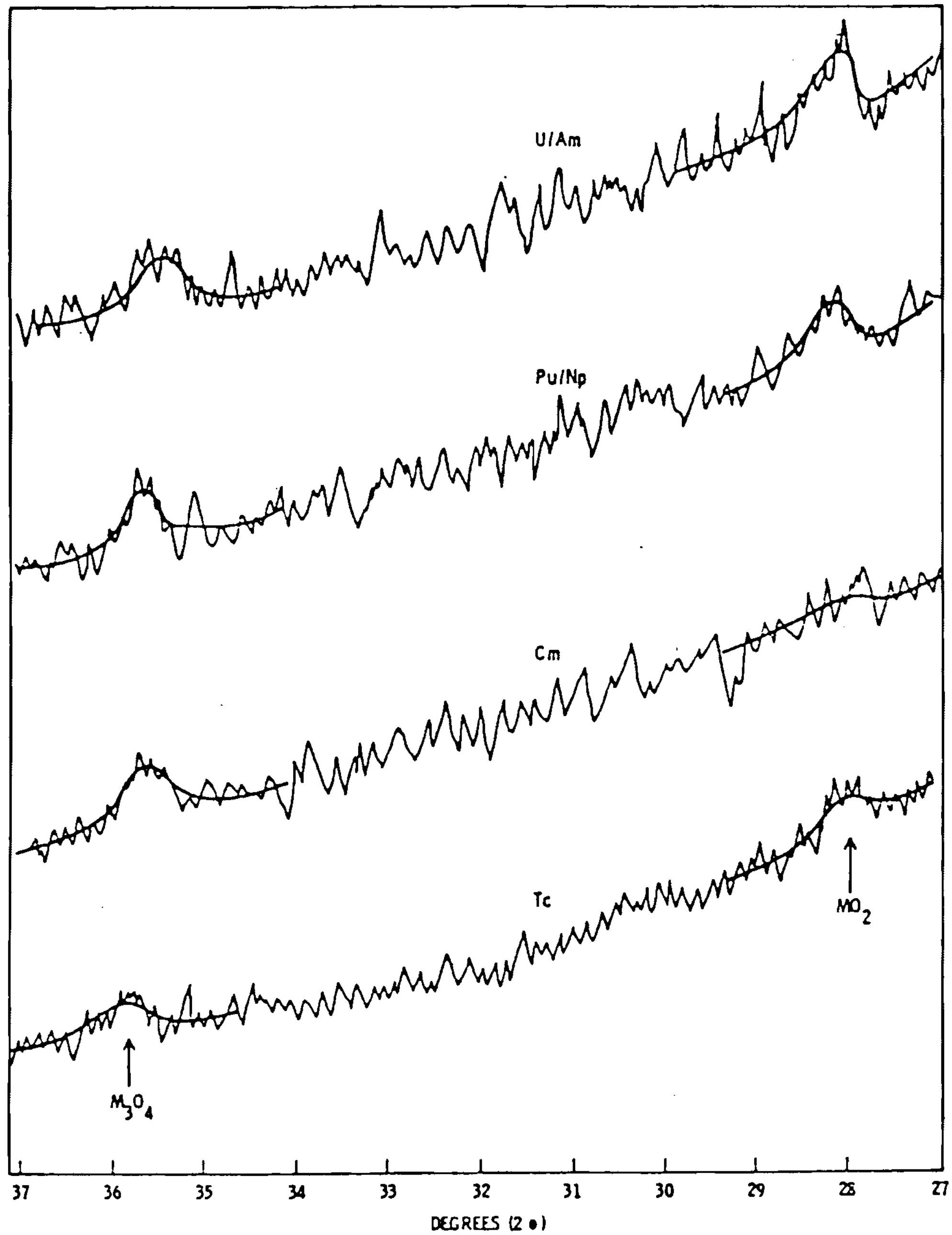


FIGURE 1. X-ray Diffraction Patterns for Doped Glasses

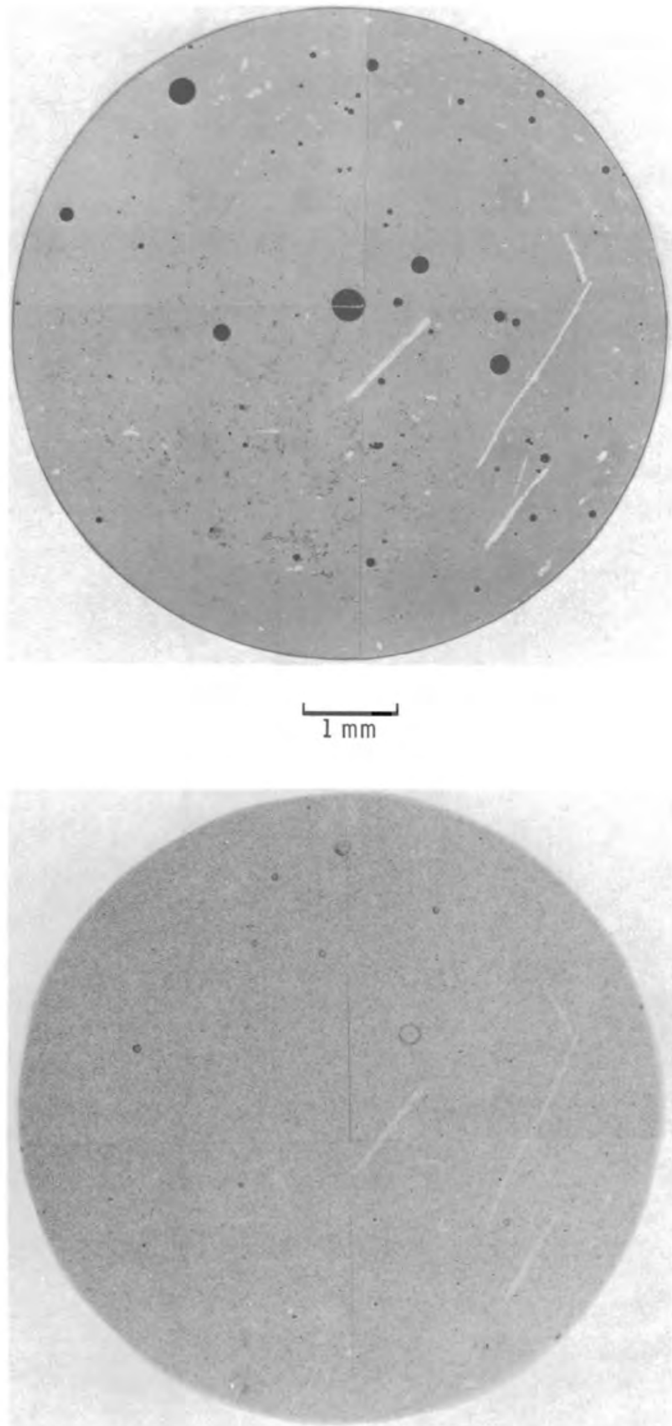
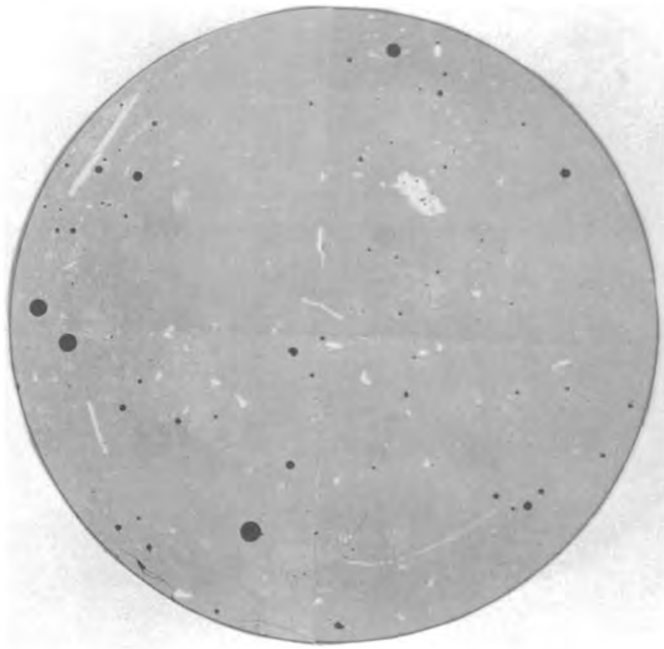


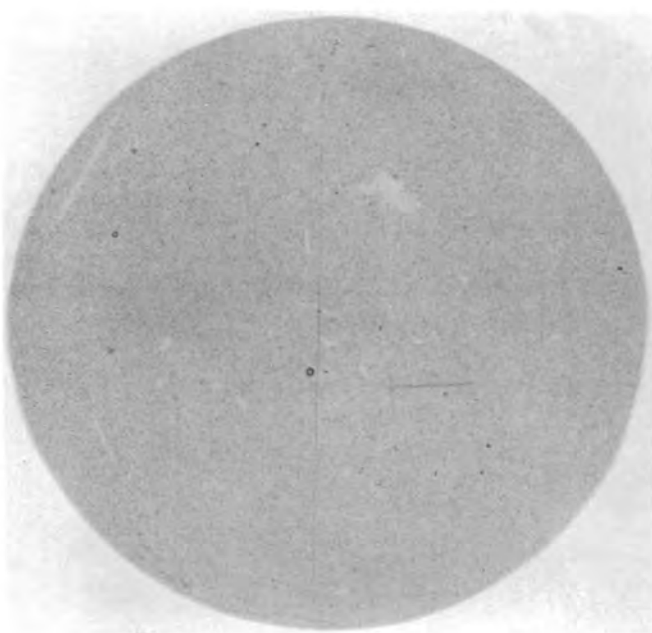
FIGURE 2. Optical Micrograph and Autoradiograph for ^{244}Cm Doped Glass

Pu, Np



OPTICAL

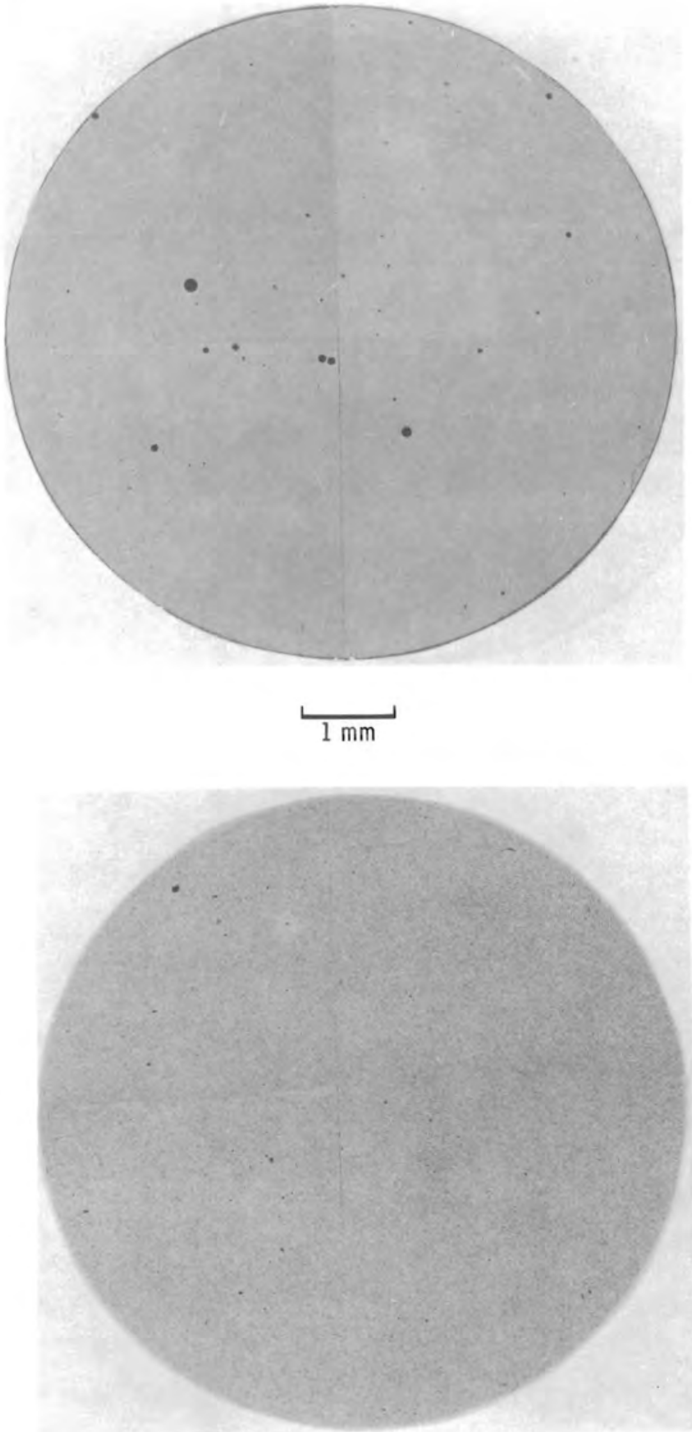
1 mm



RADIOGRAPH

FIGURE 3. Optical Micrograph and Autoradiograph for ^{239}Pu + ^{237}Np Doped Glass

Am, U



OPTICAL

RADIOGRAPH

FIGURE 4. Optical Micrograph and Autoradiograph for ^{233}U + ^{243}Am Doped Glass

Tc

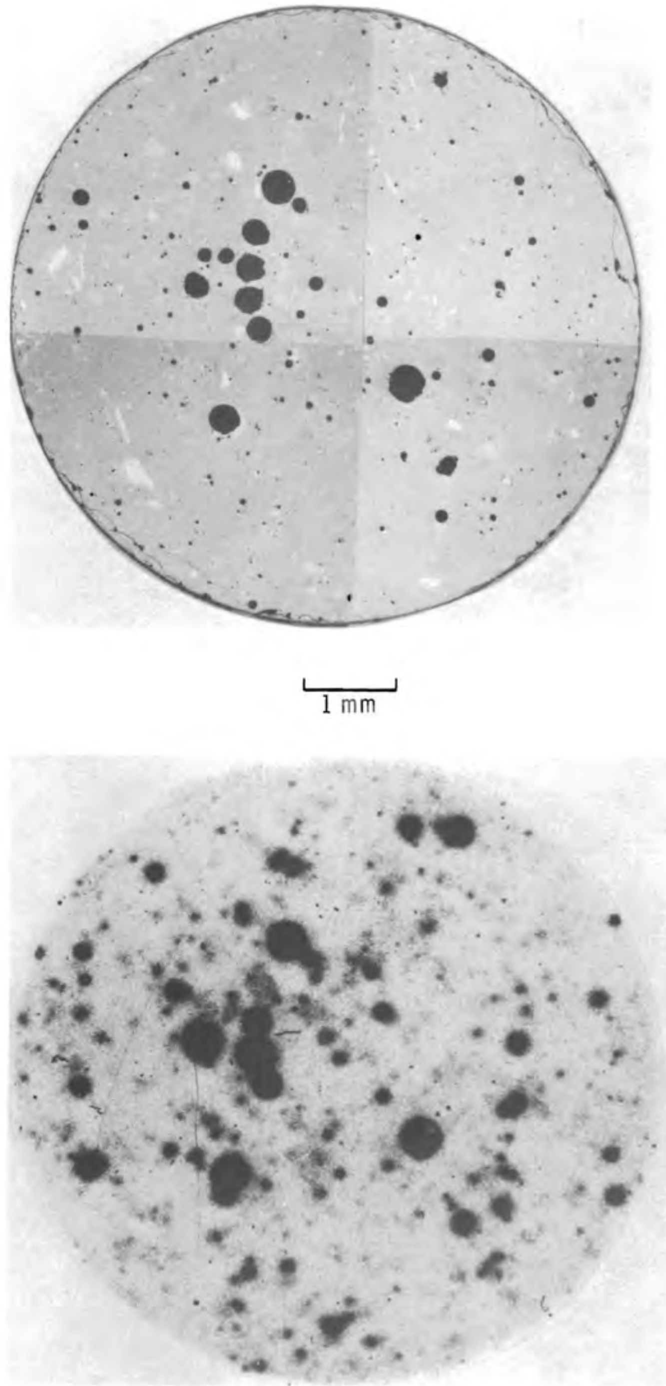


FIGURE 5. Optical Micrograph and Autoradiograph for ^{99}Tc Doped Glass

seen are anomolies of the film processing and radiography technique. Of the 20 beads examined, all were quite homogeneous in actinide distribution.

ANALYTICAL PROCEDURES FOR LEACHATE SOLUTION ANALYSIS

The leaching tests were performed in polypropylene jars with "O" rings fitted to the lids to assure a good seal. The leach container is shown in Figure 6. The temperature at which all tests were conducted was 22° ± 2°C.

Twenty beads (∿1/3 g each), which had previously been washed in ethanol and weighed, were placed in a nylon basket and suspended in the center of the test solution. Three hundred milliliters of solution were used in each test. This gave a bead surface area to solution volume ratio of 0.1 cm^{-1}.

The five solutions used are listed in Table 3. The detailed composition of WIPP "B" Brine is given in Table 4. Each combination of doped glass and test solution was run in triplicate.

The sampling schedule used, a modified version of the IAEA procedure, is shown in Table 5. On sampling days, the basket holding the beads was carefully removed and placed in a new polypropylene jar with fresh solution to continue the testing. The leachate solution was swirled, and two solution samples withdrawn. One sample was placed in a polyethylene bottle and submitted for measurement of pH, Eh, and silicon. The results of these analyses were compared to similar measurements taken on the solutions used prior to leaching. There were no significant total solution changes in pH or conductivity (measured for deionized H_2O) during the leaching period. The Eh analyses were done using platinum combination electrodes. Although there is some question as to the use and validity of Eh measurements on unpoised solutions such as these, no changes were observed during the leaching tests.

The other sample, which was analyzed for the dopant isotope(s) concentration, was put in a glass vial. To prevent the adherence of the dopant isotope(s) to the walls of the sample vial, this sample was acidified to a pH of ∿1 with concentrated nitric acid.

