3F

PNL-3173
UC-70

Waste/Rock Interactions Technology Program

Status Report on LWR Spent Fuel IAEA Leach Tests

Y. B. Katayama
D. J. Bradley
C. O. Harvey

March 1980

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
by Battelle Memorial Institute

PNL-3173

Battelle

PACIFIC NORTHWEST LABORATORY
operated by
BATTELLE
for the
UNITED STATES DEPARTMENT OF ENERGY
Under Contract DE-AC06-76RLO 1830

Printed in the United States of America
Available from
National Technical Information Service
United States Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22151

Price: Printed Copy $______*; Microfiche $3.00

*Pages	NTIS Selling Price
001-025	$4.00
026-050	$4.50
051-075	$5.25
076-100	$6.00
101-125	$6.50
126-150	$7.25
151-175	$8.00
176-200	$9.00
201-225	$9.25
226-250	$9.50
251-275	$10.75
276-300	$11.00

PNL-3173
UC-70

Waste/Rock Interactions Technology Program

STATUS REPORT ON LWR SPENT FUEL IAEA LEACH TESTS

Y. B. Katayama
D. J. Bradley
C. O. Harvey

March 1980

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

Pacific Northwest Laboratory
Richland, Washington 99352

SUMMARY

Spent light-water-reactor (LWR) fuel with an average burnup of 28,000 MWd/MTU was leach-tested at 25°C using a modified version of the International Atomic Energy Agency (IAEA) procedure. Leach rates were determined from tests conducted in five different solutions: deionized water, sodium chloride (NaCl), sodium bicarbonate ($NaHCO_3$), calcium chloride ($CaCl_2$) and Waste Isolation Pilot Plant (WIPP) "B" brine solutions. Elemental leach rates are reported based on the release of ^{90}Sr + ^{90}Y, ^{106}Ru, ^{137}Cs, ^{144}Ce, ^{154}Eu, $^{239+240}Pu$, ^{244}Cm and total uranium.

After 467 days of cumulative leaching, the elemental leach rates are highest in deionized water. The elemental leach rates in the different solutions generally decreased from deionized water to the 0.03M NaCl solution to the WIPP "B" brine solution to the 0.03M $NaHCO_3$ solution and was a factor of 20 lower in 0.015M $CaCl_2$ solution than in deionized water.

The leach rates of spent fuel and borosilicate waste-glass were also compared. In sodium bicarbonate solution, the leach rates of the two waste forms were nearly equal, but the glass was increasingly more resistant than spent fuel in calcium chloride solution, followed by sodium chloride solution, WIPP "B" brine solution and deionized water. In deionized water the glass, based on the elemental release of plutonium and curium, was 50 to 400 times more leach resistant than spent fuel.

ACKNOWLEDGMENT

This research was supported by the Waste/Rock Interactions Technology Program conducted by Pacific Northwest Laboratory. The Waste/Rock Interactions Technology program is sponsored by the Office of Nuclear Waste Isolation, which is managed by Battelle Memorial Institute for the U.S. Department of Energy under contract DE-AC06-76RLO 1830.

The authors are grateful to C. E. Bigelow, H. J. Cartmell and J. C. Nelson for their assistance in performing the experiments.

CONTENTS

SUMMARY iii

ACKNOWLEDGMENT v

INTRODUCTION 1

EXPERIMENTAL METHODS AND MATERIALS 2

MATERIAL 2

LEACH TEST. 3

ANALYTICAL PROCEDURE. 9

CALCULATIONS 13

RESULTS/DISCUSSION 14

CONCLUSIONS 34

REFERENCES. 35

APPENDIX--LEACH DATA. A-1

FIGURES

1 Appearance of Spent Fuel with a Burnup of 28,000 MWd/MTU . . . 4

2 Fragment of HB Robinson II Fuel, 28,000 MWd/MTU, Showing Microprobe Step-Scanning Path From point A to point B (Edge to Center) . . . 5

3 Microprobe Measured X-Ray Intensities for Plutonium, Technetium and Barium. . . . 6

4 Microprobe Measured X-Ray Intensities for Cesium, Zirconium and Iodine. . . . 7

5 Microprobe Measured X-Ray Intensities for Ruthenium, Tellurium and Cerium. . . . 8

6 Spent LWR Fuel Fragments Photographed Through Hot-Cell Periscope 11

7 IAEA Leach-Test Container. . . . 12

8 Leach Rate of 28,000 MWd/MTU Spent Fuel at 25°C Based on Release of Uranium 15

9 Leach Rate of 28,000 MWd/MTU Spent Fuel at 25°C Based on Release of ^{137}Cs 16

10 Leach Rate of 28,000 MWd/MTU Spent Fuel at 25°C Based on Release of ^{144}Ce 17

11 Leach Rate of 28,000 MWd/MTU Spent Fuel at 25°C Based on Release of $^{239+240}Pu$ 18

12 Leach Rate of 28,000 MWd/MTU Spent Fuel at 25°C Based on Release of ^{244}Cm 19

13 Leach Rate of 28,000 MWd/MTU Spent Fuel at 25°C Based on Release of $^{90}Sr + ^{90}Y$ 20

14 Leach Rate of 28,000 MWd/MTU Spent Fuel at 25°C Based on Release of ^{154}Eu 21

15 Leach Rate of 28,000 MWd/MTU Spent Fuel at 25°C Based on Release of ^{125}Sb 22

16 Leach Rate of 28,000 MWd/MTU Spent Fuel at 25°C Based on Release of ^{106}Ru 23

17	Leach Rate of 28,000 MWd/MTU Spent Fuel in Deionized Water at 25°C Based on Selected Elements	25
18	Leach Rate of 28,000 MWd/MTU Spent Fuel in 0.03M NaCl at 25°C Based on Selected Elements	26
19	Leach Rate of 28,000 MWd/MTU Spent Fuel in WIPP "B" Saturated Brine Solution at 25°C Based on Selected Elements	27
20	Leach Rate of 28,000 MWd/MTU Spent Fuel in 0.03M $NaHCO_3$ at 25°C Based on Selected Elements	28
21	Leach Rate of 28,000 MWd/MTU Spent Fuel in 0.015M $CaCl_2$ Solution at 25°C Based on Selected Elements	30

TABLES

1 Particle Size Distribution of 28,000 MWd/MTU Spent Fuel. . . . 2

2 IAEA Leach-Test Schedule 10

3 Chemical and Ionic Composition of Salt Brine 11

4 Results of Effects of Solution Type of Each Element . . . 24

5 Relative Leachability of Spent fuel in Various Leach Solutions Based on Average Trend of Leach Curves After 467 Days of Cumulative Leaching 31

6 Results of Comparing Incremental Leach Rates of Elements for Each Solution Type 31

7 Comparison of IAEA Leach Rates at 25°C 31

8 Comparison of IAEA Leach Rates of Paige-Test Leach Rates (467 days). 33

INTRODUCTION

In 1977, the Pacific Northwest Laboratory (PNL), operated by Battelle Memorial Institute for the Department of Energy, started a task to study spent-fuel leaching as a part of the Waste Isolation Safety Assessment Program (WISAP). Radionuclide release information from spent fuel, a candidate waste form for geologic disposal, is needed to evaluate the safety of repository storage of spent fuels. These data are used both in release-consequence modeling and as source terms for radionuclide migration experiments.

There are many types of leach tests being used to evaluate solidified waste forms (Mendel 1973). At PNL spent fuels have been leach-tested by three different procedures: Paige tests, static leach tests, and the IAEA procedure. Paige leach tests on spent LWR fuels have been previously reported (Katayama 1979), and results from static leach tests of spent fuel will be reported as radiochemical analyses of stored and ongoing leachant samples are completed. The static leach tests are designed to give detailed data on spent fuel solubilities at 25° and 75°C. This status report presents results from leach-testing LWR spent fuel with an average burnup of 28,000 MWd/MTU using a modified version of the IAEA procedure at 25°C. Release rates are discussed as a function of the duration of the leach tests and of the solution type.