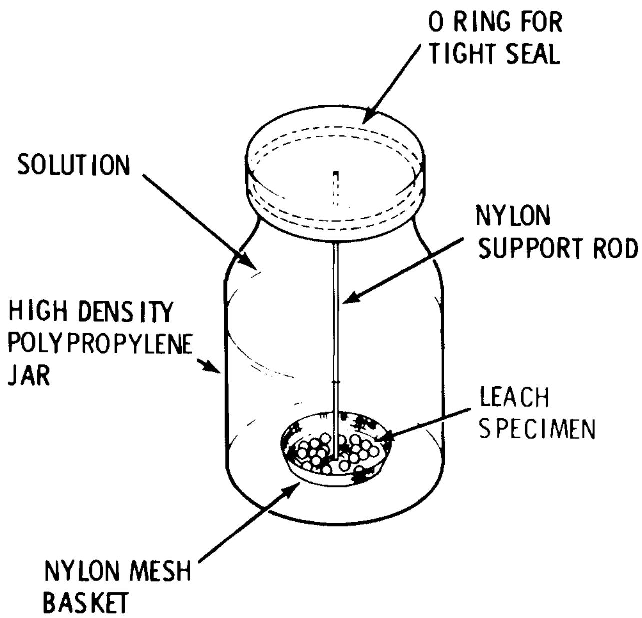


FIGURE 6. IAEA Leach Test Container

TABLE 3. Solutions Used for Leaching Study

- Salt Brine, WIPP "B" Natural Salt Brine
- Synthetic High Ionic Strength Ground Water (1.66 g/l $CaCl_2$)
- Synthetic High Bicarbonate Ground Water (2.52 g/l $NaHCO_3$)
- Synthetic High Ionic Strength Ground Water (1.76 g/l NaCl)
- Deionized Water

TABLE 4. Chemical and Ionic Composition of WIPP "B" Salt Brine

Compound	Concentration (g/ℓ)	Ion	Concentration (mole/ℓ)
NaCl	287.0	Na^+	5.0
Na_2SO_4	6.20	K^+	0.00038
$Na_2B_4O_7.10H_2O$	0.0160	Rb^+	0.000012
$NaHCO_3$	0.0140	Cs^+	0.000008
NaBr	0.5200	Mg^{++}	0.00041
KCl	0.0290	Ca^{++}	0.022
KI	0.0130	Sr^{++}	0.00017
$MgCl_2$	0.0400	Fe^{+++}	0.000036
$CaCl_2.2H_2O$	3.30	$C\ell^-$	4.94
$FeCl_3$	0.0060	Br^-	0.0050
$SrCl_2.2H_2O$	0.0330	I^-	0.000079
Rb_2SO_4	0.0016	HCO_3^-	0.00016
CsCl	0.0013	SO_4^{--}	0.036
Total Dissolved	297.2 g/ℓ	B (BO_3^{---})	0.00017
pH (adjusted)	6.5		

TABLE 5. Modified IAEA Leach Schedule

CUMULATIVE TIME	LEACH SOLUTION CHANGED	SERIES NUMBER	LEACH SOLUTIONS ANALYZED
DAYS	1	1	1
	2	2	2
	3	3	3
	4	4	4
WEEKS	2	5	5
	3	6	6
	4	7	7
	5	8	8
	6	9	
	7	10	9
	8	11	
	9	12	10
MONTHS	3	13	11
	4	14	
	5	15	12
	6	16	
	7	17	13
	8	18	
	9	19	14
	10	20	
	11	21	15
	12	22	

The used test jar was filled with 300 ml of 5M HNO_3 + 0.05 M HF solution to remove any of the nuclide of interest which had plated out on the walls. After a time equal to the original test period, the solution was sampled and submitted for analysis (plate-out sample).

The combined results from the analysis of the leachate and plate-out samples were used to calculate the leach rate.

The effectiveness of the acidification in preventing the adherence of the test nuclide(s) to the walls of the sample vial was investigated by rinsing the vial with a strong acid solution and analyzing the solution, or by filling the empty acid rinsed vial with liquid scintilator solution and counting it in a liquid scintilation counter. The treatment proved effective, as the amounts found on the walls after acidification was generally less than 1% of the activity in the sample. In the case of plutonium, the amount was below the detection limit for the procedure. (In the worst case the lower detection limit was 8% of the original activity of the sample.)

The significance of plate-out varied greatly among the different isotopes, ranging from insignificant to dominant. With few exceptions, the plate-out of an isotope was similar in all solutions.

In the plutonium-neptunium doped glass leach tests, the plutonium concentration in the plate-out solutions was mostly below the detection limit of the analysis. In those cases where the concentration was measurable, it was generally less than 10% of the leachate. The neptunium plate-out was much greater than plutonium. It ranged from less than 10% to 80% of leachate concentration.

In the curium tests, plate-out was significant in all solutions except WIPP "B" Brine. The curium concentration of the plate-out samples in some cases exceeded that of the leachate. In brine it was less than 1%.

With technetium, plate-out ranged from 0.1% in the beginning to near 1% at 500 days. In a few cases for deionized water, plate-out was found to be as large as 50% of the leachate concentration.

Plate-out of uranium was less than 1% of the leachate in brine and bicarbonate solution. It ranged from 10% to 50% of the leachate concentration in the other solutions. Americium plate-out ranged from 1% (detection limit) to 35% of the leachate. It was lowest in brine and bicarbonate.

Solvent extraction and alpha counting were used in measuring the amount of plutonium and neptunium in solution. Each isotope was individually extracted into a TTA-xylene solution (Moore and Hudgens 1957; Moore 1957). An aliquot of each organic extract was plated on a stainless steel disc and counted in a gross alpha counter.

Solutions containing ^{99}Tc or ^{244}Cm were analyzed by liquid scintillation (LS) counting. The acidified sample was added directly to the scintillator (PCS) (available from Amersham Searle) and counted. It was found that with the addition of 2 ml of water to 15 ml of PCS, as much as 1 ml of a brine sample could be counted without seriously reducing the counting efficiency.

Uranium and americium leach solutions are analyzed by ion exchange separation (Roberts and Bauer 1959), and liquid scintillation counting of the two fractions. Uranium was loaded onto Dowex-1, X-4 chloride form. The effluent containing the americium was collected in a liquid scintillation vial. The uranium was eluted from the column, and collected in a LS vial. Both solutions are evaporated to dryness, and the residue was dissolved in a small quantity of dilute acid. Liquid scintillator (PCS) is added to the vial and the sample counted.

Silicon was determined colorimetrically, using the ammonium molybdate method in which the absorbance of the reduced silicomolybdate complex measured in an absorption spectrometer is proportioned to the silica concentration of the sample solution (American Public Health Association 1971). Tests were conducted which demonstrated that all of the silicon present in the leach solutions was complexed by the ammonium molybdate.

RESULTS OF DOPED WASTE GLASS LEACHING

The equations used to calculate the isotopic leach rates and cummulative fractions leached are as follows:

$$R_i = \frac{a_o}{A_o S t}$$

$$R = \frac{\Sigma a_o}{A_o W}$$

where

R_i = incremental leach rate in g/cm^2-day
a_o = activity of isotope in leachate, sec^{-1}
A_o = specific activity of isotope in sample sec^{-1} - $gram^{-1}$
S = geometric surface area of sample, cm^2
t = leaching time, days
R = cummulative fraction leached
W = sample weight, grams.

Since the leach tests were all done in triplicate, the data were analyzed to determine the spread among triplicate results. The maximum deviation was seen to be a factor of two to three. This range was found to be the same for all solutions.

All of the leaching data presented in this report are summarized in tabular form in Appendix A.

Figures 7 through 11 show leach rates of the actinides studied as a function of leach solution. In general it can be seen that leach rates decrease by roughly an order of magnitude within the first 10 to 20 days of solution contact. This observation is also seen in virtually all past studies on waste glass including fully radioactive glass experiments (Mendel et al. 1977; Ross et al. 1978; Bradley 1978). The glass bead approach used in the present study rules out the possibility of a contribution by surface powder or imperfections from cut surfaces, which may affect some previous studies. It can be noted

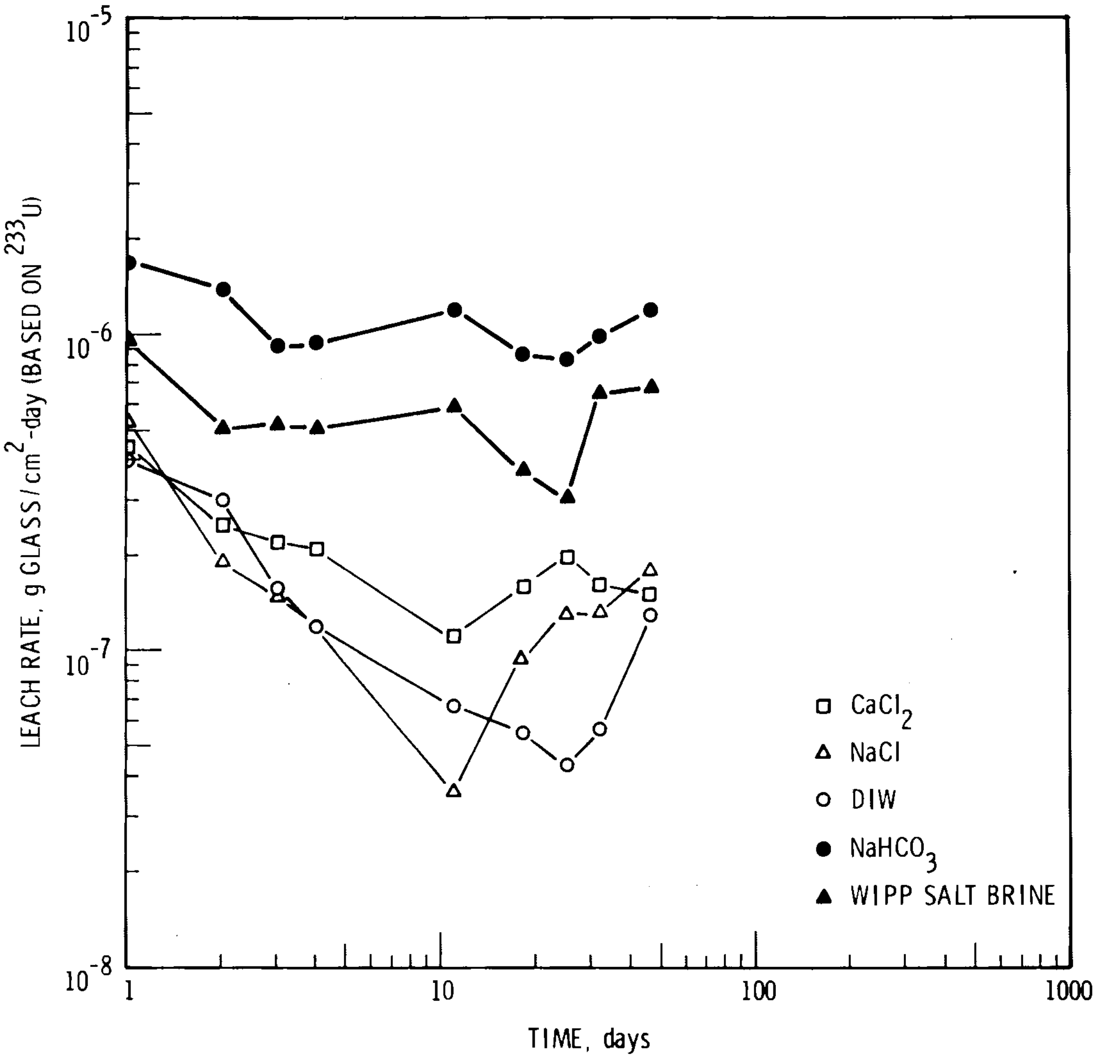


FIGURE 7. Leach Rate of ^{233}U from Simulated Waste Glass 76-68, IAEA Leach Test at 22°C

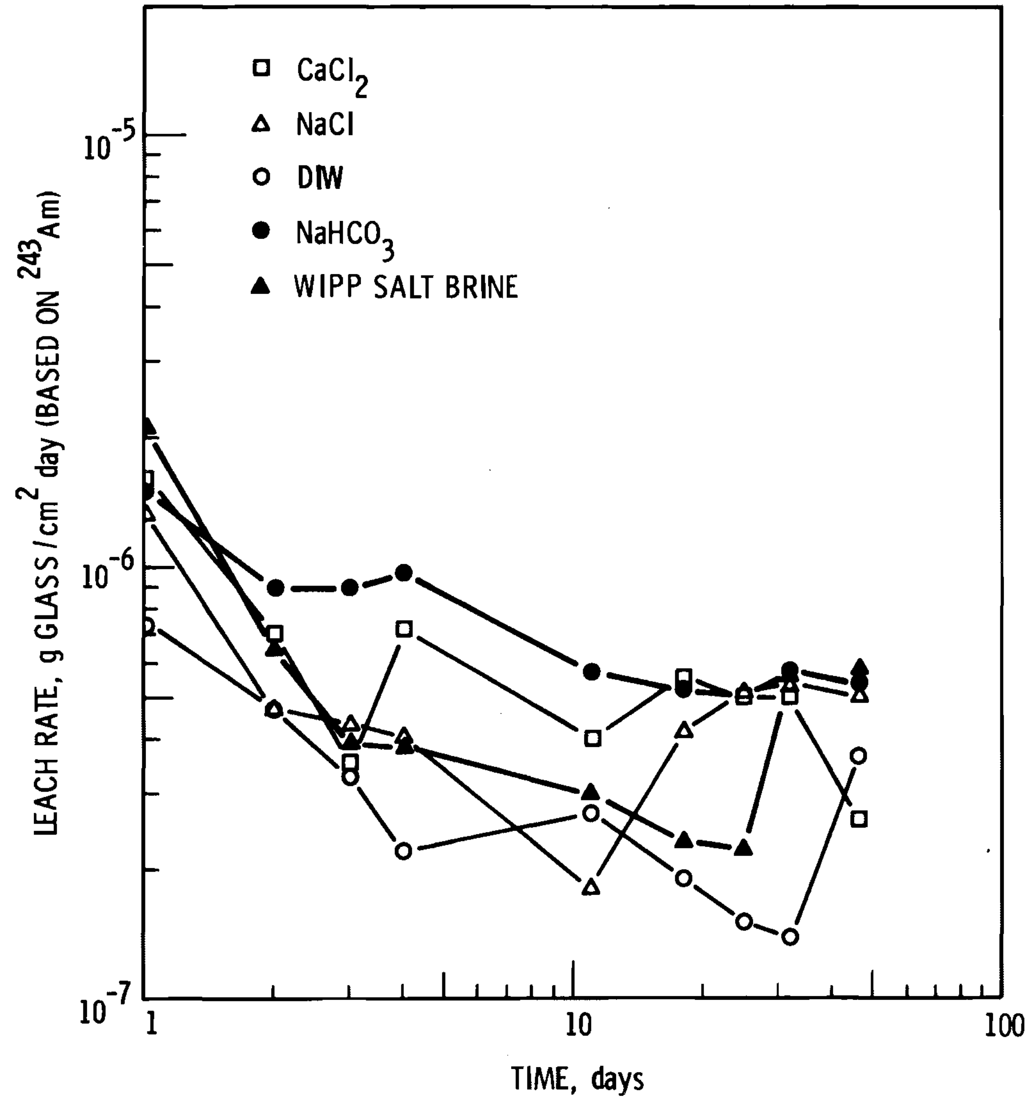


FIGURE 8. Leach Rate of ^{243}Am from Simulated Waste Glass 76-68, IAEA Leach Test at 22°C

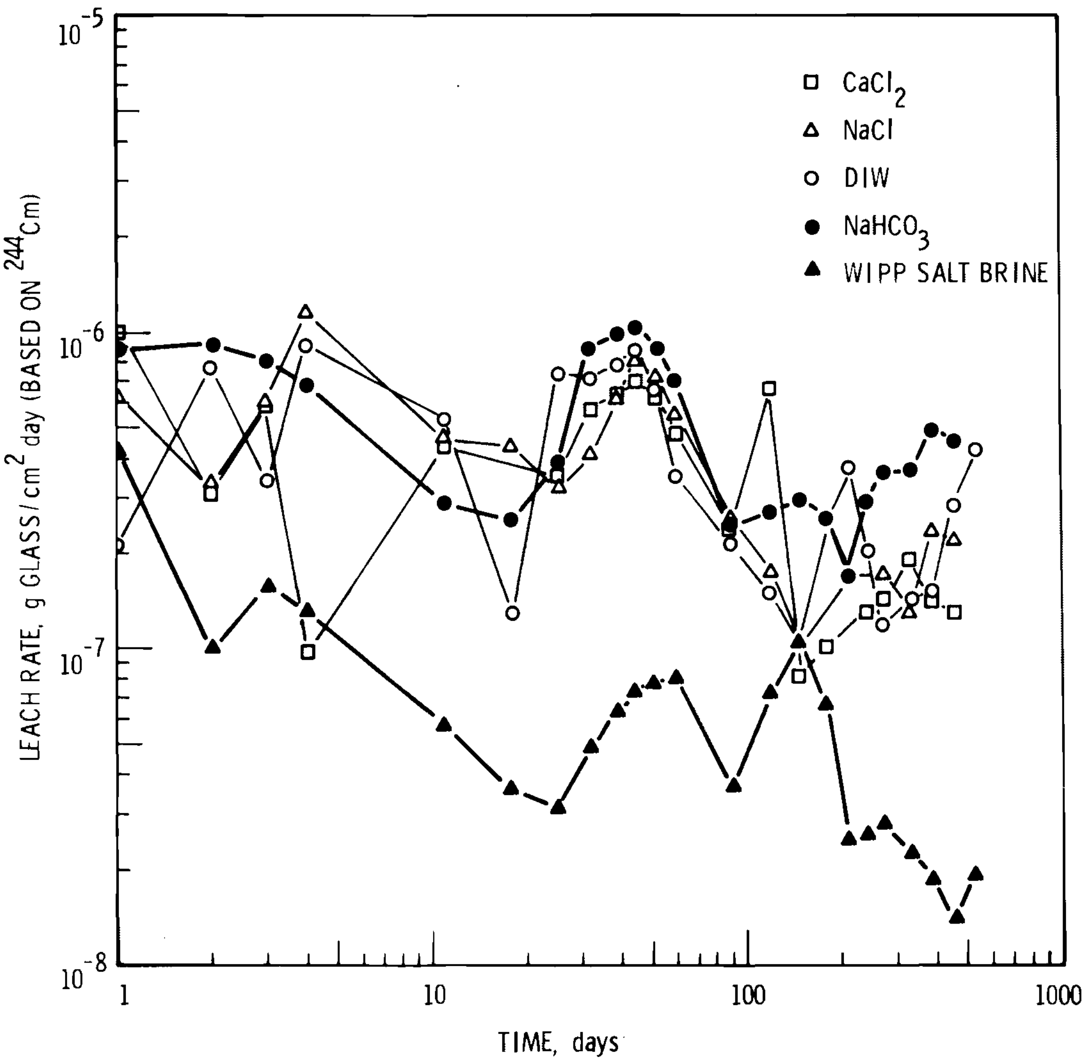


FIGURE 9. Leach Rate of ^{244}Cm from Simulated Waste Glass 76-68, IAEA Leach Test at 22°C

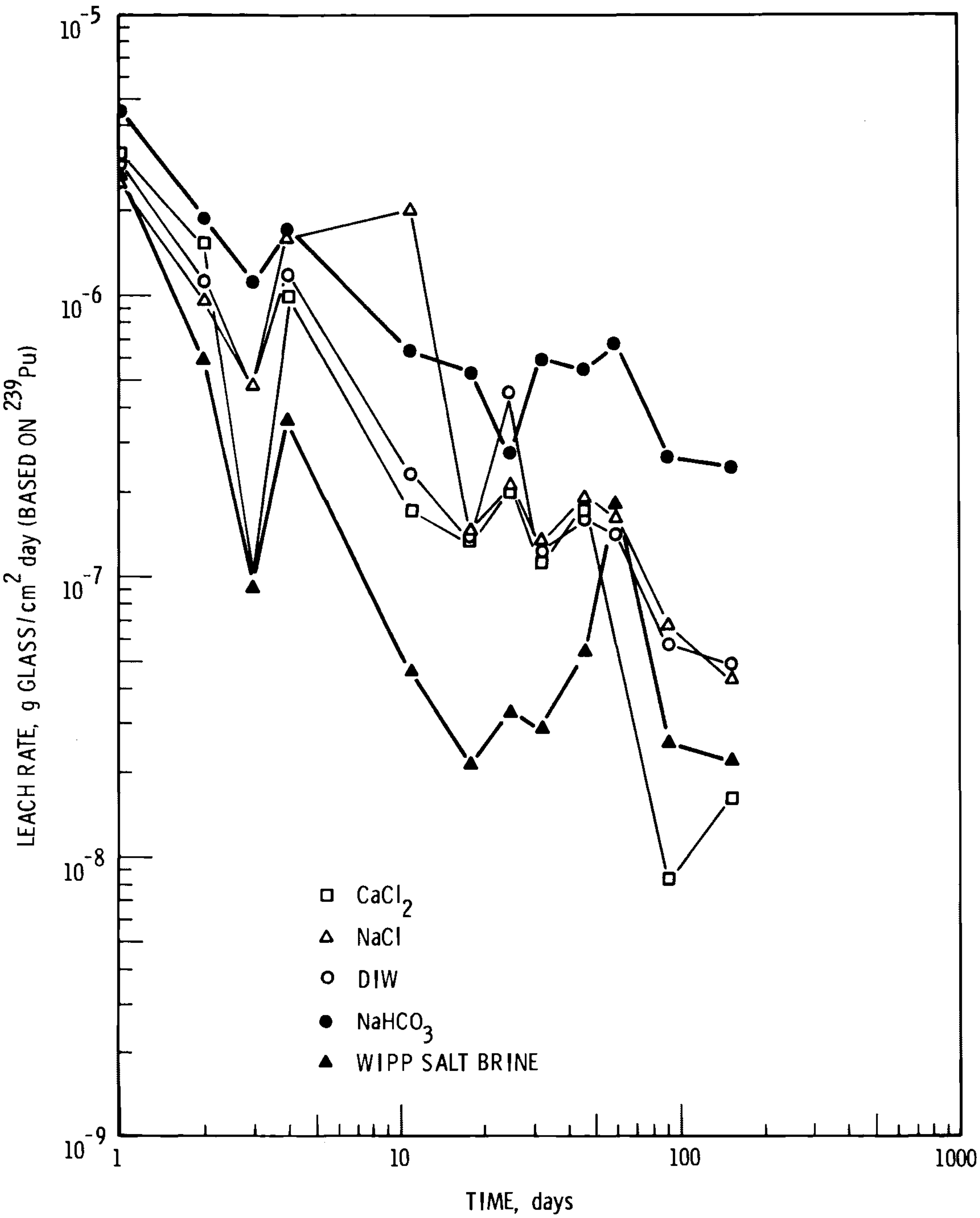


FIGURE 10. Leach Rate of ^{239}Pu from Simulated Waste Glass 76-68, IAEA Leach Test at 22°C

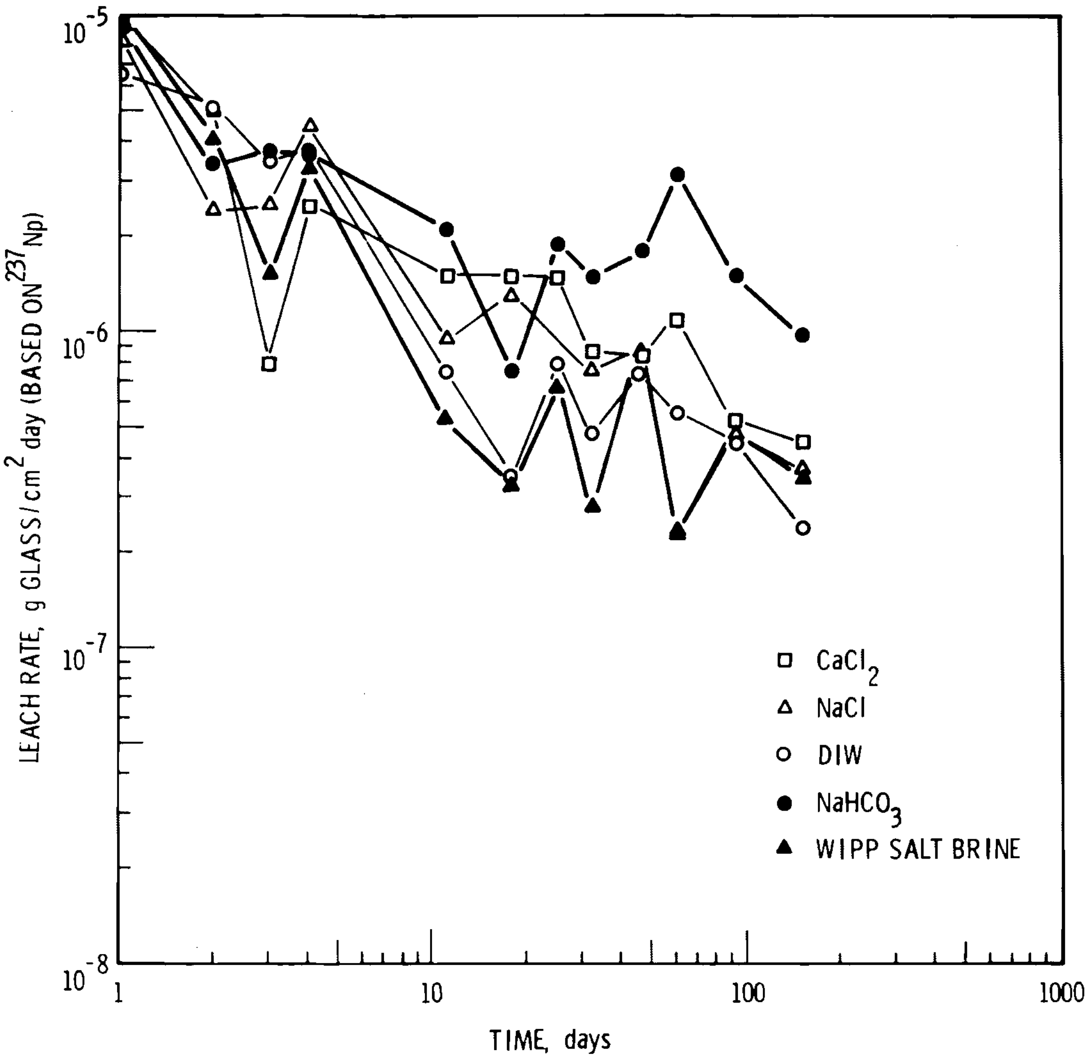


FIGURE 11. Leach Rate of ^{237}Np from Simulated Waste Glass 76-68, IAEA Leach Test at 22°C

that the leach rates for plutonium are decreasing at a faster rate than the other actinides. At this time the reason for this is unknown.

Figure 12 shows the leach rates of ^{99}Tc in WIPP"B" brine and deionized water only, as the other solutions fell between them. The trends seen here are quite different from those just discussed. The leach rates are quite high at first but then fall steadily. After 400 days the leach rate has fallen by more than a factor of 10,000. This trend is probably related to the observed accumulation of technetium on internal pore surfaces (see Figure 5) leaving a low concentration in the glass matrix. In the quenched beads, gas bubbles do not intersect the bead surface, presumably because of surface tension constraints as the molten droplet is gravity "extruded" from the crucible. We expect however that some internal bubbles are in effect collapsed at the bead surface, leaving a soluble technetium species.

The leach rate of silica in deionized water for the four doped glasses is plotted in Figure 13. Initially, differences of about a factor of eight (average) are seen among the glasses up to 20 days. After this time differences of only a factor of three are seen out to 500 days. The reason for the intial spread is unknown. The remainder of the leaching time demonstrates the expected behavior, as the four glasses have identical compositions and element doping on the level of 0.1% would not be expected to cause difference glass leaching behavior.

Since leach solutions were not analyzed every time the solution was changed, the averages of the preceeding and following solution analyses were used to enable cumulative fraction release data to be calculated.

The cumulative release fractions observed for silicon as a function of solution type is shown in Figure 14. The initial variance among solutions is small (about 1.5). This increases with time to a factor of 3.5 at nearly 600 days. Sodium bicarbonate solution shows the highest release by a factor of two at this point. This behavior is as expected since it is known that high pH (>8) solutions accelerate the attack on the silicate structure, thus releasing more silicon into solution (Hench 1977). Silicon leach information was not plotted for WIPP "B" brine because we did not obtain a complete set of

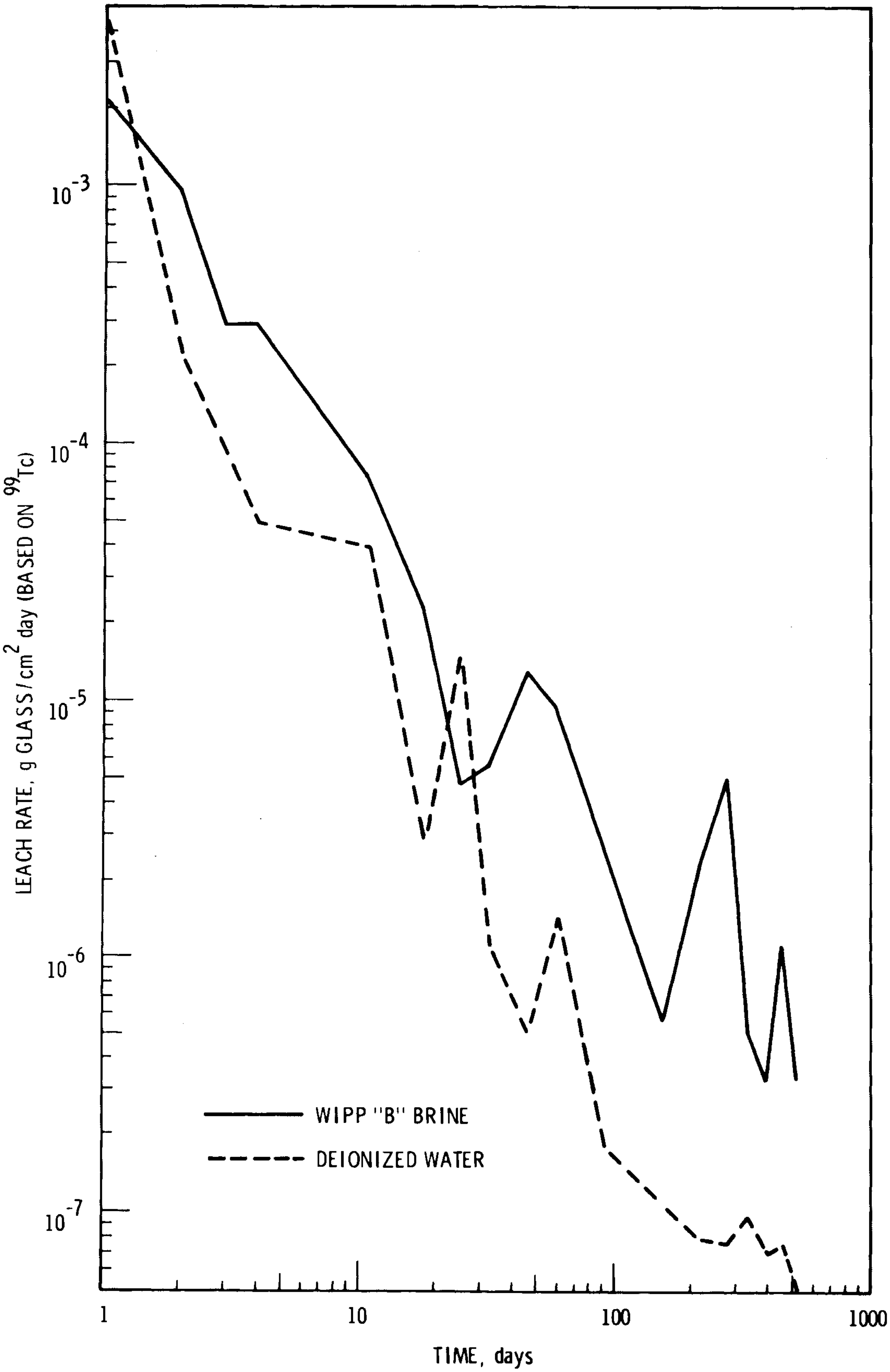


FIGURE 12. Leach Rate of ^{99}Tc from Simulated Waste Glass 76-68, IAEA Leach Test at 22°C

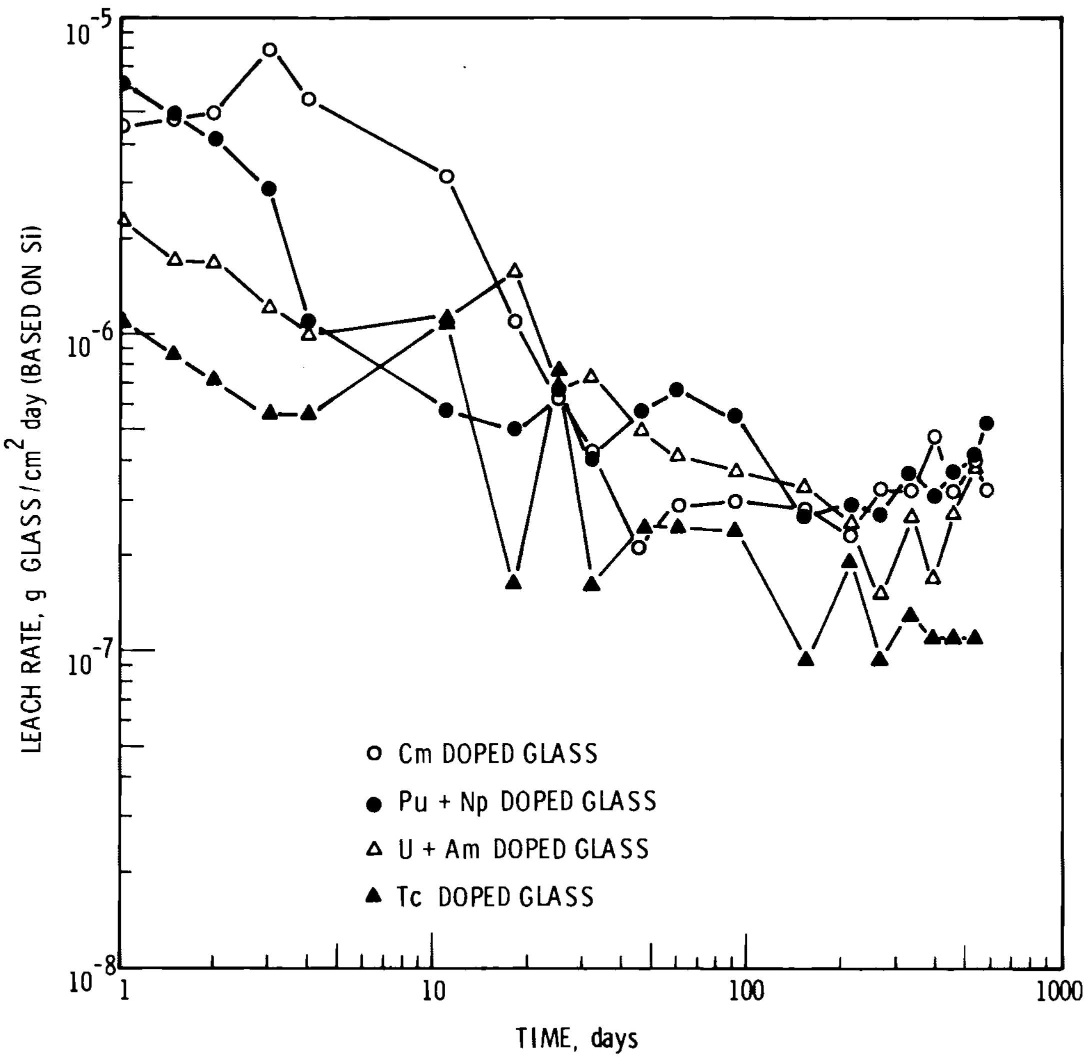


FIGURE 13. Leach Rate of Si from Simulated 76-68 Waste Glasses, IAEA Leach Test at 22°C in Deionized Water