This and future status reports on spent-fuel leaching will be reported as a part of the Waste/Rock Interactions Technology Program (WRIT). The WRIT Program, started in FY-1980 at PNL, includes Tasks 2 and 4 of WISAP.

EXPERIMENTAL METHODS AND MATERIALS

MATERIAL

The spent fuel used in this study originated from a fuel bundle discharged on June 6, 1974 from the HB Robinson II Reactor and has an average burnup of 28,000 MWd/MTU. The fuel was removed from the fuel rods at the Battelle-Columbus Hot Laboratory and was received at PNL as a mixture of unclad fuel fragments. A sample of this material was submitted to the Hanford Engineering Development Laboratory for radiochemical analysis. These analytical values for the radionuclides were used as the initial concentration in the leach rate calculations.

A randomly selected batch of the as-received fuel fragments was screened to provide a particle size distribution (see Table 1). Samples for the leach tests were taken from particle fractions retained on screens #5 and #10 (greater than 2-mm particles).

TABLE 1. Particle Size Distribution of 28,000 MWd/MTU Spent Fuel

Sieve Number	Sieve Opening, mm	Weight, g	Fraction Retained
3	6.73	0	0
4	4.73	192.883	1.007×10^{-1}
5	4.00	634.765	3.331×10^{-1}
10	2.00	1031.170	5.384×10^{-1}
20	0.841	35.205	1.838×10^{-2}
40	0.420	11.242	5.869×10^{-3}
60	0.250	4.979	2.599×10^{-3}
80	0.177	1.424	7.434×10^{-4}
100	0.149	1.042	5.440×10^{-4}
140	0.105	1.204	6.286×10^{-4}
200	0.074	0.769	4.015×10^{-4}
200	0.074	0.737	3.848×10^{-4}

Samples of the as-received spent fuel were also mounted, polished and examined by metallography for microstructural characterization of the fuel. Metallographic examination of spent-fuel fragments from the HB Robinson II reactor showed the presence of closed porosity (see Figure 1). Cathodic etching of the surface showed equiaxed grains with little change in size from the center of the pellet to the outer edge (magnified portion in Figure 1 is typical of entire sample).

Chemical-concentration profiles for selected radionuclides were recorded as fluorescence X-ray intensities on a shielded electron-beam microprobe X-ray analyzer. Various fuel fragments, typical of the samples in our leach tests, are now being analyzed. The data presented here are for a fragment of fuel with a burnup of 28,000 MWd/MTU. Figure 2 shows a segment of a transverse section. The microprobe was programmed to step-scan the sample from point A at the outside diameter of the pellet to point B near the center of the pellet. Concentration profiles for elements measured by step scanning are expressed as X-ray intensities in Figures 3, 4 and 5.

Plutonium, cesium, ruthenium, technetium, barium, zirconium and cerium showed enrichment near the outer edge (point A). Iodine and tellurium showed no indication of enrichment at the outer edge. Plutonium showed the highest gradient from edge to center (point A to point B), with a 47% reduction in X-ray intensity 300 μm inward from point A and an additional 28% drop over the next 3600 μm to point B.

LEACH TEST

The modified IAEA leach test involves the immersion of a spent-fuel sample in a solution according to a fixed ratio of 1:10 (cm^2/cm^3) of exposed surface area of sample to volume of solution. The solution is then left in contact with the sample for progressively longer time intervals. The solution is changed after each time interval. Between each sampling period the system is not disturbed via stirring or shaking. Although the leach container is left static during the leaching interval, the thermal power of the spent fuel of about 5 cal h^{-1} g^{-1} is a source of convective (thermal) agitation at the