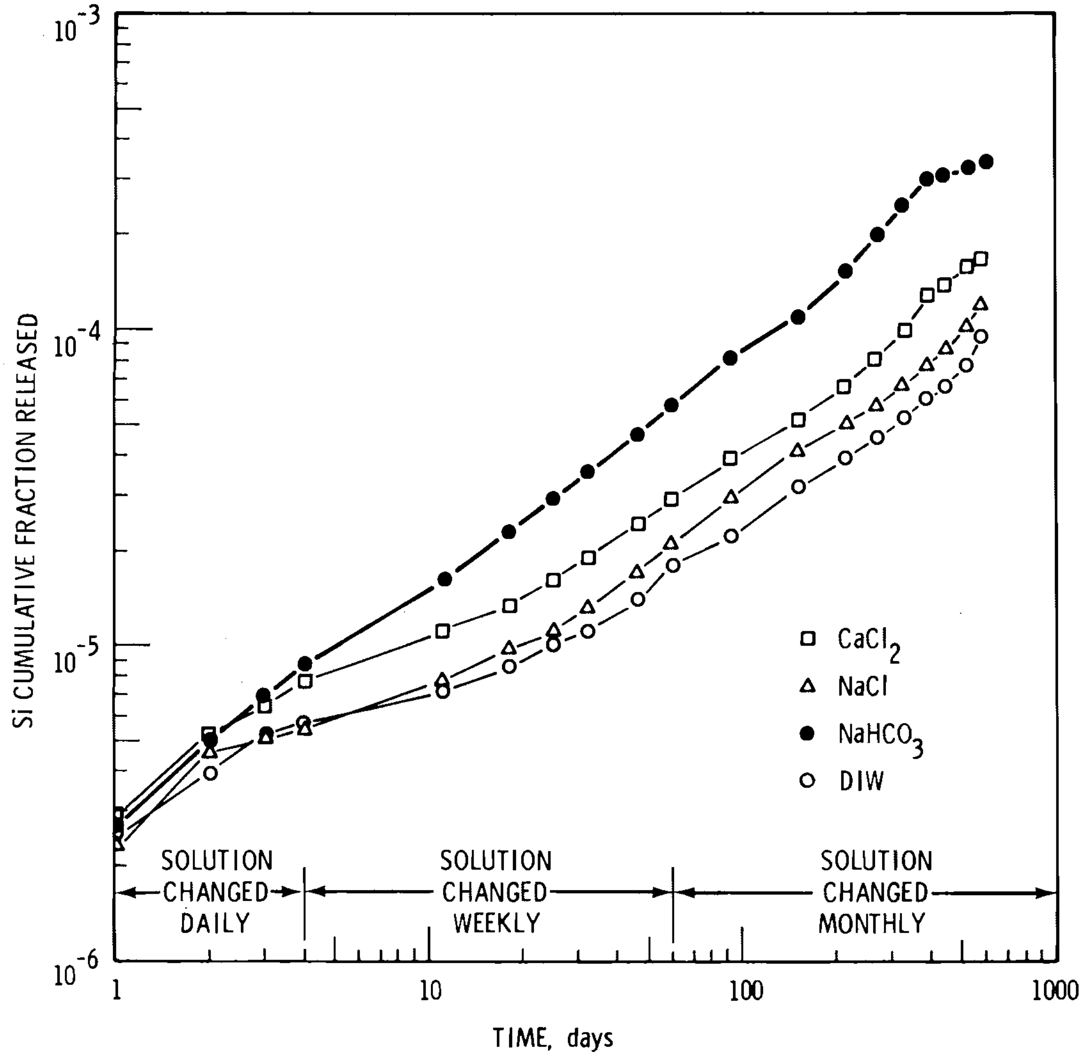


FIGURE 14. Cumulative Fraction Si Released from ^{239}Pu + ^{237}Np Doped Simulated Waste Glass 76-68, IAEA Leach Test at 22°C

silicon data. The limited data that was obtained shows that the brine follows closely the trend seen for the $CaCl_2$/NaCl solutions.

Bead samples from all of the solutions have been examined by metallography after 400 days leaching. Although a thin film causes surface discoloration the polished cross sections show no measurable reaction zone into the glass, or any buildup of products on the surface. Electron microscopy will be used to assist in further evaluation of leached beads but on fracture surfaces, so that the surface film will not be damaged by polishing.

Figures 15 through 19 depict the actinide cumulative release fractions as a function of leach solution. These are complimented by Figures 20 through 24 which show for a given leach solution, the cumulative fractions released for each actinide element. The data presented in these figures are summarized in Tables 6 and 7. Ratios of the highest to the lowest element release at the beginning and end of the reported leaching duration were calculated for both tables, to indicate trends in actinide element release with time. From the cumulative fraction released figures and resulting tables the following observations result:

- For all elements, sodium bicarbonate solution gives the highest releases. In general deionized water or WIPP "B" brine give the lowest values of release. For plutonium and neptunium, WIPP "B" brine gives the lowest release while curium, uranium and americium are lowest in deionized water.
- For all solutions except $CaCl_2$, neptunium shows the highest actinide release rate. In general curium or uranium show the lowest release. Uranium shows a high release rate in $NaHCO_3$, which may be due to the known affinity for uranium to form soluble carbonate complexes (Grandstaff 1976). Curium displays a high release rate in $CaCl_2$ and deionized water leach solutions, while americium has the highest of the actinide release rates in $CaCl_2$ solution.

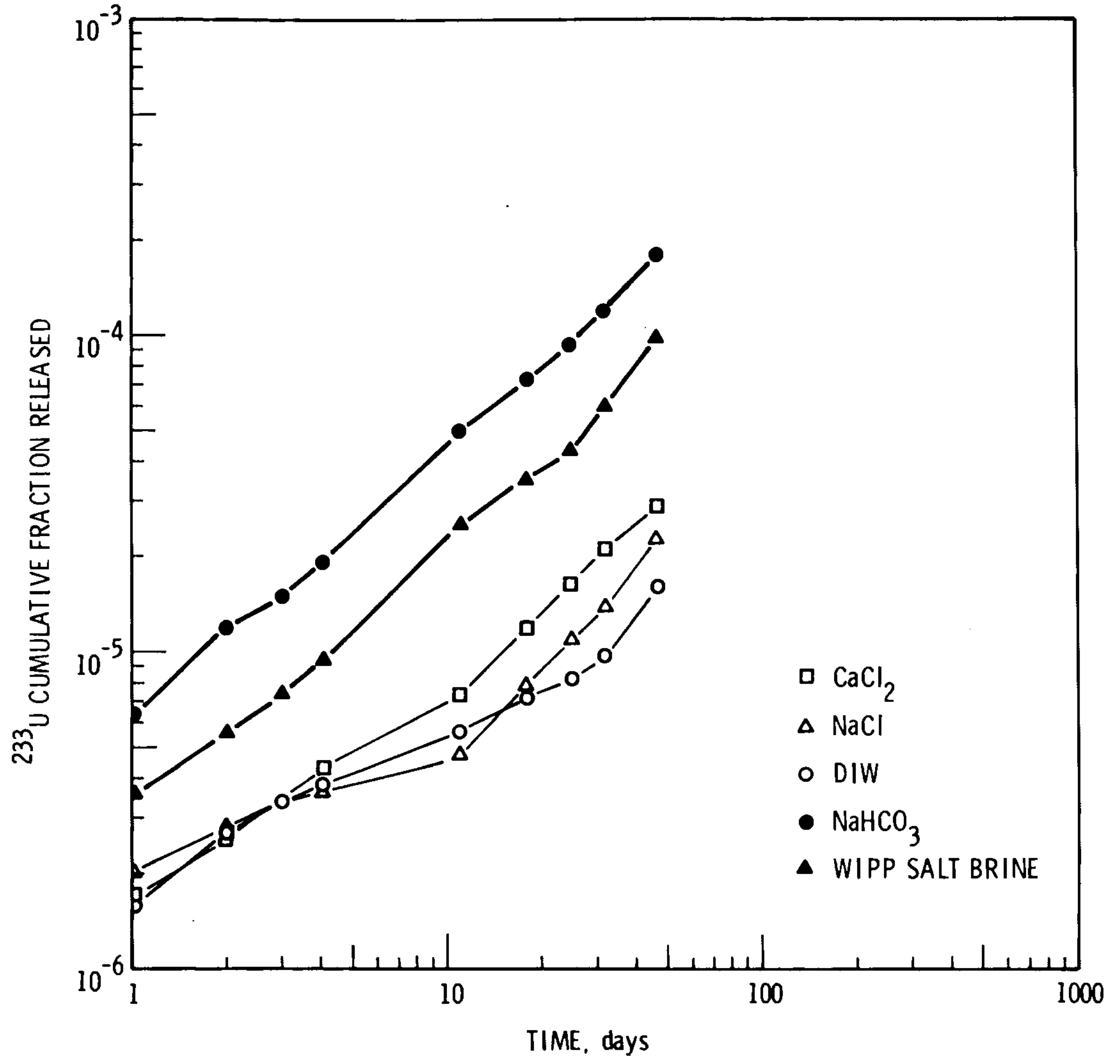


FIGURE 15. Cumulative Fraction of ^{233}U Released from Simulated Waste Glass 76-68 IAEA Leach Test at 22°C

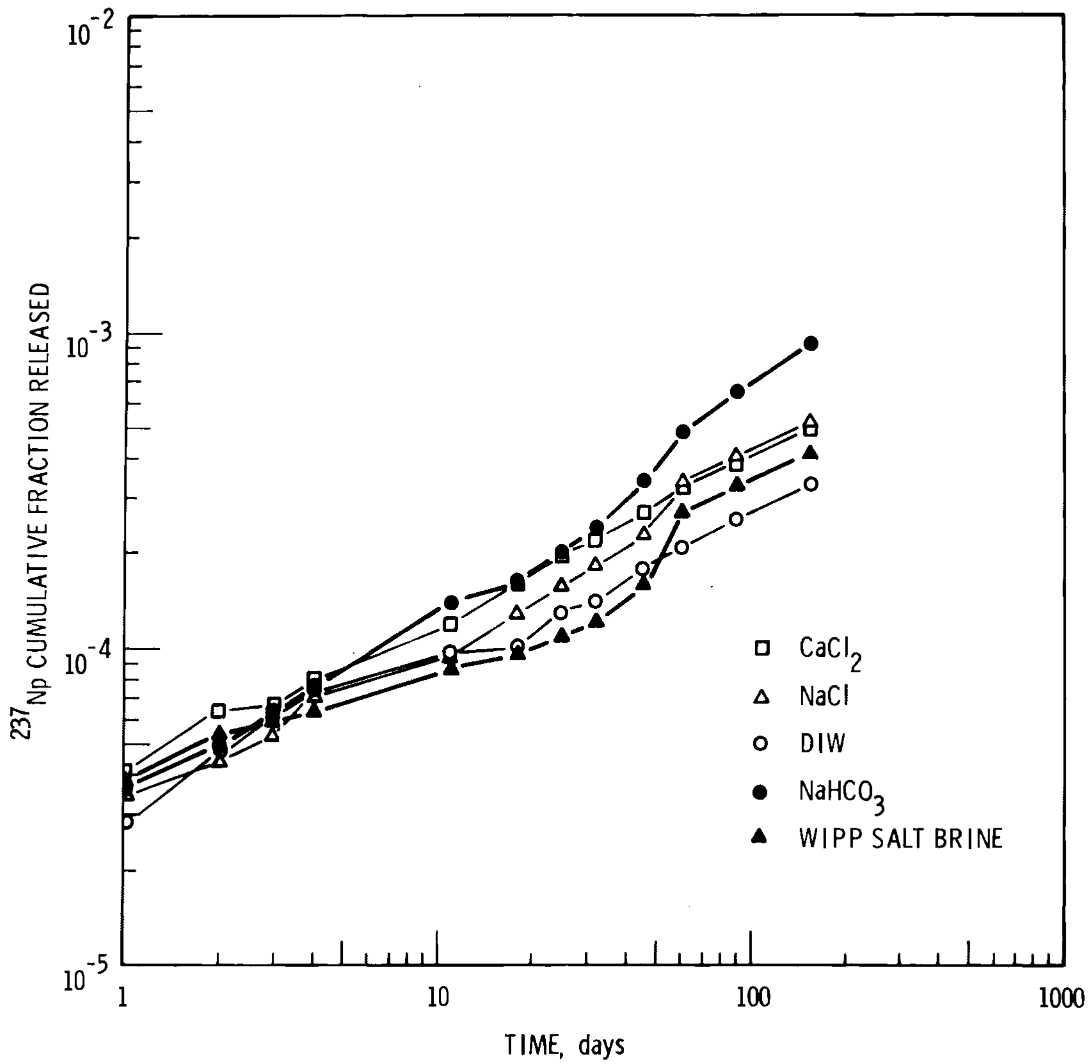


FIGURE 16. Cumulative Fraction of ^{237}Np Released from Simulated Waste Glass 76-68 IAEA Leach Test at 22°C

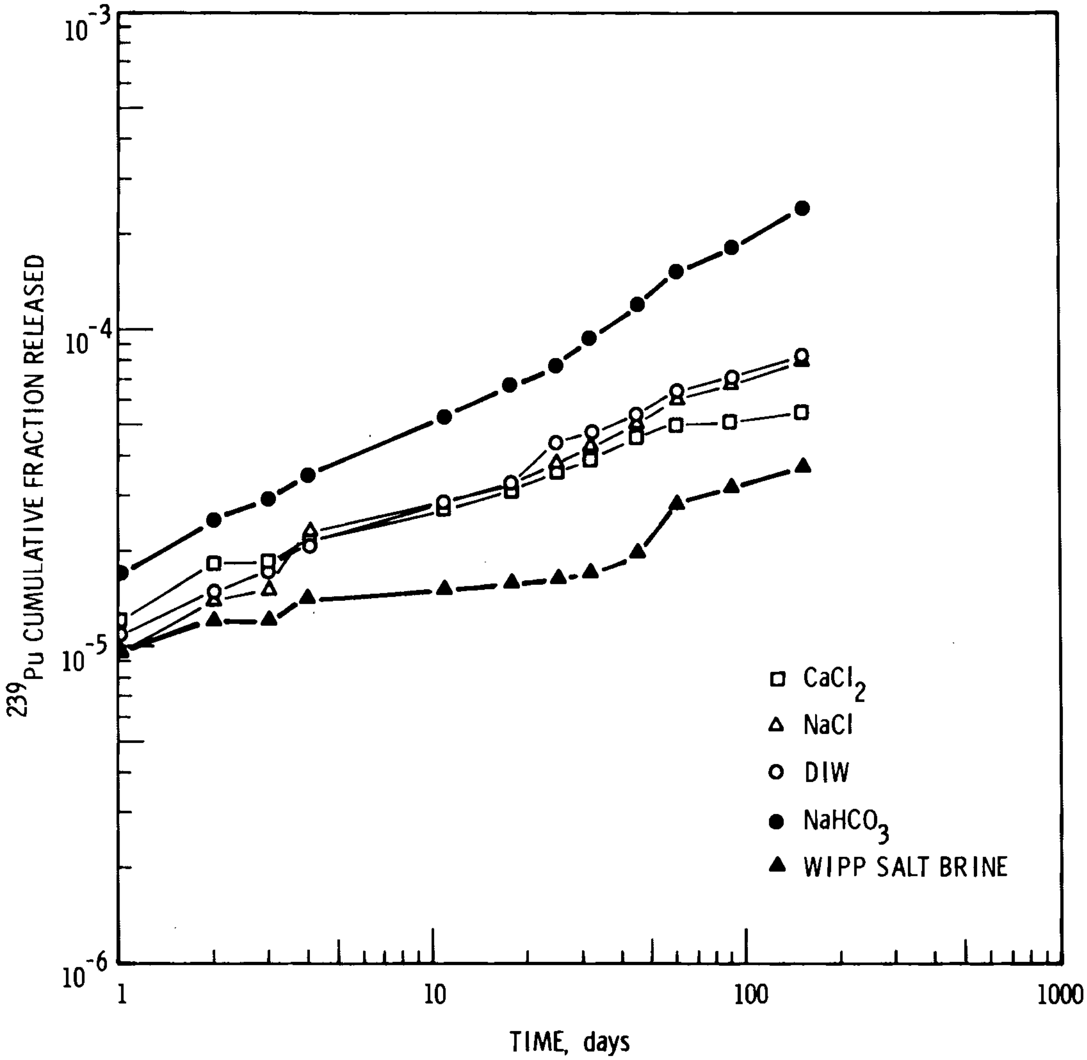


FIGURE 17. Cumulative Fraction of ^{239}Pu Released from Simulated Waste Glass 76-68 IAEA Leach Test at 22°C

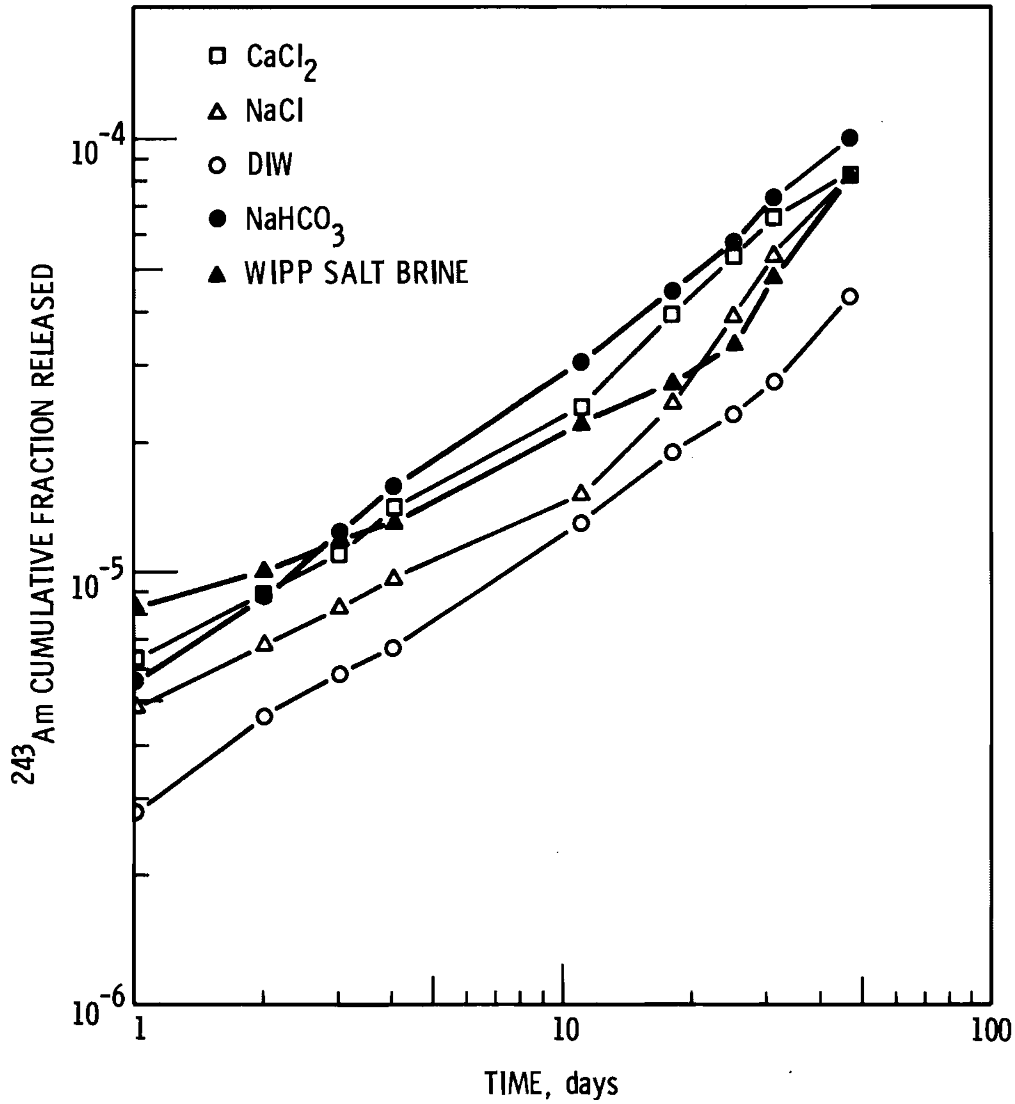


FIGURE 18. Cumulative Fraction of ^{243}Am Released from Simulated Waste Glass 76-68 IAEA Leach Test at 22°C

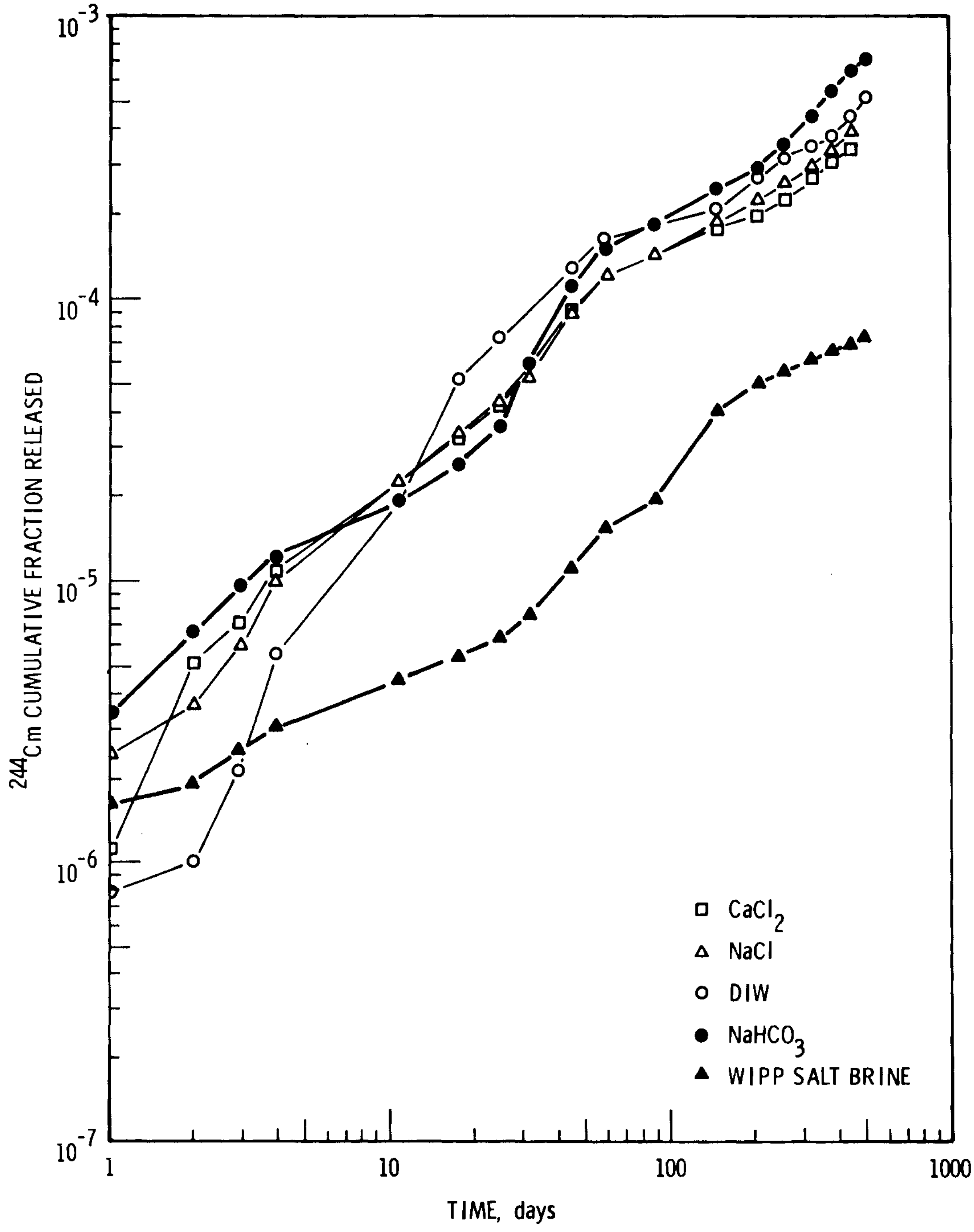


FIGURE 19. Cumulative Fraction of ^{244}Cm Released from Simulated Waste Glass 76-68 IAEA Leach Test at 22°C

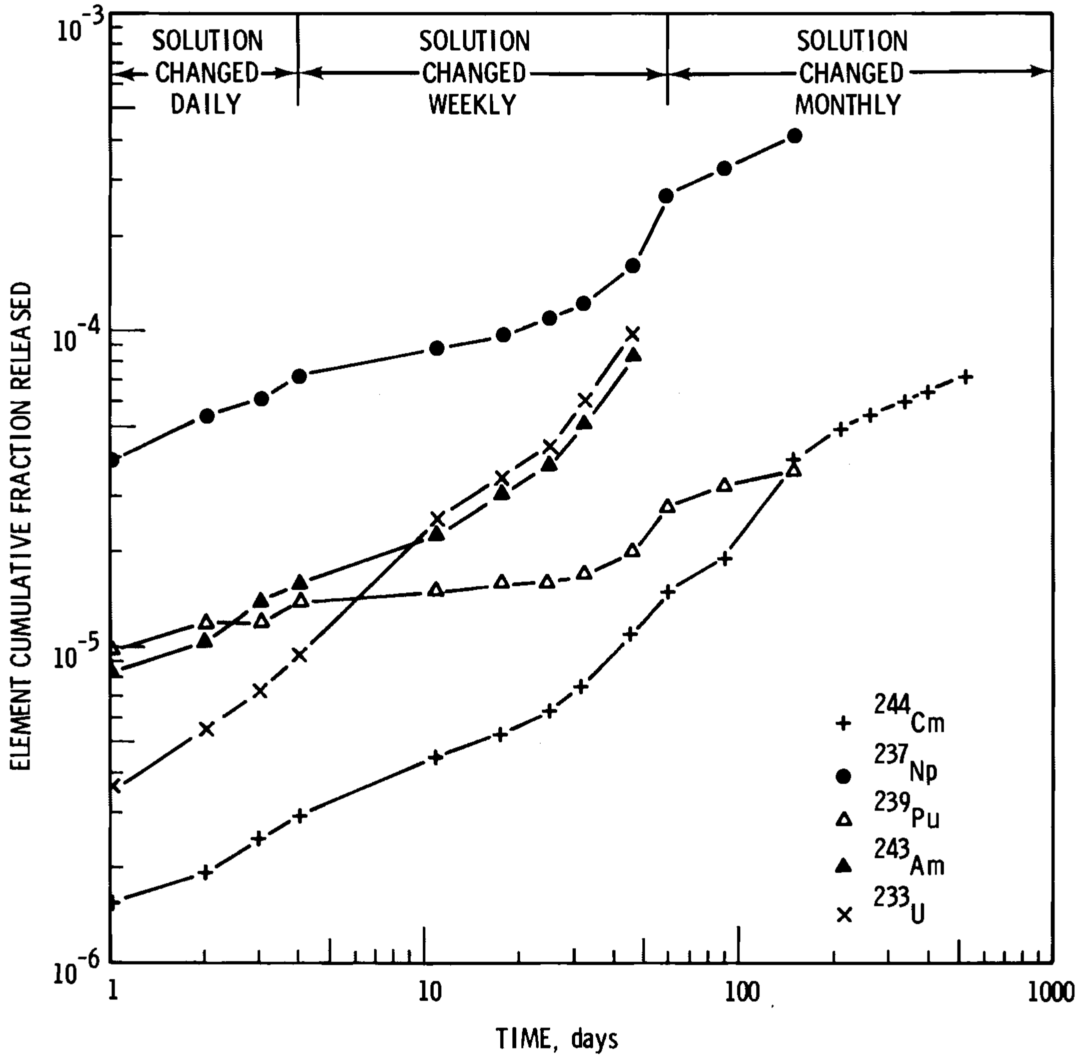


FIGURE 20. Cumulative Fraction of Actinide Isotopes Released in WIPP "B" Brine from Simulated 76-68 Waste Glass, IAEA Test at 22°C

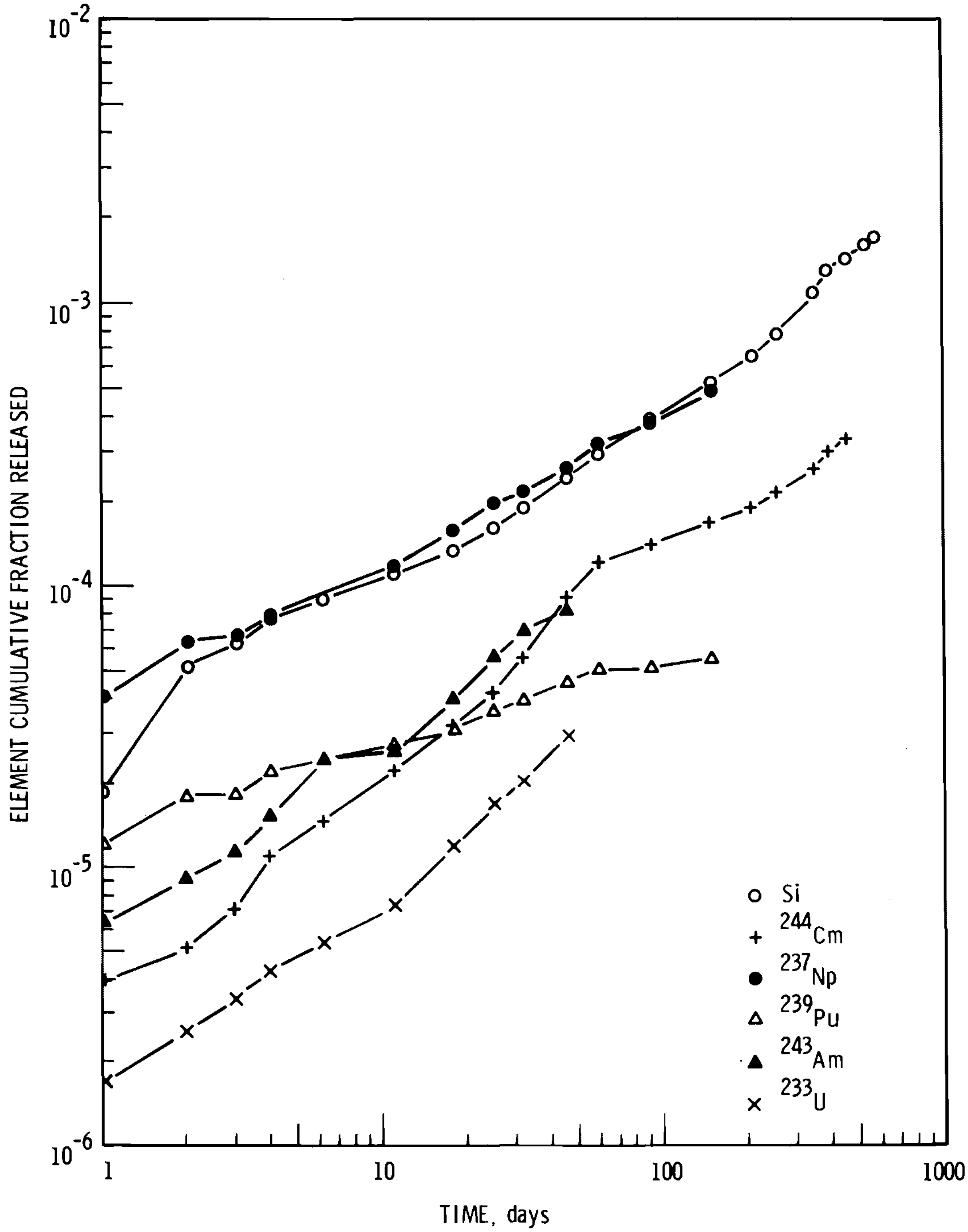


FIGURE 21. Cumulative Fraction of Actinide Isotopes Released in $CaCl_2$ Solution from Simulated 76-68 Waste Glass, IAEA Test at 22°C

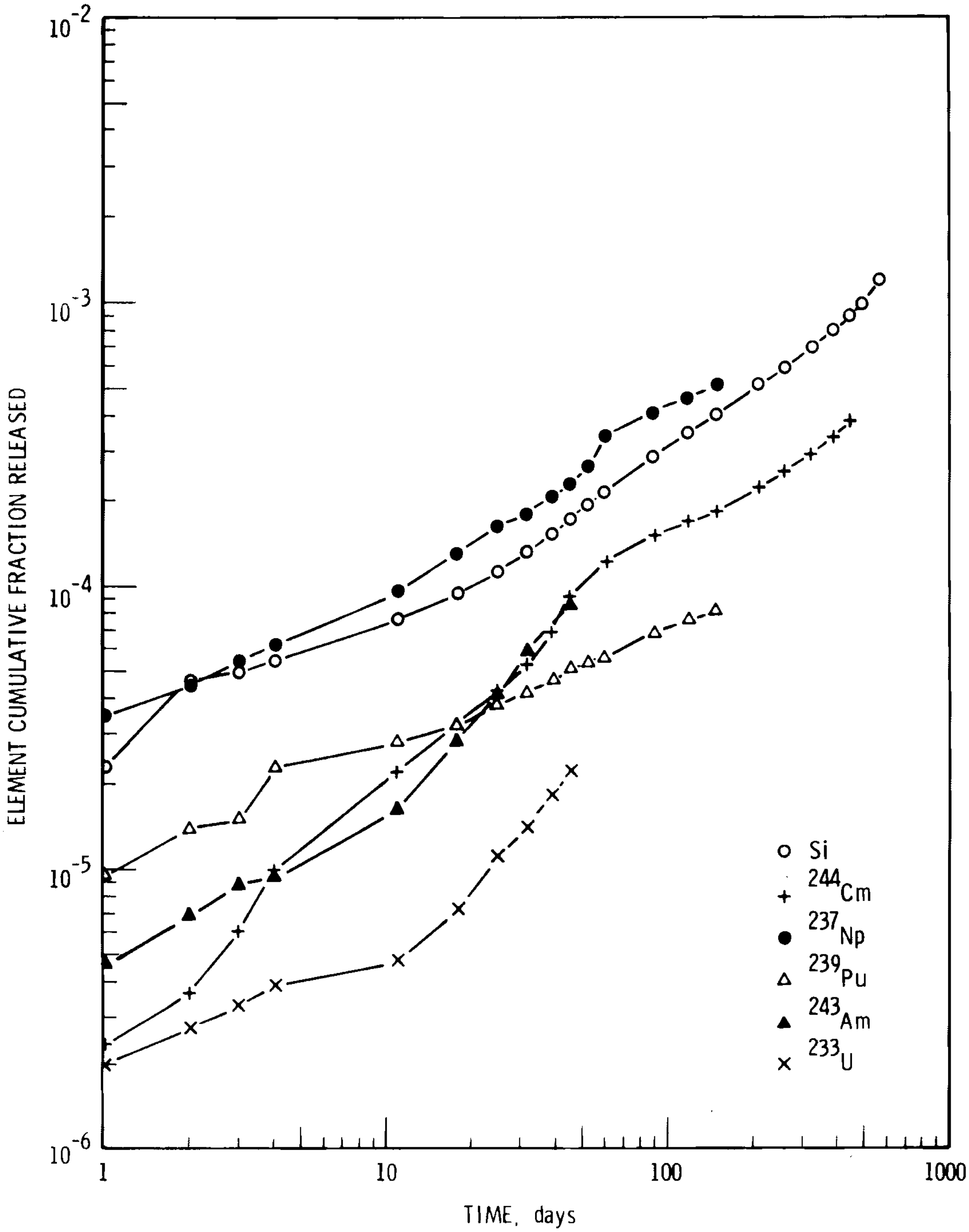


FIGURE 22. Cumulative Fraction of Actinide Isotopes Released in NaCl Solution from Simulated 76-68 Waste Glass, IAEA Test at 22°C