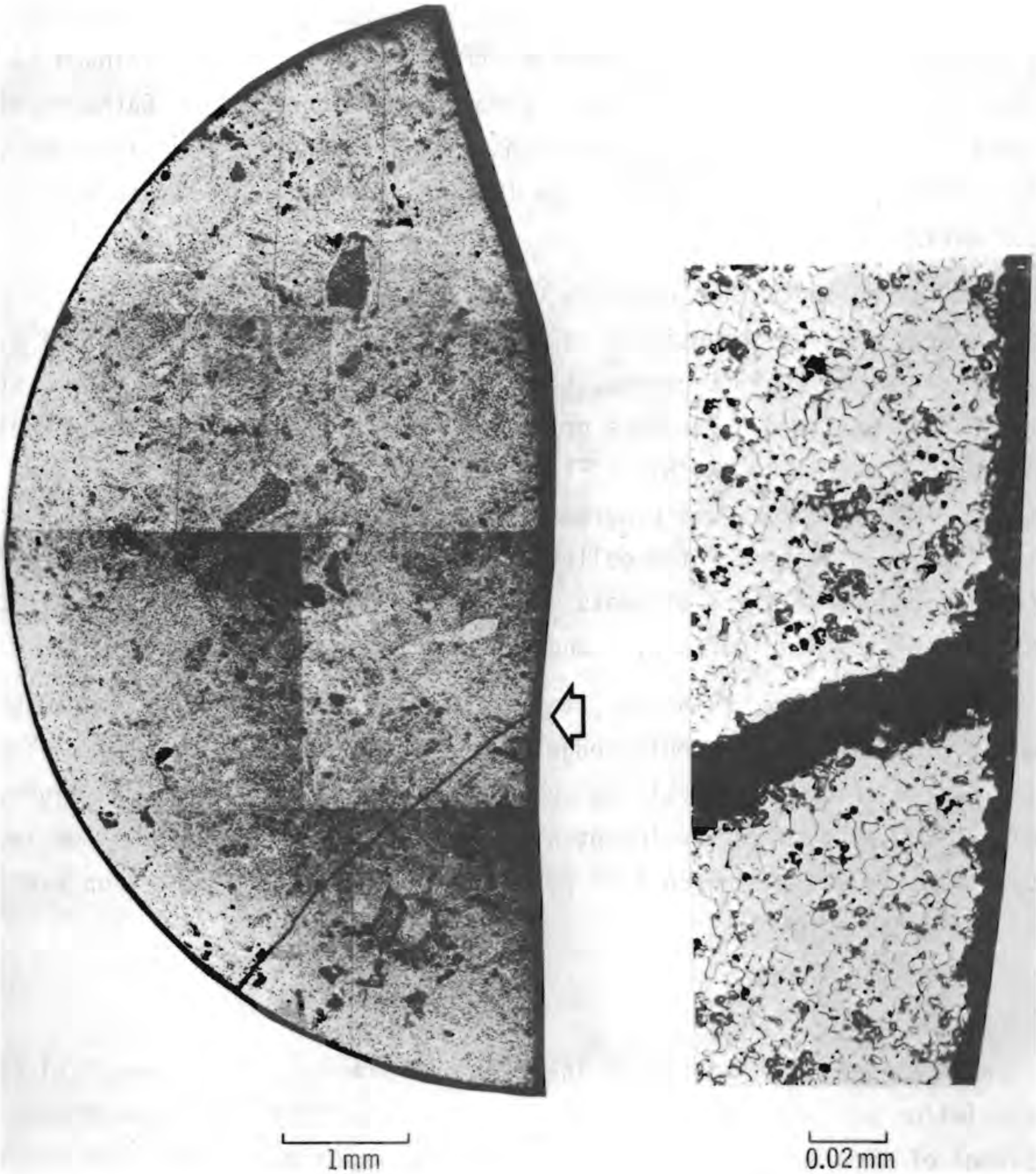

FIGURE 1. Appearance of Spent Fuel with a Burnup of 28,000 MWd/MTU

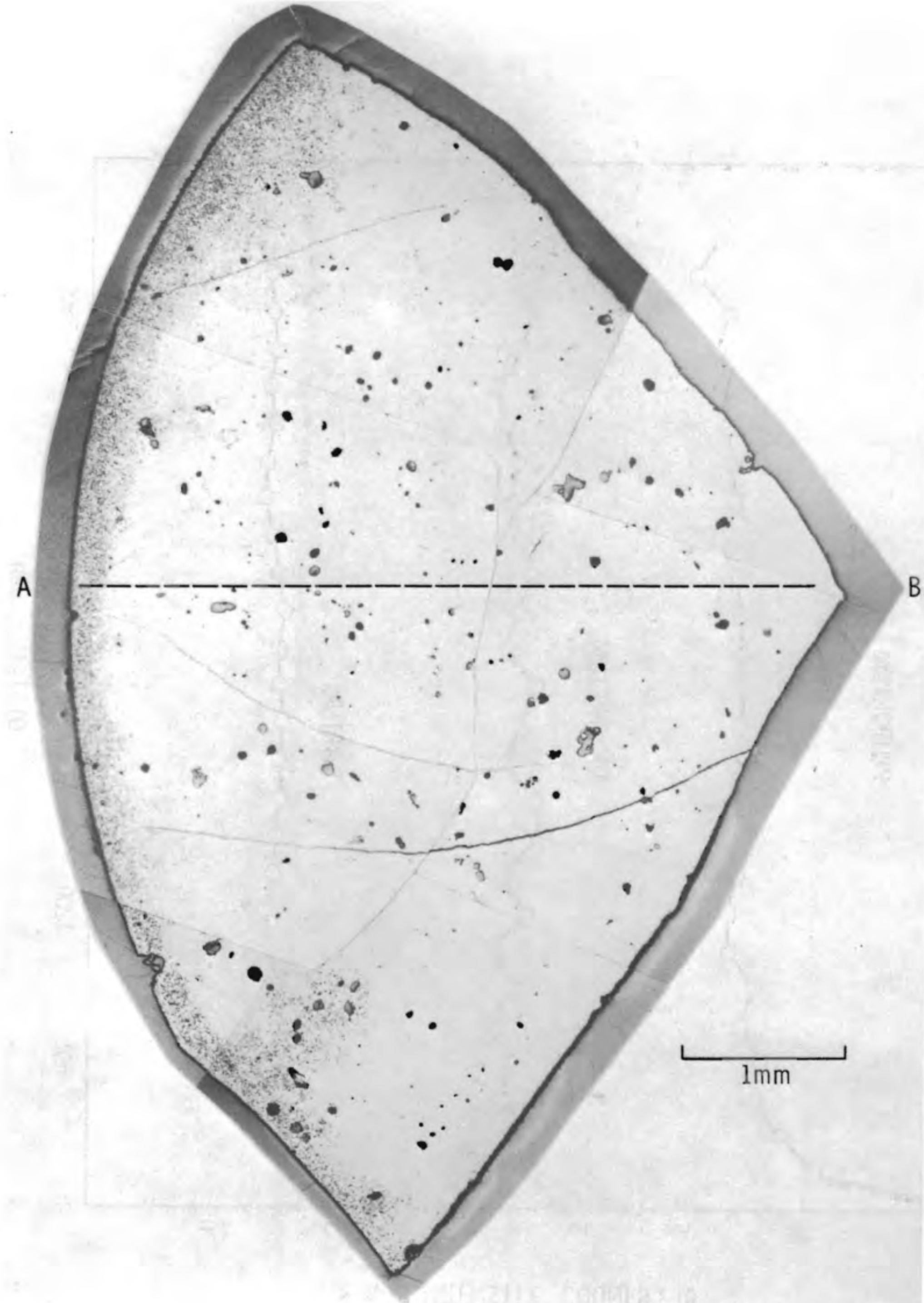

FIGURE 2. Fragment of HB Robinson II Fuel, 28,000 MWd/MTU, Showing Microprobe Step-Scanning Path From point A to point B (Edge to Center)

A-B ≅ 4.7 mm

DOE-Richland, WA

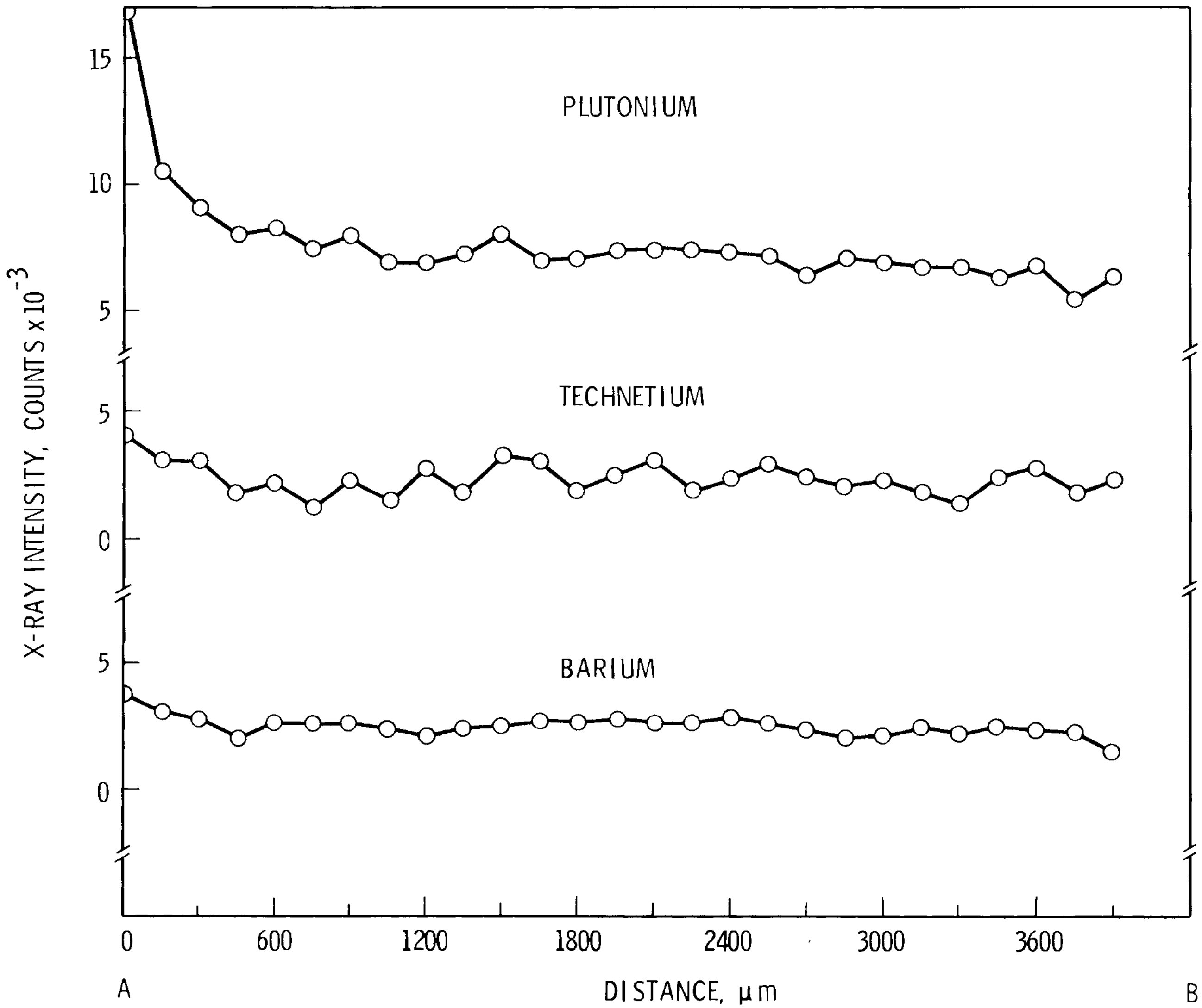

FIGURE 3. Microprobe Measured X-Ray Intensities for Plutonium, Technetium and Barium

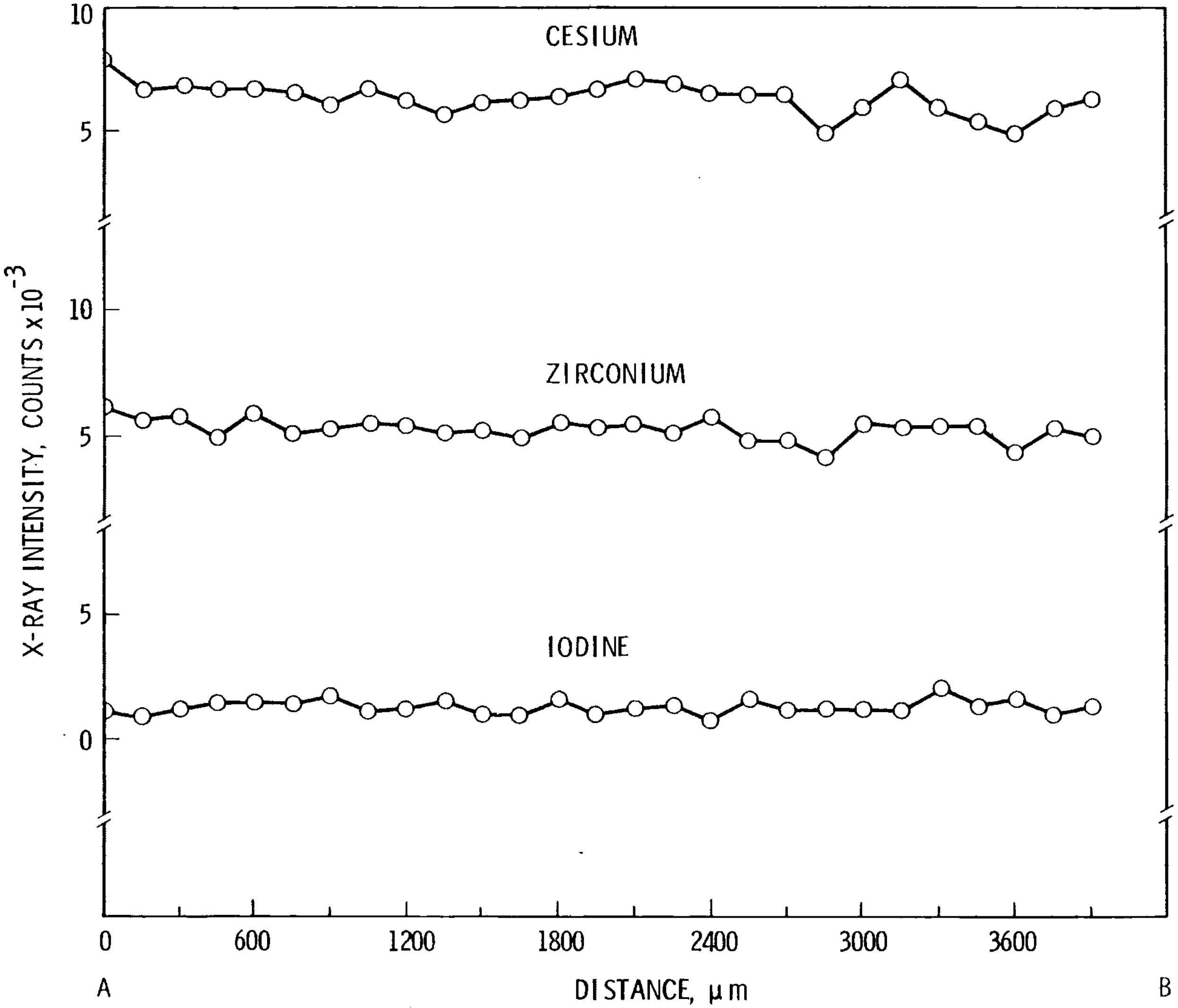

FIGURE 4. Microprobe Measured X-Ray Intensities for Cesium, Zirconium and Iodine

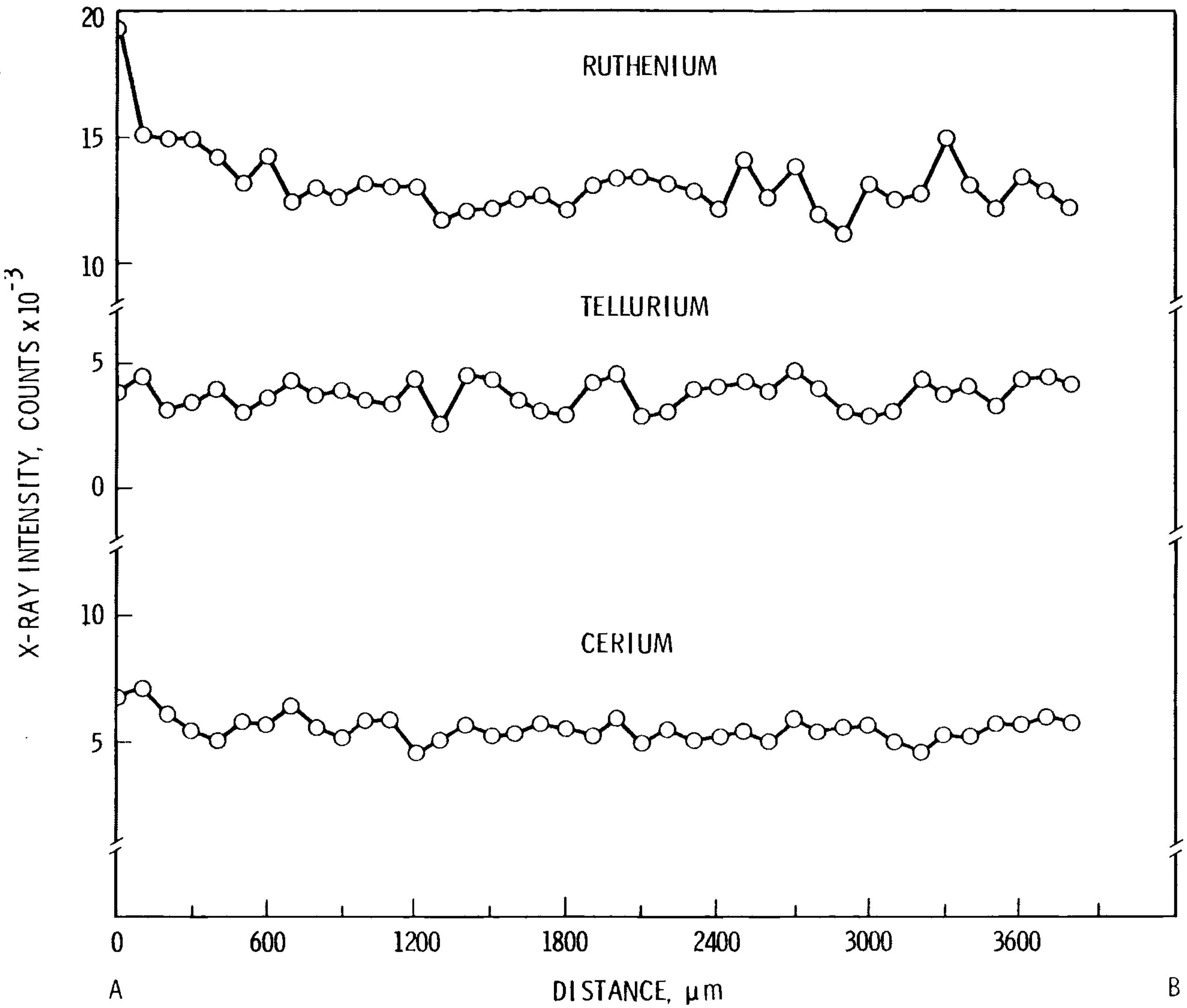

FIGURE 5. Microprobe Measured X-Ray Intensities for Ruthenium, Tellurium and Cerium

fuel-solution interface. Thus, the transfer of radionuclides to the bulk of the leach solution may not be wholly dependent on diffusion. Table 2 shows the sampling schedule for the IAEA test used.