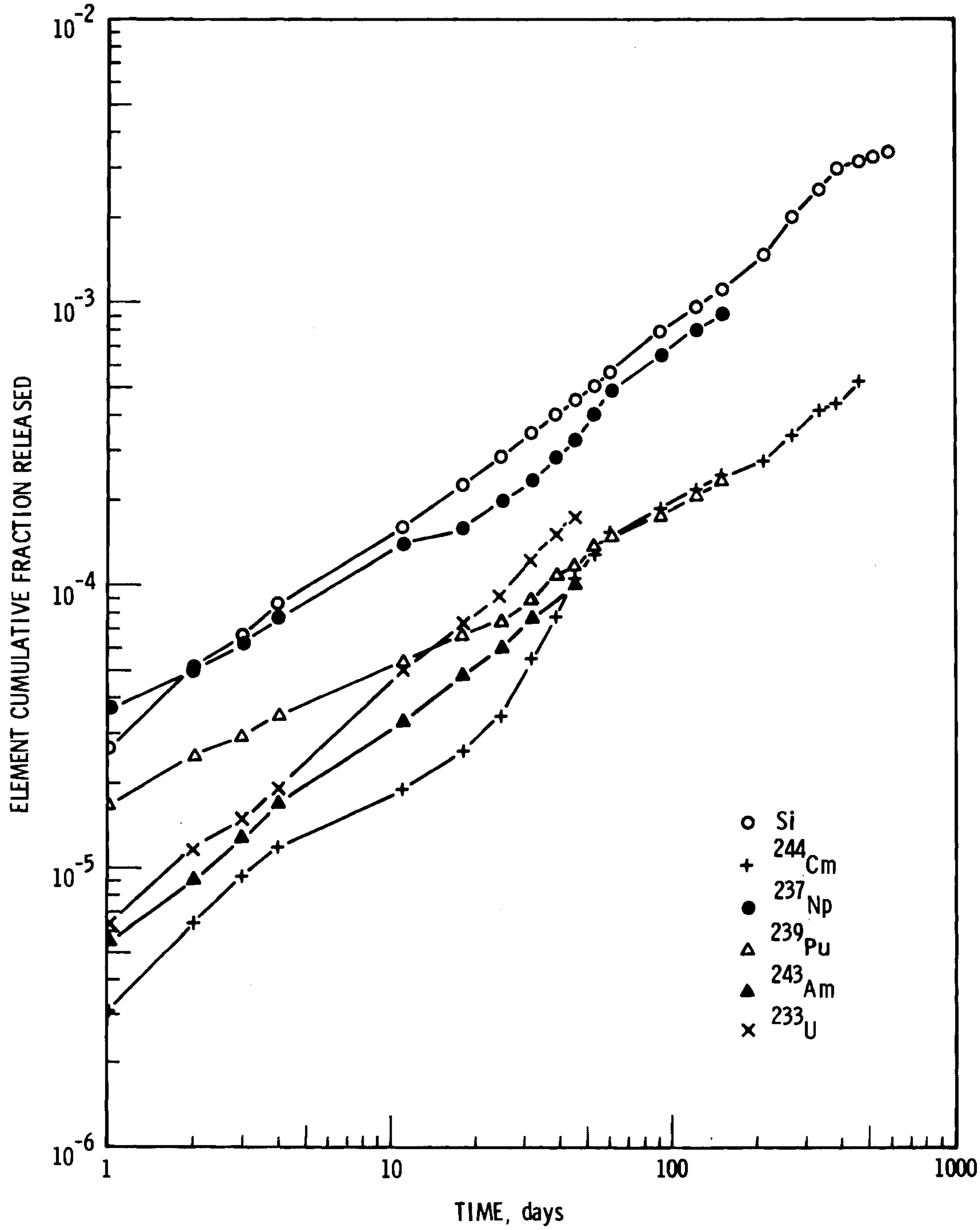


FIGURE 23. Cumulative Fraction of Actinide Isotopes Released in $NaHCO_3$ Solution from Simulated 76-68 Waste Glass, IAEA Test at 22°C

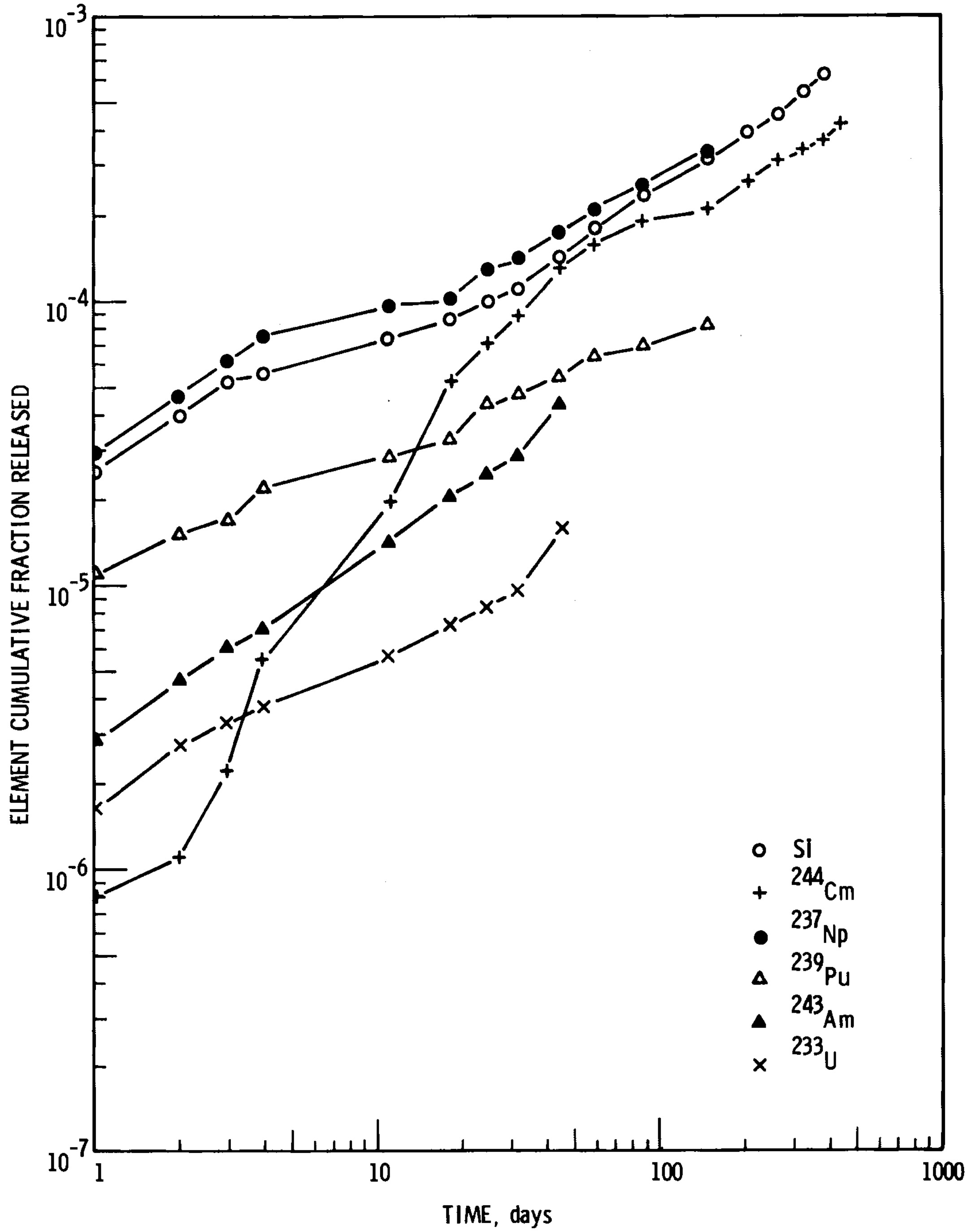


FIGURE 24. Cumulative Fraction of Actinide Isotopes Released in Deionized Water from Simulated 76-68 Waste Glass, IAEA Test at 22°C

TABLE 6. Results of Single Element Cumulative Fraction Release as a Function of Multiple Solution Type (Figures 15-19)

Element	Observed Ranking of Solutions from Highest to Lowest Element Release	Ratio of Highest to Lowest Element Release at Time - 1 Day	Ratio of Highest to Lowest Element Release at end of Leach Time
U	$NaHCO_3$, WIPP Brine, $CaCl_2$, NaCl, DIW	4.0	12.0
Am	$NaHCO_3$, BaCl, $CaCl_2$, WIPP Brine, DIW	3.0	2.5
Pu	$NaHCO_3$, DIW, NaCl, $CaCl_2$, WIPP Brine	1.8	6.6
Np	$NaHCO_3$, NaCl, $CaCl_2$, WIPP Brine, DIW	1.5	2.8
Cm	$NaHCO_3$, DIW, NaCl, $CaCl_2$, WIPP Brine	4.4	9.4

DIW = Deionized water

TABLE 7. Results of Multiple Element Cumulative Fraction Release as a Function of a Single Solution Type (Figures 20-24)

Element	Observed Ranking of Element Release from Highest to Lowest Element Release	Ratio of Highest to Lowest Element Release at Time - 1 Day	Ratio of Highest to Lowest Element Release at end of Leach Time
WIPP Brine	Np, Am,. U, Pu, Cm	36	20
$CaCl_2$	Am, Cm, Pu, Np, U	7	5
NaCl	Np, Am, Cm, Pu, U	18	10
$NaHCO_3$	Np, U, Am, Pu, Cm	12	3
Deionized water	Np, Cm, Pu, Am, U	37	11

- The release mechanism for actinides is dependent upon the leach solution composition. The evidence for this can be seen from Table 6 where the trend with increasing leaching time is toward larger differences between leach solutions.
- For a particular leach solution, actinides element release appears to be approaching a common mechanism at extended leach times. As Table 7 shows, actinide release is tending toward convergence when leached from the same solution.
- It is particularly important that the actinides as a class, are leached at rates below that of the silica matrix. This means they are effectively being left behind in a chemical/physical form yet to be defined. As discussed earlier, post-leach surface characterization is just beginning. After two years at 25°C, no gel layer or residual heavy metal film is optically detected.

Figure 25 shows the cumulative fraction leached for ^{99}Tc. It is nearly flat as expected in view of the large drop in leach rate with time. It is interesting to note that after nearly 500 days of leaching, its fraction leached is practically the same as cesium and strontium from a fully radioactive glass sample leached for nearly the same time (Bradley 1978). The silicon leach rate also plotted in Figure 25 will likely cross the techneticum curve at about four years leach time. Near this time, as the glass continues to erode, (and depending on the bubble distribution) technetium will be released in bursts as individual pools are reached.

Finally, we would point out that results using this modified IAEA procedure have been related to continous flow tests (Weed et al. 1979). Previous studies and the present work all show indications that the frequency of changing solutions has a direct effect on the apparent leach rate. The cumulative release fraction curves for neptunium and plutonium depicted in Figure 20 for example, indicate that apparent rate changes correspond to the frequency of solution changes, i.e. daily, weekly or monthly. The changes are in the direction of decreasing leach rate with decreasing rate of solution exchange. Static and continuous flow leach tests are in progress to further clarify

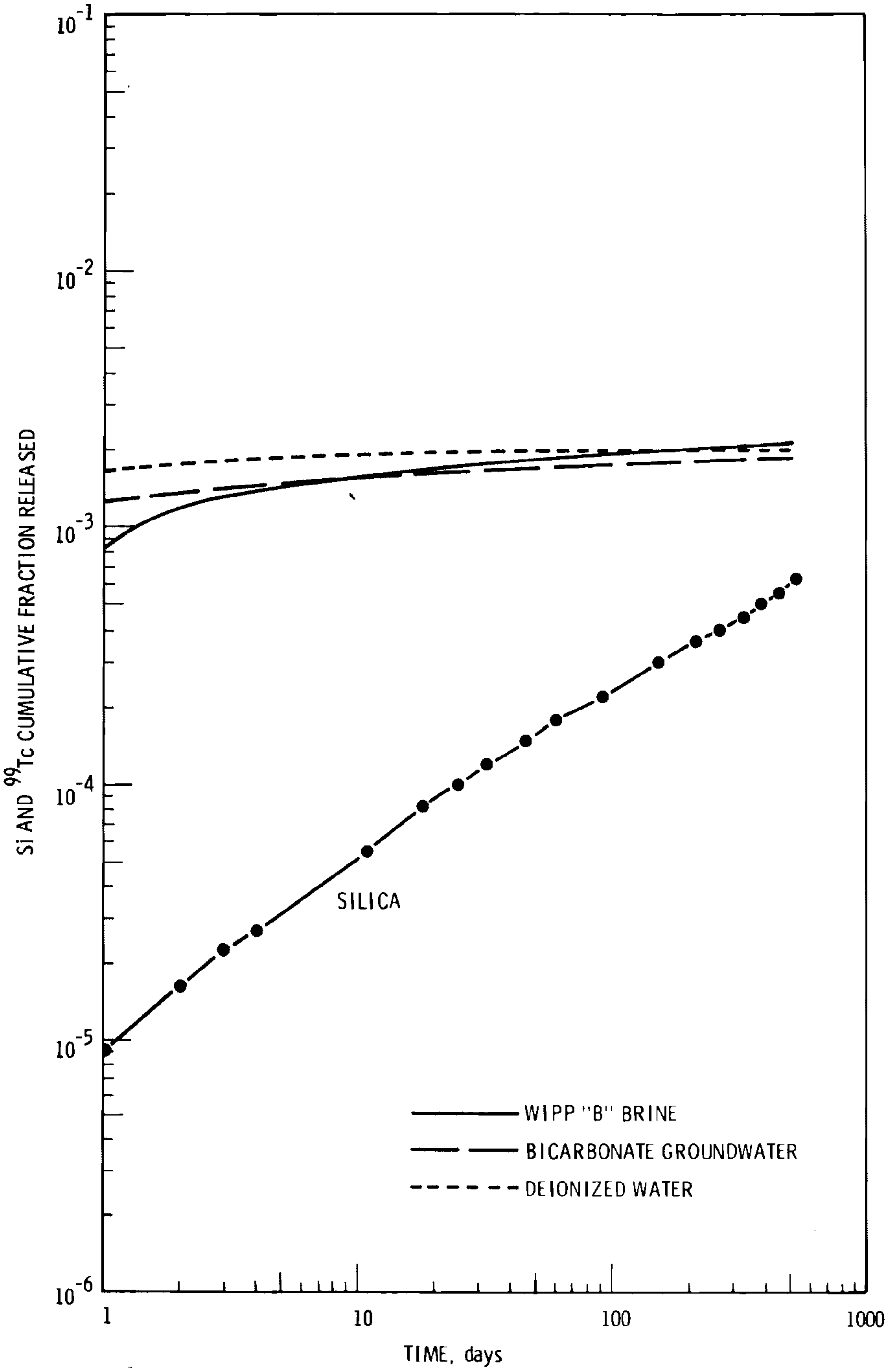


FIGURE 25. Cumulative Fractions of ^{99}Tc Released from Simulated Waste Glass 76-68, IAEA Leach Test at 22°C

(and quantify) this dependence. It is interesting to note that the cumulative fraction release curves for silicon in Figure 14 do not show clear changes related to the solution change frequency.

With respect to completing the current work, a selection of the leach solutions generated will be analyzed by induction coupled plasma (ICP) spectroscopy to obtain leach rates for some $\sim$20 elements present in the glass. In addition, the leached glass surface will be studied by scanning electron microscopy, microradiography, and alpha spectroscopy. Selected samples will also be evaluated by scanning Auger/ESCA, using ion milling techniques.

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APPENDIX A

LEACHING DATA

Leach Rate of Silicon from various doped glasses in Deionized water

Series #	Cumulative Days	Leach rate from ^{244}Cm glass, g/cm^2-day	Leach rate from $^{239}Pu + ^{237}Np$ glass, g/cm^2-day	Leach rate from ^{99}Tc glass, g/cm^2-day	Leach rate from $^{233}U + ^{243}Am$ glass, g/cm^2-day
1	1	4.5×10^{-6}	6.4×10^{-6}	2.3×10^{-6}	1.1×10^{-6}
2	2	5.0×10^{-6}	4.1×10^{-6}	1.7×10^{-6}	*
3	3	8.0×10^{-6}	2.9×10^{-6}	1.7×10^{-6}	5.6×10^{-7}
4	4	6.4×10^{-6}	1.2×10^{-6}	1.2×10^{-6}	5.6×10^{-7}
5	11	3.2×10^{-6}	5.8×10^{-7}	9.9×10^{-7}	1.1×10^{-6}
6	18	1.1×10^{-6}	5.0×10^{-7}	1.1×10^{-6}	1.6×10^{-7}
7	25	6.4×10^{-7}	6.6×10^{-6}	6.6×10^{-7}	8.0×10^{-7}
8	32	4.3×10^{-7}	4.1×10^{-7}	7.4×10^{-7}	1.6×10^{-7}
10	46	2.1×10^{-7}	5.8×10^{-7}	4.8×10^{-7}	2.4×10^{-7}
12	60	2.9×10^{-7}	6.6×10^{-7}	4.1×10^{-7}	2.4×10^{-7}
13	91	3.0×10^{-7}	5.6×10^{-7}	3.7×10^{-7}	2.4×10^{-7}
15	151	2.8×10^{-7}	2.7×10^{-7}	3.3×10^{-7}	9.3×10^{-8}
17	212	2.3×10^{-7}	2.9×10^{-7}	2.5×10^{-7}	1.9×10^{-7}
19	269	3.3×10^{-7}	2.7×10^{-7}	1.5×10^{-7}	9.3×10^{-8}
21	331	3.2×10^{-7}	3.7×10^{-7}	2.7×10^{-7}	1.3×10^{-7}
23	391	4.8×10^{-7}	3.1×10^{-7}	1.7×10^{-7}	1.1×10^{-7}
25	454	3.2×10^{-7}	2.7×10^{-7}	2.7×10^{-7}	1.1×10^{-7}
27	517	4.0×10^{-7}	4.1×10^{-7}	3.7×10^{-7}	1.1×10^{-7}
29	577	3.3×10^{-7}	5.2×10^{-7}		

* Data point not available

Release of Silicon in $CaCl_2$ solution from
^{239}Pu + ^{237}Np doped glass

Series #	Cumulative Days	Leach Rate g/cm²-day	Cumulative Fraction Leached
1	1	7.5×10^{-6}	2.9×10^{-5}
2	2	5.8×10^{-6}	5.2×10^{-5}
3	3	2.9×10^{-6}	6.3×10^{-5}
4	4	3.5×10^{-6}	7.7×10^{-5}
5	11	1.2×10^{-6}	1.1×10^{-4}
6	18	7.5×10^{-7}	1.3×10^{-4}
7	25	1.2×10^{-6}	1.6×10^{-4}
8	32	9.1×10^{-7}	1.9×10^{-4}
10	46	9.9×10^{-7}	2.2×10^{-4}
12	60	9.9×10^{-7}	2.9×10^{-4}
13	91	8.1×10^{-7}	3.9×10^{-4}
15	151	5.2×10^{-7}	5.3×10^{-4}
17	212	5.4×10^{-7}	6.5×10^{-4}
19	269	7.9×10^{-7}	8.2×10^{-4}
21	331	1.1×10^{-6}	1.0×10^{-3}
23	391	1.0×10^{-6}	1.3×10^{-3}
25	454	5.4×10^{-7}	1.4×10^{-3}
27	517	*	1.6×10^{-3}
29	577	7.2×10^{-7}	1.7×10^{-3}