Approximately 15 g of fuel fragments (see Figure 6), having a geometric surface area of approximately 30 cm^2, were used per test. Figure 7 shows the details of the leach-test container. For these tests the solution volume was 300 ml. The solutions used for the spent-fuel leach tests were:

- salt brine, WIPP "B" salt brine (Dosch and Lynch 1978)
- synthetic high-ionic-strength calcium groundwater (0.015$\underline{M}$ $CaCl^2$)
- synthetic high-bicarbonate groundwater (0.03$\underline{M}$ $NaHCO_3$)
- synthetic high-ionic-strength sodium groundwater (0.03$\underline{M}$ NaCl)
- deionized water.

Table 3 shows the composition of WIPP "B" salt brine.

ANALYTICAL PROCEDURE

The tests were run in triplicate, making a total of 15 tests. On each sample collection day, the basket holding the spent-fuel fragments was carefully removed, and after swirling the jar of solution, a 10-ml sample of solution was withdrawn. This sample was then acidified to approximately pH 1 using concentrated nitric acid to prevent radionuclides from adhering to the walls of the glass sample-container. The addition of nitric acid was found to be effective in preventing nuclide plateout in IAEA leach testing of doped glass beads (Bradley, Harvey and Turcotte 1979).

After discarding the remaining leach solution, the leach-test polypropylene jars (see Figure 7) were then filled with 300 ml of 5$\underline{M}$ HNO_3 + 0.5$\underline{M}$ HF. This solution was used to remove any radionuclides that had adhered to the leach-test jar walls. After one to two days, a 10-ml sample was withdrawn and analyzed. The result of this radionuclide concentration analysis was added to that from the original leach solution to arrive at a leach rate of a given radionuclide from the spent fuel. The measurements of radionuclides were all made using common radiochemical analysis methods. Gamma emitters were measured by gamma-energy analysis using a multi-channel analyzer with a Ge-Li detector.

TABLE 2. IAEA Leach-Test Schedule

Cumulative Days Leached	Solution Change, Series Number	Leach Solutions to be Analyzed
1	1	x[a]
2	2	x[a]
3	3	x[a]
4	4	x[a]
11	5	x[a]
18	6	x
25	7	x[a]
32	8	x
39	9	
46	10	x
53	11	
60	12	x[a]
91	13	x
122	14	
154	15	x[a]
187	16	
215	17	x
246	18	
277	19	x[a]
310	20	
341	21	x
374	22	
404	23	x[a]
433	24	
467	25	x[a]
495	26	
529	27	x

(a) Results presented in this report.

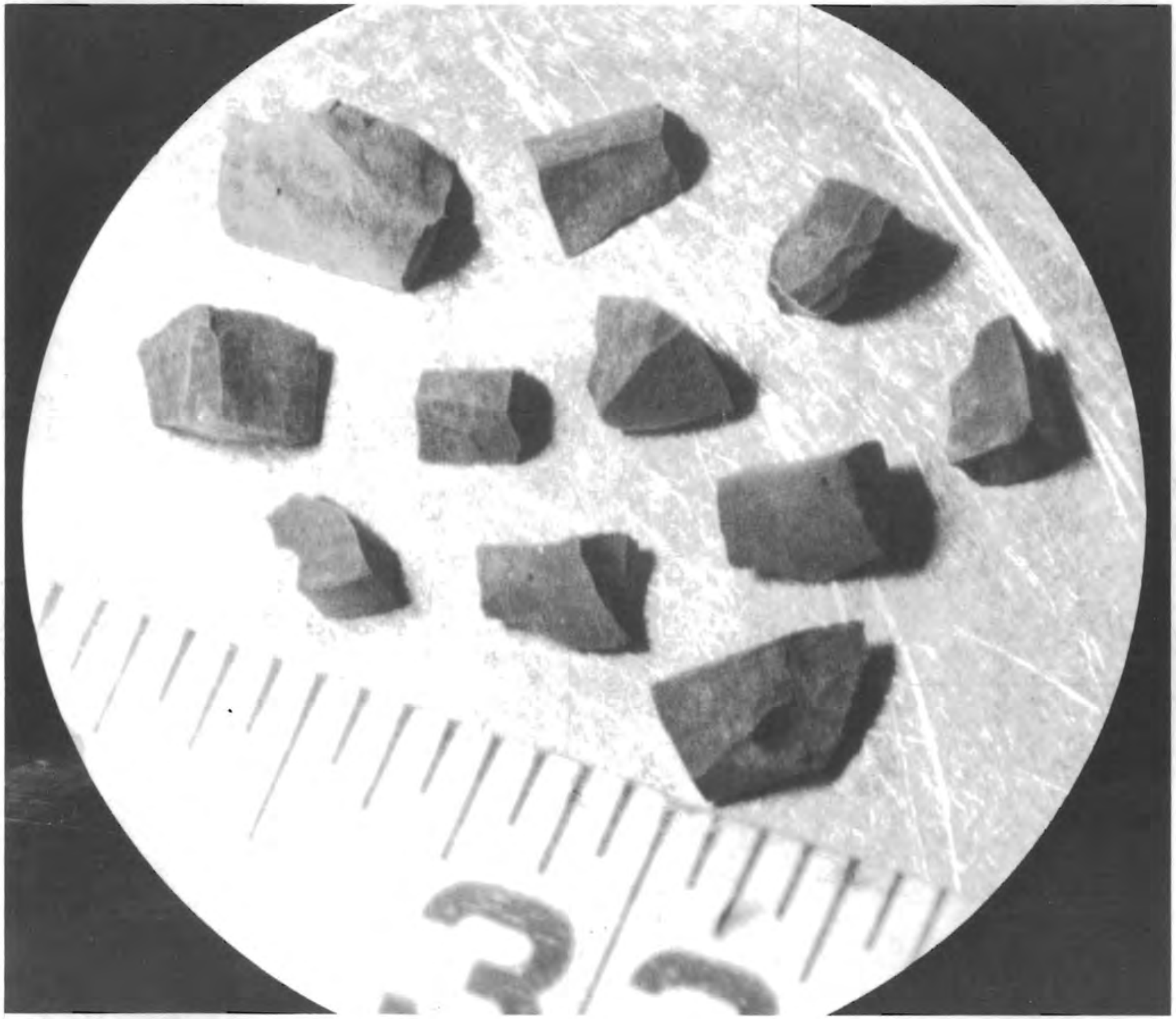

FIGURE 6. Spent LWR Fuel Fragments Photographed Through Hot-Cell Periscope[a]

(a) each division on scale equals 1/16 in.

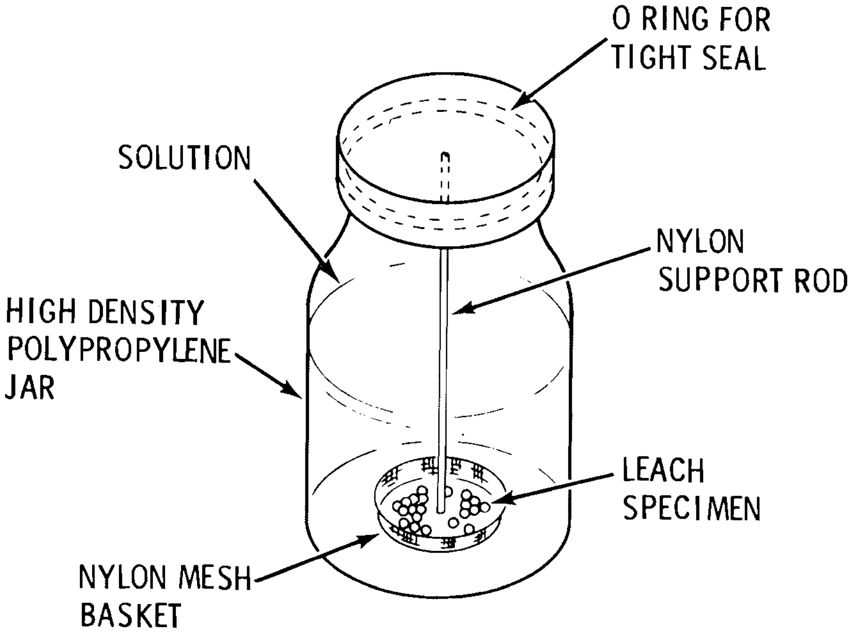

FIGURE 7. IAEA Leach-Test Container

TABLE 3. Chemical and Ionic Composition of Salt Brine[a]

Compound	Concentration, g/L	Ion	Concentration, mole/L
$NaCl$	287.0	Na^+	5.0
Na_2SO_4	6.2	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
$CaCl_2$ $2H_2O$	0.0033	Cl^-	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 solids	297.2	B (BO_3^{---})	0.00017

(a) pH (adjusted) = 6.5

To improve the measurement of minor constituents, the majority of the cesium was removed by extraction with tetraphenylboron in amylacetate (Finston 1961) and the sample recounted.

Plutonium and curium analyses, with the exception of brine samples, were done by alpha-energy analysis of a direct mount of the sample. In brine samples, the plutonium was extracted into TTA-xylene (Moore and Hudgens 1957), plated and counted on an alpha proportional counter. Curium was separated by ion exchange, plated and alpha counted.

Strontium was separated by ion exchange and was beta-counted. Repeat counts were made and the ^{90}Sr was calculated from the ^{90}Y ingrowth (Koltoff and Elving 1966). Uranium analysis was done by fluorometry (Centanni and DeSesa 1956; Price, Perritti and Swaity et al. 1953).

CALCULATIONS

All the leach rates in this report are incremental leach rates and are average leach rates for the sampling interval. The equation used to calculate the incremental leach rate is as follows:

$$R_i = \frac{a_i}{A_0 S t} \text{ , where}$$

R_i = incremental leach rate, g/cm^2-day

a_i = activity of isotope in leachate, counts s^{-1}

A_0 = specific activity of isotope in sample before leaching, counts s^{-1} g^{-1}

S = geometric surface area of sample, cm^2

t = incremental leaching period, days.

RESULTS/DISCUSSION

Figures 8 through 16 are graphs of incremental leach rates for nine radioactive elements plotted as a function of time. In each figure, leach curves for the five leachants are shown for one element. Each data point represents the average of three samples, and tabulations of the data are given in the Appendix. The elemental leach curves for the five solutions diverge with cumulative leaching time, and for the elements analyzed the spread in the leach rate curves is greatest at the last data point reported (467 days). The leach rates based on cesium, antimony and ruthenium release appear to be the least affected by the different leach solutions. For the other six elements, the leach rate curves for the five different leach solutions are spread out up to 2-1/2 orders of magnitude. The leach rate curves in Figures 8 through 16 are summarized in Table 4. Table 4 contains a tabulation of the observed ranking of leach solutions from the highest to lowest elemental releases. Deionized water had the highest release for all the isotopes. The lowest release for the isotopes was in $CaCl_2$ solution except for cerium, ruthenium and europium where the $CaCl_2$ solution was next to lowest.

Figure 17 is a graph of leach rate curves in deionized water for the nine elements measured. The spread in the curves at the first day of leaching covers two orders of magnitude and decreases to a spread of one order of magnitude at 467 days of cumulative leaching. Cesium has the highest initial leach rate and continues to exhibit a high leach rate at the last sampling period. Ruthenium has the lowest initial leach rate and continues to exhibit the lowest leach rate after 467 days.

Figure 18 is a graph of leach rate curves in 0.03M NaCl solution for the nine elements measured. The spread in the leach curves at the first day of leaching covers about 1-1/2 order of magnitude and decreases to about one order of magnitude at 467 days of cumulative leaching. Cesium tends to have the highest leach rate and ruthenium the lowest leach rate during the 467 days, which is the same general behavior as in deionized water.

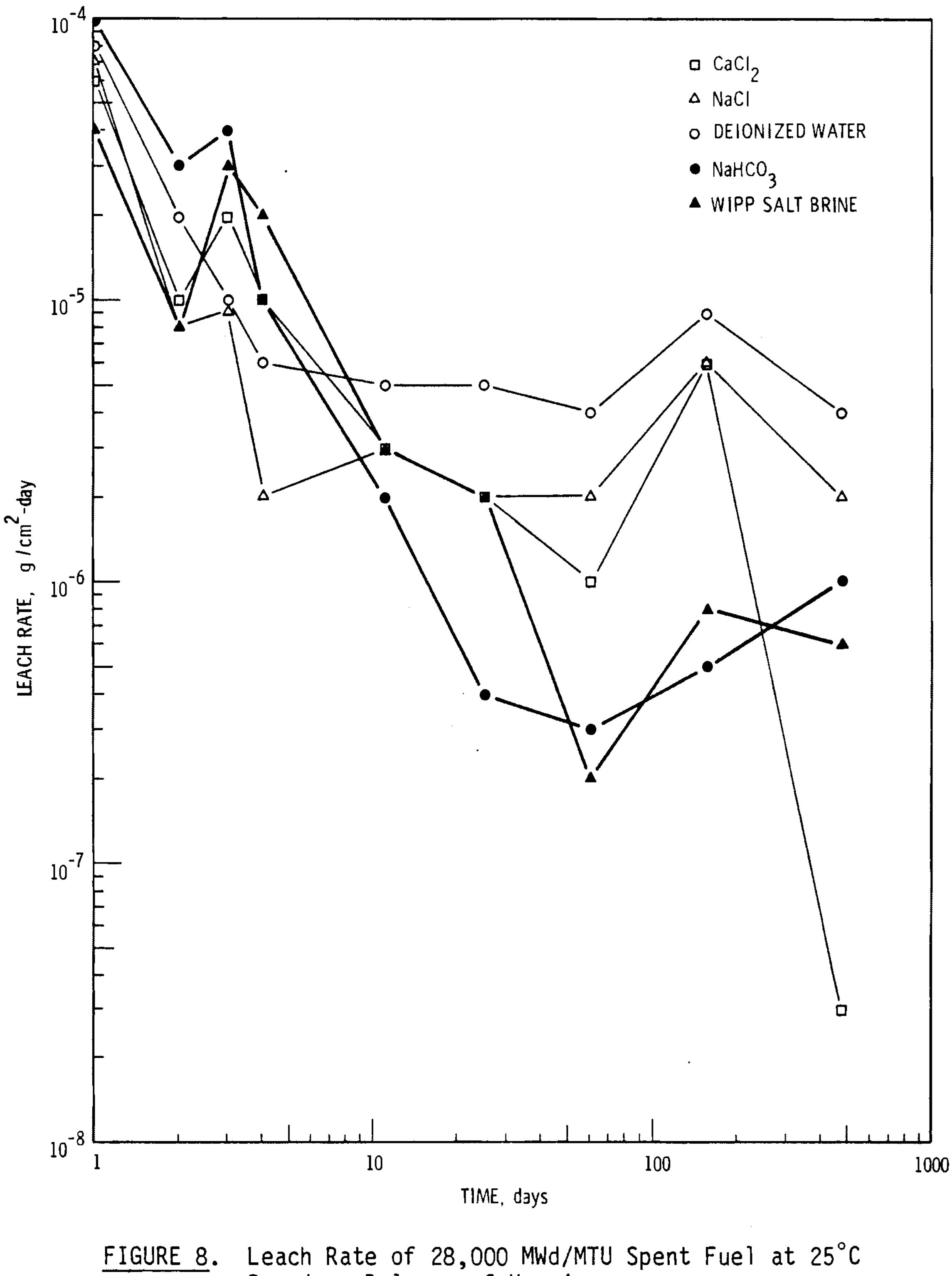

FIGURE 8. Leach Rate of 28,000 MWd/MTU Spent Fuel at 25°C Based on Release of Uranium