* data point not available

Release of Silicon in NaCl solution from

$^{239}Pu + ^{237}Np$ doped glass

Series #	Cumulative Days	Leach Rate g/cm^2-day	Cumulative Fraction Leached
1	1	5.8×10^{-6}	2.3×10^{-5}
2	2	4.1×10^{-6}	4.6×10^{-5}
3	3	2.9×10^{-6}	5.0×10^{-5}
4	4	1.2×10^{-6}	5.5×10^{-5}
5	11	7.5×10^{-7}	7.6×10^{-5}
6	18	1.4×10^{-7}	9.2×10^{-5}
7	25	8.3×10^{-7}	1.1×10^{-4}
8	32	6.6×10^{-7}	1.3×10^{-4}
10	46	7.5×10^{-7}	1.7×10^{-4}
12	60	7.5×10^{-7}	2.1×10^{-4}
13	91	6.8×10^{-7}	2.9×10^{-4}
15	151	4.6×10^{-7}	4.2×10^{-4}
17	212	3.3×10^{-7}	5.0×10^{-4}
19	269	2.9×10^{-7}	5.7×10^{-4}
21	331	4.8×10^{-7}	6.8×10^{-4}
23	391	4.6×10^{-7}	7.9×10^{-4}
25	454	4.4×10^{-7}	8.9×10^{-4}
27	517	6.4×10^{-7}	1.0×10^{-4}
29	577	7.0×10^{-7}	1.2×10^{-4}

Release of Silicon in $NaHCO_3$ solution from ^{239}Pu + ^{237}Np doped glass

Series #	Cumulative Days	Leach Rate g/cm²-day	Cumulative Fraction Leached
1	1	6.7×10^{-6}	2.7×10^{-5}
2	2	6.4×10^{-6}	5.1×10^{-5}
3	3	4.6×10^{-6}	6.9×10^{-5}
4	4	5.2×10^{-6}	8.9×10^{-5}
5	11	2.6×10^{-6}	1.6×10^{-4}
6	18	2.7×10^{-6}	2.3×10^{-4}
7	25	2.1×10^{-6}	2.9×10^{-4}
8	32	2.6×10^{-6}	3.6×10^{-4}
10	46	1.7×10^{-6}	4.6×10^{-4}
12	60	2.6×10^{-6}	5.8×10^{-4}
13	91	2.0×10^{-6}	8.1×10^{-4}
15	151	1.2×10^{-6}	1.1×10^{-3}
17	212	1.8×10^{-6}	1.5×10^{-3}
19	269	2.0×10^{-6}	2.0×10^{-3}
21	331	2.2×10^{-6}	2.5×10^{-3}
23	391	2.7×10^{-6}	3.0×10^{-3}
25	454	1.5×10^{-7}	3.1×10^{-3}
27	517	2.4×10^{-6}	3.3×10^{-3}
29	577	2.1×10^{-7}	3.4×10^{-3}

Release of Silicon in Deionized water from

^{99}Tc doped glass

Series #	Cumulative Days	Leach Rate g/cm²-day	Cumulative Fraction Leached
1	1	2.3×10^{-6}	9.0×10^{-6}
2	2	1.7×10^{-6}	1.6×10^{-5}
3	3	1.7×10^{-6}	2.3×10^{-5}
4	4	1.2×10^{-6}	2.7×10^{-5}
5	11	9.9×10^{-7}	5.4×10^{-5}
6	18	1.1×10^{-6}	8.4×10^{-5}
7	25	6.6×10^{-7}	1.0×10^{-4}
8	32	7.4×10^{-7}	1.2×10^{-4}
10	46	4.9×10^{-7}	1.5×10^{-4}
12	60	4.1×10^{-7}	1.8×10^{-4}
13	91	3.7×10^{-7}	2.2×10^{-4}
15	151	3.3×10^{-7}	3.0×10^{-4}
17	212	2.5×10^{-7}	3.6×10^{-4}
19	269	1.5×10^{-7}	4.0×10^{-4}
21	331	2.7×10^{-7}	4.6×10^{-4}
23	391	1.7×10^{-7}	5.1×10^{-4}
25	454	2.7×10^{-7}	5.1×10^{-4}
27	517	3.7×10^{-7}	6.4×10^{-4}
29	577		

Release of Silicon in Deionized water from

$^{239}Pu + ^{237}Np$ doped glass

Series #	Cumulative Days	Leach Rate g/cm^2-day	Cumulative Fraction Leached
1	1	6.4×10^{-6}	2.5×10^{-5}
2	2	4.1×10^{-6}	4.0×10^{-5}
3	3	2.9×10^{-6}	5.2×10^{-5}
4	4	1.2×10^{-6}	5.6×10^{-5}
5	11	5.8×10^{-7}	7.2×10^{-5}
6	18	5.0×10^{-7}	8.5×10^{-5}
7	25	6.6×10^{-7}	1.0×10^{-4}
8	32	4.1×10^{-7}	1.1×10^{-4}
10	46	5.8×10^{-7}	1.4×10^{-4}
12	60	6.6×10^{-7}	1.8×10^{-4}
13	91	5.6×10^{-7}	2.4×10^{-4}
15	151	2.7×10^{-7}	3.2×10^{-4}
17	212	2.9×10^{-7}	3.9×10^{-4}
19	269	2.7×10^{-7}	4.5×10^{-4}
21	331	3.7×10^{-7}	5.3×10^{-4}
23	391	3.1×10^{-7}	6.1×10^{-4}
25	454	2.7×10^{-7}	6.7×10^{-4}
27	517	4.1×10^{-7}	7.6×10^{-4}
29	577	5.2×10^{-7}	8.7×10^{-4}

Release of ^{99}Tc in WIPP Brine

Series #	Cumulative Days	Leach Rate g/cm^2-day	Cumulative Fraction Leached
1	1	2.2×10^{-3}	8.46×10^{-3}
2	2	9.5×10^{-4}	1.22×10^{-2}
3	3	2.9×10^{-4}	1.33×10^{-2}
4	4	2.9×10^{-4}	1.44×10^{-2}
5	11	7.4×10^{-5}	1.64×10^{-2}
6	18	2.3×10^{-5}	1.71×10^{-2}
7	25	4.7×10^{-6}	1.72×10^{-2}
8	32	5.5×10^{-6}	1.74×10^{-2}
10	46	1.3×10^{-5}	1.80×10^{-2}
12	60	9.5×10^{-6}	1.85×10^{-2}
13	92	2.6×10^{-6}	1.88×10^{-2}
15	156	5.7×10^{-7}	1.91×10^{-2}
17	215	2.3×10^{-6}	1.95×10^{-2}
19	276	5.0×10^{-6}	2.06×10^{-2}
21	337	5.0×10^{-7}	2.09×10^{-2}
23	397	3.3×10^{-7}	2.10×10^{-2}
25	459	1.1×10^{-6}	2.12×10^{-2}
27	516	3.3×10^{-7}	2.13×10^{-2}

Release of ^{99}Tc in $CaCl_2$

Series #	Cumulative Days	Leach Rate g/cm^2-day	Cumulative Fraction Leached
1	1	2.4×10^{-3}	8.45×10^{-3}
2	2	3.0×10^{-4}	9.64×10^{-3}
3	3	1.4×10^{-4}	1.02×10^{-2}
4	4	1.5×10^{-4}	1.08×10^{-2}
5	11	3.2×10^{-5}	1.16×10^{-2}
6	18	2.1×10^{-5}	1.22×10^{-2}
7	25	1.4×10^{-5}	1.26×10^{-2}
8	32	7.7×10^{-6}	1.28×10^{-2}
10	46	3.9×10^{-6}	1.31×10^{-2}
12	60	4.4×10^{-6}	1.33×10^{-2}
13	92	3.8×10^{-6}	1.36×10^{-2}
15	156	2.6×10^{-6}	1.43×10^{-2}
17	215	2.1×10^{-6}	1.48×10^{-2}
19	276	1.7×10^{-6}	1.53×10^{-2}
21	337	1.1×10^{-6}	1.56×10^{-2}
23	397	8.1×10^{-7}	1.58×10^{-2}
25	459	8.9×10^{-7}	1.60×10^{-2}
27	516	1.3×10^{-6}	1.62×10^{-2}

Release of ^{99}Tc in NaCl

Series #	Cumulative Days	Leach Rate g/cm^2-day	Cumulative Fraction Leached
1	1	4.6×10^{-3}	1.8×10^{-2}
2	2	8.9×10^{-4}	2.15×10^{-2}
3	3	2.1×10^{-4}	2.23×10^{-2}
4	4	1.2×10^{-4}	2.28×10^{-2}
5	11	4.0×10^{-5}	2.39×10^{-2}
6	18	2.0×10^{-5}	2.45×10^{-2}
7	25	1.3×10^{-5}	2.48×10^{-2}
8	32	1.3×10^{-5}	2.51×10^{-2}
10	46	9.8×10^{-6}	2.57×10^{-2}
12	60	9.5×10^{-6}	2.62×10^{-2}
13	92	3.1×10^{-6}	2.66×10^{-2}
15	156	7.5×10^{-7}	2.70×10^{-2}
17	215	8.9×10^{-7}	2.72×10^{-2}
19	276	7.8×10^{-7}	2.73×10^{-2}
21	337	6.5×10^{-7}	2.75×10^{-2}
23	397	6.6×10^{-7}	2.77×10^{-2}
25	459	8.9×10^{-7}	2.79×10^{-2}
27	516	5.5×10^{-7}	2.80×10^{-2}

Release of ^{99}Tc in $NaHCO_3$

Series #	Cumulative Days	Leach Rate g/cm^2-day	Cumulative Fraction Leached
1	1	3.3×10^{-3}	1.29×10^{-2}
2	2	2.0×10^{-4}	1.37×10^{-2}
3	3	9.7×10^{-5}	1.41×10^{-2}
4	4	8.1×10^{-5}	1.44×10^{-2}
5	11	4.7×10^{-5}	1.57×10^{-2}
6	18	1.1×10^{-5}	1.60×10^{-2}
7	25	3.5×10^{-6}	1.61×10^{-2}
8	32	2.0×10^{-6}	1.61×10^{-2}
10	46	9.9×10^{-7}	1.62×10^{-2}
12	60	1.1×10^{-6}	1.63×10^{-2}
13	92	1.1×10^{-6}	1.64×10^{-2}
15	156	1.3×10^{-6}	1.67×10^{-2}
17	215	1.3×10^{-6}	1.70×10^{-2}
19	276	2.5×10^{-6}	1.75×10^{-2}
21	337	1.9×10^{-6}	1.79×10^{-2}
23	397	8.0×10^{-7}	1.82×10^{-2}
25	459	1.0×10^{-6}	1.84×10^{-2}
27	516	1.0×10^{-6}	1.86×10^{-2}

Release of ^{99}Tc in Deionized Water

Series #	Cumulative Days	Leach Rate g/cm²-day	Cumulative Fraction Leached
1	1	4.4×10^{-3}	1.69×10^{-2}
2	2	2.2×10^{-4}	1.78×10^{-2}
3	3	9.5×10^{-5}	1.82×10^{-2}
4	4	4.9×10^{-5}	1.84×10^{-2}
5	11	3.9×10^{-5}	1.94×10^{-2}
6	18	2.8×10^{-6}	1.95×10^{-2}
7	25	1.5×10^{-5}	1.99×10^{-2}
8	32	1.1×10^{-6}	2.00×10^{-2}
10	46	4.9×10^{-7}	2.00×10^{-2}
12	60	1.4×10^{-6}	2.01×10^{-2}
13	92	1.8×10^{-7}	2.01×10^{-2}
15	156	1.1×10^{-7}	2.01×10^{-2}
17	215	8.0×10^{-8}	2.01×10^{-2}
19	276	7.6×10^{-8}	2.02×10^{-2}
21	337	9.7×10^{-8}	2.02×10^{-2}
23	397	7.0×10^{-8}	2.02×10^{-2}
25	459	7.6×10^{-8}	2.02×10^{-2}
27	516	5.2×10^{-8}	2.02×10^{-2}

Release of ^{233}U in WIPP Brine

Series #	Cumulative Days	Leach Rate g/cm^2-day	Cumulative Fraction Leached
1	1	9.5×10^{-7}	3.6×10^{-6}
2	2	5.0×10^{-7}	5.5×10^{-6}
3	3	5.2×10^{-7}	7.2×10^{-6}
4	4	5.0×10^{-7}	9.4×10^{-6}
5	11	5.9×10^{-7}	2.5×10^{-5}
6	18	3.7×10^{-7}	3.5×10^{-5}
7	25	3.0×10^{-7}	4.3×10^{-5}
8	32	6.5×10^{-7}	6.0×10^{-5}
10	46	6.9×10^{-7}	9.6×10^{-5}

Release of ^{233}U in $CaCl_2$

Series #	Cumulative Days	Leach Rate g/cm^2-day	Cumulative Fraction Leached
1	1	4.4×10^{-7}	1.7×10^{-6}
2	2	$2.5 \times 10^{-}$	2.6×10^{-6}
3	3	2.2×10^{-7}	3.4×10^{-6}
4	4	2.1×10^{-7}	4.3×10^{-6}
5	11	1.1×10^{-7}	7.3×10^{-6}
6	18	1.6×10^{-7}	1.2×10^{-5}
7	25	2.0×10^{-7}	1.7×10^{-5}
8	32	1.6×10^{-7}	2.1×10^{-5}
10	46	1.5×10^{-7}	2.9×10^{-5}

Release of ^{233}U in NaCl

Series #	Cumulative Days	Leach Rate g/cm²-day	Cumulative Fraction Leached
1	1	5.3×10^{-7}	2.0×10^{-6}
2	2	1.9×10^{-7}	2.7×10^{-6}
3	3	1.5×10^{-7}	3.3×10^{-6}
4	4	1.2×10^{-7}	3.8×10^{-6}
5	11	3.6×10^{-8}	4.7×10^{-6}
6	18	9.3×10^{-8}	7.2×10^{-6}
7	25	1.3×10^{-7}	1.1×10^{-5}
8	32	1.3×10^{-7}	1.4×10^{-5}
10	46	1.7×10^{-7}	2.3×10^{-5}

Release of ^{233}U in $NaHCO_3$

Series #	Cumulative Days	Leach Rate g/cm^2-day	Cumulative Fraction Leached
1	1	1.7×10^{-6}	6.4×10^{-6}
2	2	1.4×10^{-6}	1.2×10^{-5}
3	3	9.2×10^{-7}	1.5×10^{-5}
4	4	9.5×10^{-7}	1.9×10^{-5}
5	11	1.2×10^{-6}	5.0×10^{-5}
6	18	8.6×10^{-7}	7.3×10^{-5}
7	25	8.3×10^{-7}	9.4×10^{-5}
8	32	9.9×10^{-7}	1.2×10^{-4}
10	46	1.2×10^{-6}	1.8×10^{-4}

Release of ^{233}U in Deionized Water

Series #	Cumulative Days	Leach Rate g/cm^2-day	Cumulative Fraction Leached
1	1	4.0×10^{-7}	1.6×10^{-6}
2	2	3.0×10^{-7}	2.7×10^{-6}
3	3	1.6×10^{-7}	3.3×10^{-6}
4	4	1.2×10^{-7}	3.8×10^{-6}
5	11	6.7×10^{-8}	5.6×10^{-6}
6	18	5.8×10^{-8}	7.2×10^{-6}
7	25	4.4×10^{-8}	8.2×10^{-6}
8	32	5.7×10^{-8}	9.6×10^{-6}
10	46	1.3×10^{-7}	1.6×10^{-5}

Release of ^{237}Np in WIPP Brine

Series #	Cumulative Days	Leach Rate g/cm²-day	Cumulative Fraction Leached
1	1	1.0×10^{-5}	3.9×10^{-5}
2	2	4.0×10^{-6}	5.4×10^{-5}
3	3	1.5×10^{-6}	6.0×10^{-5}
4	4	3.3×10^{-6}	7.3×10^{-5}
5	11	5.3×10^{-7}	8.7×10^{-5}
6	18	3.3×10^{-7}	9.6×10^{-5}
7	25	6.6×10^{-7}	1.1×10^{-4}
8	32	2.8×10^{-7}	1.2×10^{-4}
10	46	9.7×10^{-7}	1.6×10^{-4}
12	60	2.3×10^{-7}	2.7×10^{-4}
13	91	4.8×10^{-7}	3.3×10^{-4}
15	151	3.4×10^{-7}	4.1×10^{-4}

Release of ^{237}Np in $CaCl_2$

Series #	Cumulative Days	Leach Rate g/cm^2-day	Cumulative Fraction Leached
1	1	1.0×10^{-5}	4.1×10^{-5}
2	2	5.9×10^{-6}	6.4×10^{-5}
3	3	7.9×10^{-7}	6.7×10^{-5}
4	4	3.5×10^{-6}	8.0×10^{-5}
5	11	1.5×10^{-6}	1.2×10^{-4}
6	18	1.5×10^{-6}	1.6×10^{-4}
7	25	1.5×10^{-6}	2.0×10^{-4}
8	32	8.7×10^{-7}	2.2×10^{-4}
10	46	8.5×10^{-7}	2.7×10^{-4}
12	60	1.1×10^{-6}	3.3×10^{-4}
13	91	5.2×10^{-7}	3.9×10^{-4}
15	151	4.5×10^{-7}	5.0×10^{-4}

Release of ^{237}Np in NaCl

Series #	Cumulative Days	Leach Rate g/cm^2-day	Cumulative Fraction Leached
1	1	8.9×10^{-6}	3.5×10^{-5}
2	2	2.4×10^{-6}	4.5×10^{-5}
3	3	2.4×10^{-6}	5.4×10^{-5}
4	4	4.5×10^{-6}	7.2×10^{-5}
5	11	8.4×10^{-7}	9.5×10^{-5}
6	18	1.3×10^{-6}	1.3×10^{-4}
7	25	9.6×10^{-7}	1.6×10^{-4}
8	32	7.5×10^{-7}	1.8×10^{-4}
10	46	8.9×10^{-7}	2.3×10^{-4}
12	60	2.3×10^{-7}	3.3×10^{-4}
13	91	5.9×10^{-7}	4.1×10^{-4}
15	151	3.6×10^{-7}	5.1×10^{-4}

Release of ^{237}Np in $NaHCO_3$

Series #	Cumulative Days	Leach Rate g/cm²-day	Cumulative Fraction Leached
1	1	9.6×10^{-6}	3.7×10^{-5}
2	2	3.4×10^{-6}	5.0×10^{-5}
3	3	3.7×10^{-6}	6.4×10^{-5}
4	4	3.7×10^{-6}	7.8×10^{-5}
5	11	2.1×10^{-6}	1.4×10^{-4}
6	18	7.5×10^{-7}	1.6×10^{-4}
7	25	1.9×10^{-6}	2.0×10^{-4}
8	32	1.5×10^{-6}	2.4×10^{-4}
10	46	1.8×10^{-6}	3.4×10^{-4}
12	60	3.2×10^{-6}	4.9×10^{-4}
13	91	1.5×10^{-6}	6.6×10^{-4}
15	151	9.9×10^{-7}	9.2×10^{-4}