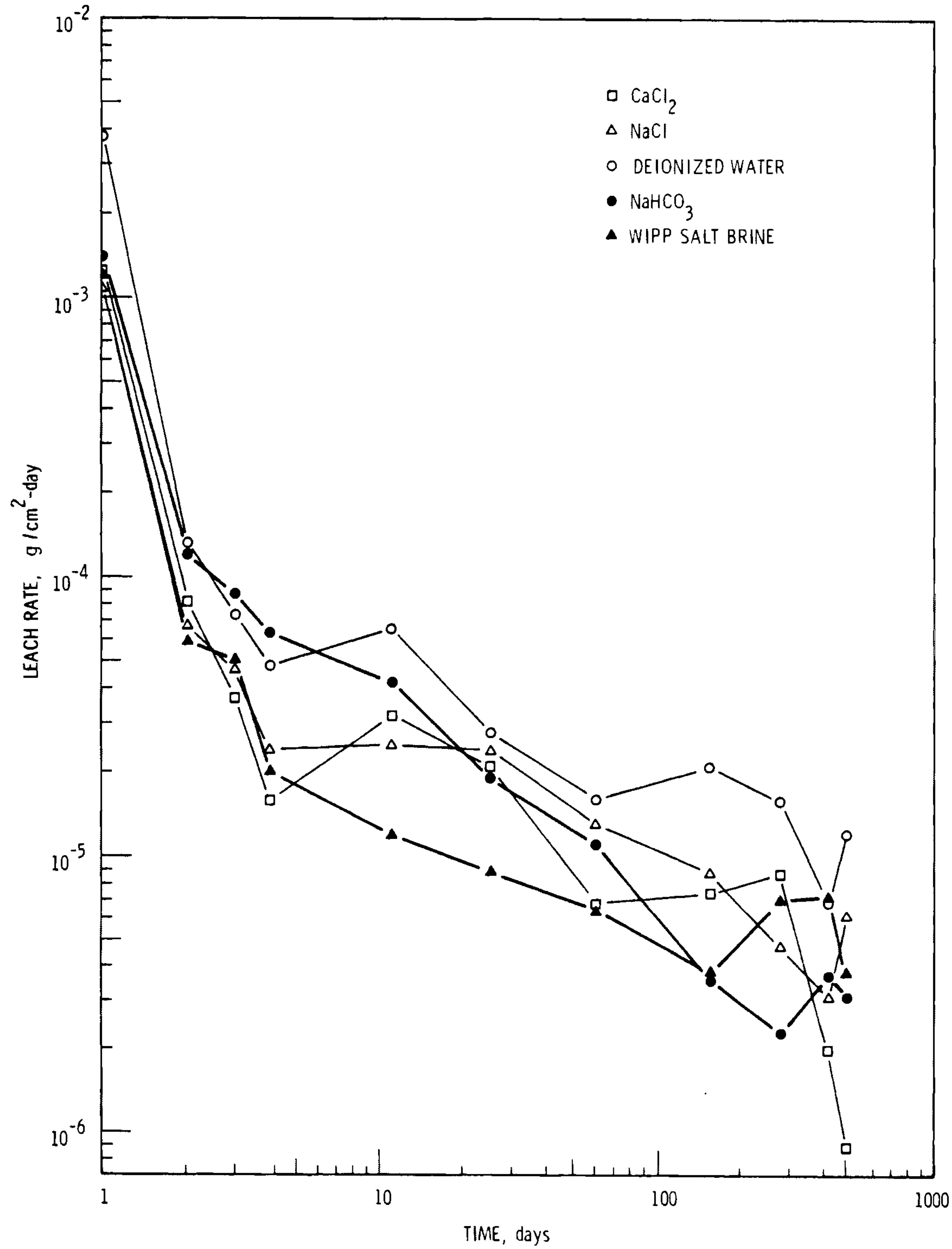

FIGURE 9. Leach Rate of 28,000 MWd/MTU Spent Fuel at 25°C Based on Release of ^{137}Cs

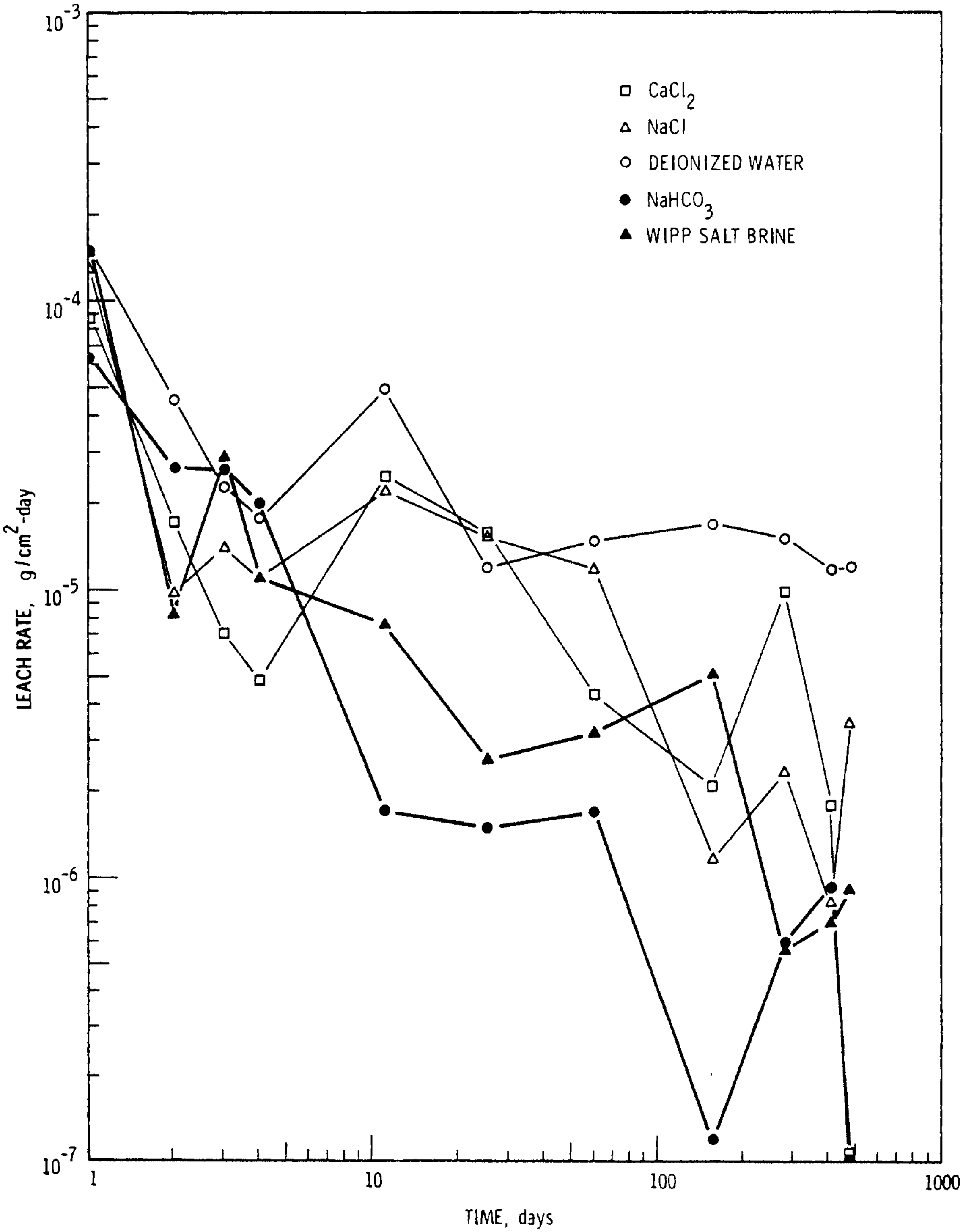

FIGURE 10. Leach Rate of 28,000 MWd/MTU Spent Fuel at 25°C Based on Release of ^{144}Ce