Release of ^{237}Np in NaCl

Series #	Cumulative Days	Leach Rate g/cm²-day	Cumulative Fraction Leached
1	1	8.9×10^{-6}	3.5×10^{-5}
2	2	2.4×10^{-6}	4.5×10^{-5}
3	3	2.4×10^{-6}	5.4×10^{-5}
4	4	4.5×10^{-6}	7.2×10^{-5}
5	11	8.4×10^{-7}	9.5×10^{-5}
6	18	1.3×10^{-6}	1.3×10^{-4}
7	25	9.6×10^{-7}	1.6×10^{-4}
8	32	7.5×10^{-7}	1.8×10^{-4}
10	46	8.9×10^{-7}	2.3×10^{-4}
12	60	2.3×10^{-7}	3.3×10^{-4}
13	91	5.9×10^{-7}	4.1×10^{-4}
15	151	3.6×10^{-7}	5.1×10^{-4}

Release of ^{237}Np in $NaHCO_3$

Series #	Cumulative Days	Leach Rate g/cm^2-day	Cumulative Fraction Leached
1	1	9.6×10^{-6}	3.7×10^{-5}
2	2	3.4×10^{-6}	5.0×10^{-5}
3	3	3.7×10^{-6}	6.4×10^{-5}
4	4	3.7×10^{-6}	7.8×10^{-5}
5	11	2.1×10^{-6}	1.4×10^{-4}
6	18	7.5×10^{-7}	1.6×10^{-4}
7	25	1.9×10^{-6}	2.0×10^{-4}
8	32	1.5×10^{-6}	2.4×10^{-4}
10	46	1.8×10^{-6}	3.4×10^{-4}
12	60	3.2×10^{-6}	4.9×10^{-4}
13	91	1.5×10^{-6}	6.6×10^{-4}
15	151	9.9×10^{-7}	9.2×10^{-4}

^{237}Np in Deionized Water

Series #	Cumulative Days	Leach Rate g/cm²-day	Cumulative Fraction Leached
1	1	7.4×10^{-6}	2.9×10^{-5}
2	2	5.1×10^{-6}	4.8×10^{-5}
3	3	3.5×10^{-6}	6.2×10^{-5}
4	4	3.7×10^{-6}	7.5×10^{-5}
5	11	7.5×10^{-7}	9.5×10^{-5}
6	18	3.4×10^{-7}	1.0×10^{-4}
7	25	8.0×10^{-7}	1.3×10^{-4}
8	32	4.8×10^{-7}	1.4×10^{-4}
10	46	7.5×10^{-7}	1.8×10^{-4}
12	60	5.6×10^{-7}	2.1×10^{-4}
13	91	4.6×10^{-7}	2.6×10^{-4}
14	121	[illegible] x 10^{-}	3.1×10^{-4}
15	151	2.4×10^{-7}	3.3×10^{-4}

Release of ^{237}Np in Deionized Water

Series #	Cumulative Days	Leach Rate g/cm^2-day	Cumulative Fraction Leached
1	1	7.4×10^{-6}	2.9×10^{-5}
2	2	5.1×10^{-6}	4.8×10^{-5}
3	3	3.5×10^{-6}	6.2×10^{-5}
4	4	3.7×10^{-6}	7.5×10^{-5}
5	11	7.5×10^{-7}	9.5×10^{-5}
6	18	3.4×10^{-7}	1.0×10^{-4}
7	25	8.0×10^{-7}	1.3×10^{-4}
8	32	4.8×10^{-7}	1.4×10^{-4}
10	46	7.5×10^{-7}	1.8×10^{-4}
12	60	5.6×10^{-7}	2.1×10^{-4}
13	91	4.6×10^{-7}	2.6×10^{-4}
15	151	2.4×10^{-7}	3.3×10^{-4}

Release of ^{239}Pu in WIPP Brine

Series #	Cumulative Days	Leach Rate g/cm²-day	Cumulative Fraction Leached
1	1	2.6×10^{-6}	9.8×10^{-6}
2	2	5.8×10^{-7}	1.2×10^{-5}
3	3	9.1×10^{-8}	1.2×10^{-5}
4	4	3.6×10^{-7}	1.4×10^{-5}
5	11	4.6×10^{-8}	1.5×10^{-5}
6	18	2.1×10^{-8}	1.6×10^{-5}
7	25	3.2×10^{-8}	1.6×10^{-5}
8	32	2.8×10^{-8}	1.7×10^{-5}
10	46	5.3×10^{-8}	2.0×10^{-5}
12	60	1.8×10^{-7}	2.8×10^{-5}
13	91	2.5×10^{-8}	3.1×10^{-5}
15	151	2.2×10^{-8}	3.6×10^{-5}

Release of ^{239}Pu in $CaCl_2$

Series #	Cumulative Days	Leach Rate g/cm^2-day	Cumulative Fraction Leached
1	1	3.2×10^{-6}	1.2×10^{-5}
2	2	1.5×10^{-6}	1.8×10^{-5}
3	3	8.9×10^{-8}	1.8×10^{-5}
4	4	1.0×10^{-6}	2.2×10^{-5}
5	11	1.7×10^{-7}	2.7×10^{-5}
6	18	1.3×10^{-7}	3.1×10^{-5}
7	25	2.0×10^{-7}	3.6×10^{-5}
8	32	1.1×10^{-7}	3.9×10^{-5}
10	46	1.4×10^{-7}	4.6×10^{-5}
12	60	6.1×10^{-8}	5.0×10^{-5}
13	91	8.4×10^{-9}	5.1×10^{-5}
15	151	1.6×10^{-8}	5.5×10^{-5}

Release of ^{239}Pu in NaCl

Series #	Cumulative Days	Leach Rate g/cm²-day	Cumulative Fraction Leached
1	1	2.5×10^{-6}	9.7×10^{-6}
2	2	9.6×10^{-7}	1.4×10^{-5}
3	3	4.8×10^{-7}	1.5×10^{-5}
4	4	1.9×10^{-6}	2.3×10^{-5}
5	11	2.0×10^{-6}	2.8×10^{-5}
6	18	1.4×10^{-7}	3.2×10^{-5}
7	25	2.1×10^{-7}	3.8×10^{-5}
8	32	1.3×10^{-7}	4.2×10^{-5}
10	46	1.9×10^{-7}	5.1×10^{-5}
12	60	1.6×10^{-7}	6.0×10^{-5}
13	91	6.8×10^{-8}	6.8×10^{-5}
15	151	4.3×10^{-8}	8.0×10^{-5}

Release of ^{239}Pu in $NaHCO_3$

Series #	Cumulative Days	Leach Rate g/cm^2-day	Cumulative Fraction Leached
1	1	4.6×10^{-6}	1.7×10^{-5}
2	2	1.9×10^{-6}	2.5×10^{-5}
3	3	1.1×10^{-6}	2.9×10^{-5}
4	4	1.7×10^{-6}	3.5×10^{-5}
5	11	6.4×10^{-7}	5.3×10^{-5}
6	18	5.3×10^{-7}	6.7×10^{-5}
7	25	3.3×10^{-7}	7.6×10^{-5}
8	32	5.9×10^{-7}	9.1×10^{-5}
10	46	5.4×10^{-7}	1.2×10^{-4}
12	60	6.7×10^{-7}	1.5×10^{-4}
13	91	2.6×10^{-7}	1.8×10^{-4}
15	151	2.2×10^{-7}	2.4×10^{-4}

Release of ^{239}Pu in Deionized Water

Series #	Cumulative Days	Leach Rate g/cm^2-day	Cumulative Fraction Leached
1	1	2.9×10^{-6}	1.1×10^{-5}
2	2	1.1×10^{-6}	1.5×10^{-5}
3	3	4.6×10^{-7}	1.7×10^{-5}
4	4	1.2×10^{-6}	2.2×10^{-5}
5	11	2.3×10^{-7}	2.8×10^{-5}
6	18	1.3×10^{-7}	3.2×10^{-5}
7	25	4.5×10^{-7}	4.4×10^{-5}
8	32	1.2×10^{-7}	4.7×10^{-5}
10	46	1.6×10^{-7}	5.5×10^{-5}
12	60	1.3×10^{-7}	6.3×10^{-5}
13	91	5.7×10^{-8}	6.9×10^{-5}
15	151	4.8×10^{-8}	8.1×10^{-5}

Release of ^{243}Am in WIPP Brine

Series #	Cumulative Days	Leach Rate g/cm^2-day	Cumulative Fraction Leached
1	1	2.1×10^{-6}	8.2×10^{-6}
2	2	6.5×10^{-7}	1.0×10^{-5}
3	3	3.9×10^{-7}	1.2×10^{-5}
4	4	3.8×10^{-7}	1.3×10^{-5}
5	11	3.0×10^{-7}	2.2×10^{-5}
6	18	2.3×10^{-7}	2.7×10^{-5}
7	25	2.2×10^{-7}	3.3×10^{-5}
8	32	5.6×10^{-7}	4.8×10^{-5}
10	46	5.9×10^{-7}	8.2×10^{-5}

Release of ^{243}Am in $CaCl_2$

Series #	Cumulative Days	Leach Rate g/cm^2-day	Cumulative Fraction Leached
1	1	1.6×10^{-6}	6.3×10^{-6}
2	2	7.0×10^{-7}	8.9×10^{-6}
3	3	3.5×10^{-7}	1.1×10^{-5}
4	4	7.2×10^{-7}	1.4×10^{-5}
5	11	4.0×10^{-7}	2.4×10^{-5}
6	18	5.5×10^{-7}	3.9×10^{-5}
7	25	5.0×10^{-7}	5.3×10^{-5}
8	32	5.1×10^{-7}	6.6×10^{-5}
10	46	2.6×10^{-7}	8.2×10^{-5}

Release of ^{243}Am in NaCl

Series #	Cumulative Days	Leach Rate g/cm^2-day	Cumulative Fraction Leached
1	1	1.3×10^{-6}	4.9×10^{-6}
2	2	4.7×10^{-7}	6.7×10^{-6}
3	3	4.3×10^{-7}	8.2×10^{-6}
4	4	4.1×10^{-7}	9.6×10^{-6}
5	11	1.8×10^{-7}	1.5×10^{-5}
6	18	4.1×10^{-7}	2.5×10^{-5}
7	25	5.1×10^{-7}	3.9×10^{-5}
8	32	5.3×10^{-7}	5.3×10^{-5}
10	46	5.0×10^{-7}	8.2×10^{-5}

Release of ^{243}Am in $NaHCO_3$

Series #	Cumulative Days	Leach Rate g/cm^2-day	Cumulative Fraction Leached
1	1	1.5×10^{-6}	5.6×10^{-6}
2	2	8.9×10^{-7}	8.9×10^{-6}
3	3	8.9×10^{-7}	1.2×10^{-5}
4	4	9.6×10^{-7}	1.6×10^{-5}
5	11	5.7×10^{-7}	3.1×10^{-5}
6	18	5.2×10^{-7}	4.5×10^{-5}
7	25	5.0×10^{-7}	5.8×10^{-5}
8	32	5.6×10^{-7}	7.3×10^{-5}
10	46	5.3×10^{-7}	1.0×10^{-4}

Release of ^{243}Am in Deionized Water

Series #	Cumulative Days	Leach Rate g/cm^2-day	Cumulative Fraction Leached
1	1	7.3×10^{-7}	2.8×10^{-6}
2	2	4.7×10^{-7}	4.6×10^{-6}
3	3	3.3×10^{-7}	5.9×10^{-6}
4	4	2.2×10^{-7}	6.6×10^{-6}
5	11	2.7×10^{-7}	1.3×10^{-5}
6	18	1.9×10^{-7}	1.9×10^{-5}
7	25	1.5×10^{-7}	2.3×10^{-5}
8	32	1.4×10^{-7}	2.7×10^{-5}
10	46	3.7×10^{-7}	4.3×10^{-5}

Release of ^{244}Cm in WIPP Brine

Series #	Cumulative Days	Leach Rate g/cm²-day	Cumulative Fraction Leached
1	1	4.3×10^{-7}	1.6×10^{-6}
2	2	9.9×10^{-8}	1.9×10^{-6}
3	3	1.6×10^{-7}	2.5×10^{-6}
4	4	1.3×10^{-7}	3.0×10^{-6}
5	11	5.7×10^{-8}	4.5×10^{-6}
6	18	3.6×10^{-8}	5.4×10^{-6}
7	25	3.1×10^{-8}	6.2×10^{-6}
8	32	4.8×10^{-8}	7.5×10^{-6}
10	45	7.5×10^{-8}	1.1×10^{-5}
12	60	8.1×10^{-8}	1.5×10^{-5}
13	90	3.6×10^{-8}	1.9×10^{-5}
15	150	1.1×10^{-7}	3.9×10^{-5}
17	212	2.5×10^{-8}	4.9×10^{-5}
19	269	2.8×10^{-8}	5.5×10^{-5}
21	331	2.2×10^{-8}	6.0×10^{-5}
23	391	1.8×10^{-8}	6.4×10^{-5}
25	454	1.4×10^{-8}	6.7×10^{-5}
27	517	1.9×10^{-8}	7.1×10^{-5}

Release of ^{244}Cm in $CaCl_2$

Series #	Cumulative Days	Leach Rate g/cm^2-day	Cumulative Fraction Leached
1	1	1.1×10^{-6}	3.9×10^{-6}
2	2	3.1×10^{-7}	5.1×10^{-6}
3	3	5.9×10^{-7}	7.2×10^{-6}
4	4	9.9×10^{-8}	1.1×10^{-5}
5	11	4.4×10^{-7}	2.2×10^{-5}
6	18	3.8×10^{-7}	3.2×10^{-5}
7	25	3.5×10^{-7}	4.1×10^{-5}
8	32	5.6×10^{-7}	5.6×10^{-5}
10	45	7.0×10^{-7}	9.0×10^{-5}
12	60	4.8×10^{-7}	1.2×10^{-5}
13	90	2.3×10^{-7}	1.4×10^{-5}
15	150	8.2×10^{-8}	1.7×10^{-5}
17	212	1.2×10^{-7}	1.9×10^{-5}
19	269	1.4×10^{-7}	2.2×10^{-5}
21	331	1.9×10^{-7}	2.6×10^{-5}
23	391	1.4×10^{-7}	3.0×10^{-5}
25	454	1.3×10^{-7}	3.3×10^{-5}

Release of ^{244}Cm in $NaHCO_3$

Series #	Cumulative Days	Leach Rate g/cm^2-day	Cumulative Fraction Leached
1	1	8.8×10^{-7}	3.2×10^{-6}
2	2	9.2×10^{-7}	6.5×10^{-6}
3	3	8.1×10^{-7}	9.5×10^{-6}
4	4	6.9×10^{-7}	1.2×10^{-5}
5	11	2.9×10^{-7}	1.9×10^{-5}
6	18	2.5×10^{-7}	2.6×10^{-5}
7	25	3.7×10^{-7}	3.5×10^{-5}
8	32	8.9×10^{-7}	5.8×10^{-5}
10	45	1.1×10^{-6}	1.1×10^{-4}
12	60	6.9×10^{-7}	1.5×10^{-4}
13	90	2.4×10^{-7}	1.8×10^{-4}
15	150	3.0×10^{-7}	2.4×10^{-4}
17	212	1.7×10^{-7}	2.8×10^{-4}
19	269	3.6×10^{-7}	3.5×10^{-4}
21	331	3.7×10^{-7}	4.3×10^{-4}
23	391	4.9×10^{-7}	5.3×10^{-4}
25	454	4.5×10^{-7}	6.3×10^{-4}
27	517	1.9×10^{-7}	6.9×10^{-4}

Release of ^{244}Cm in NaCl

Series #	Cumulative Days	Leach Rate g/cm^2-day	Cumulative Fraction Leached
1	1	6.4×10^{-7}	2.4×10^{-6}
2	2	3.3×10^{-7}	3.6×10^{-6}
3	3	6.9×10^{-7}	6.1×10^{-6}
4	4	1.2×10^{-6}	1.0×10^{-5}
5	11	4.6×10^{-7}	2.2×10^{-5}
6	18	4.3×10^{-7}	3.3×10^{-5}
7	25	3.2×10^{-7}	4.2×10^{-5}
8	32	4.0×10^{-7}	5.2×10^{-5}
10	45	8.6×10^{-7}	9.0×10^{-5}
12	60	5.4×10^{-7}	1.2×10^{-4}
13	90	2.6×10^{-7}	1.5×10^{-4}
15	150	1.0×10^{-7}	1.8×10^{-4}
17	212	1.7×10^{-7}	2.2×10^{-4}
19	269	1.7×10^{-7}	2.5×10^{-4}
21	331	1.3 x 10-7	2.9×10^{-4}
23	391	2.4×10^{-7}	3.3×10^{-4}
25	454	2.2×10^{-7}	3.8×10^{-4}

Release of ^{244}Cm in Deionized Water

Series #	Cumulative Days	Leach Rate g/cm^2-day	Cumulative Fraction Leached
1	1	2.2×10^{-7}	7.9×10^{-7}
2	2	7.8×10^{-7}	1.1×10^{-6}
3	3	3.3×10^{-7}	2.3×10^{-6}
4	4	9.1×10^{-7}	5.6×10^{-6}
5	11	5.3×10^{-7}	1.9×10^{-5}
6	18	1.3×10^{-6}	5.2×10^{-5}
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8	32	7.1×10^{-7}	8.9×10^{-5}
10	45	8.7×10^{-7}	1.3×10^{-4}
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13	90	2.1×10^{-7}	1.8×10^{-4}
15	150	9.3×10^{-8}	2.1×10^{-4}
17	212	3.8×10^{-7}	2.7×10^{-4}
19	269	1.2×10^{-7}	3.1×10^{-4}
21	331	1.4×10^{-7}	3.4×10^{-4}
23	391	1.5×10^{-7}	3.7×10^{-4}
25	454	2.7×10^{-7}	4.3×10^{-4}
27	517	4.1×10^{-7}	5.1×10^{-4}

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