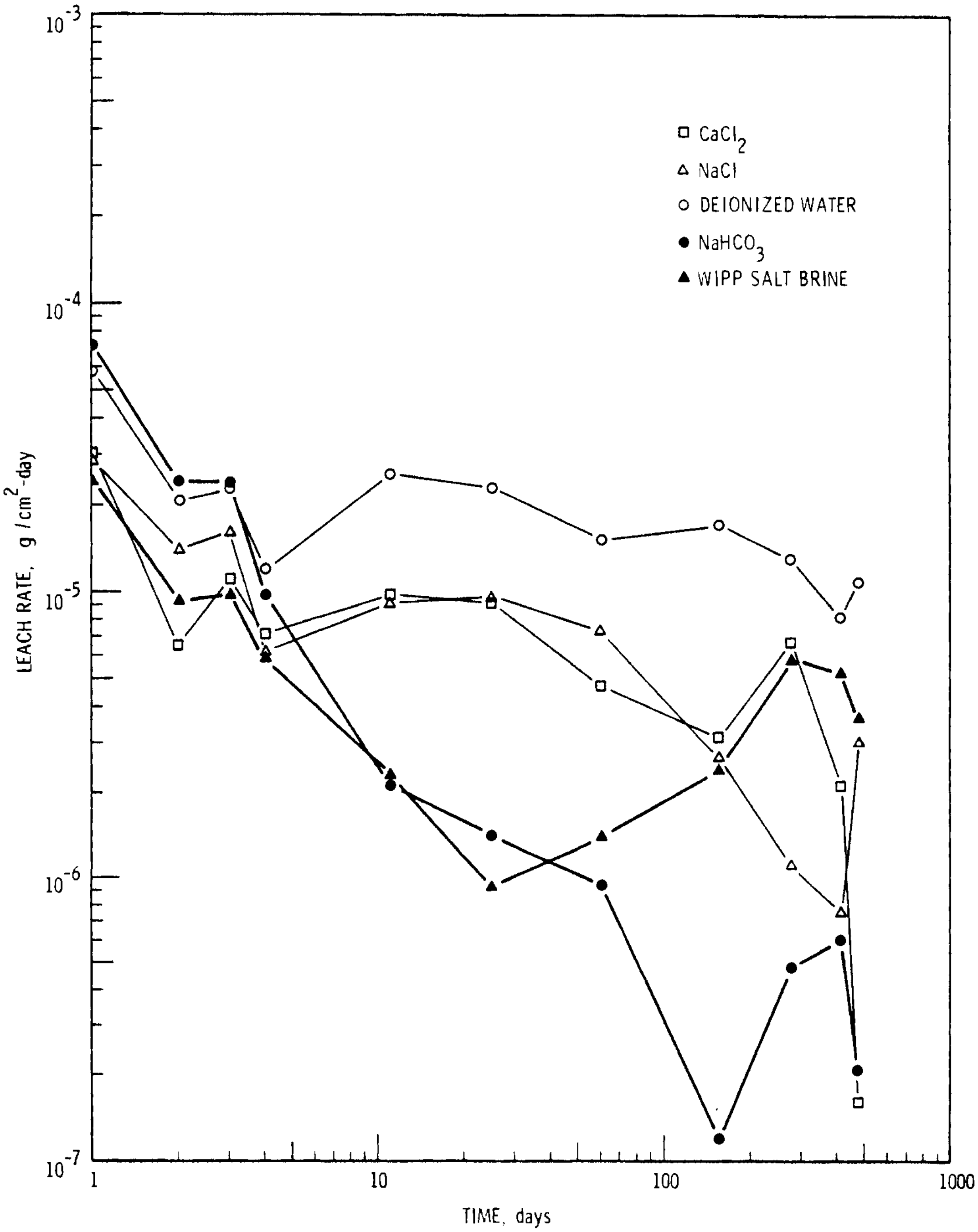

FIGURE 11. Leach Rate of 28,000 MWd/MTU Spent Fuel at 25°C Based on Release of $^{239+240}Pu$

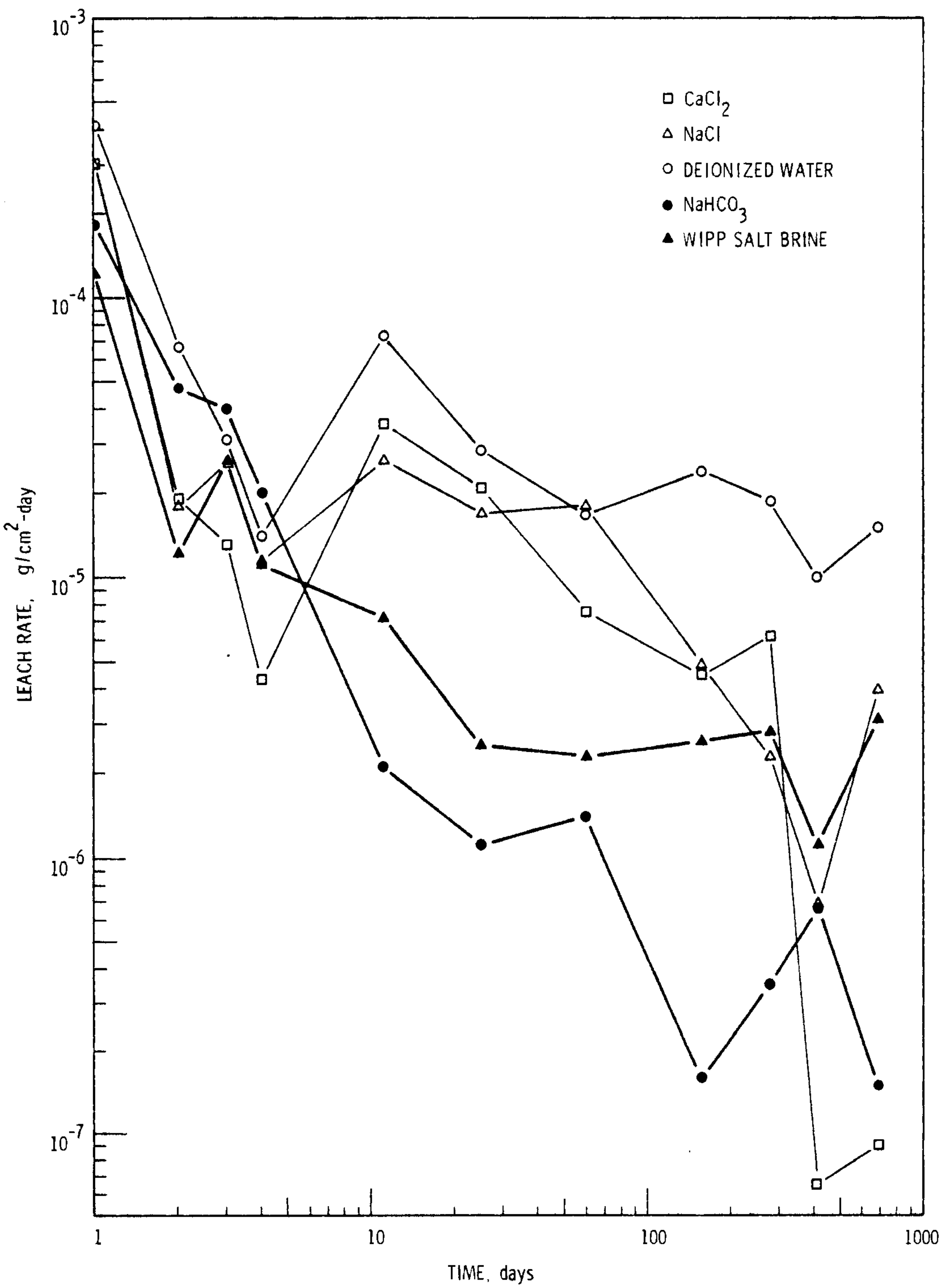

FIGURE 12. Leach Rate of 28,000 MWd/MTU Spent Fuel at 25°C Based on Release of ^{244}Cm

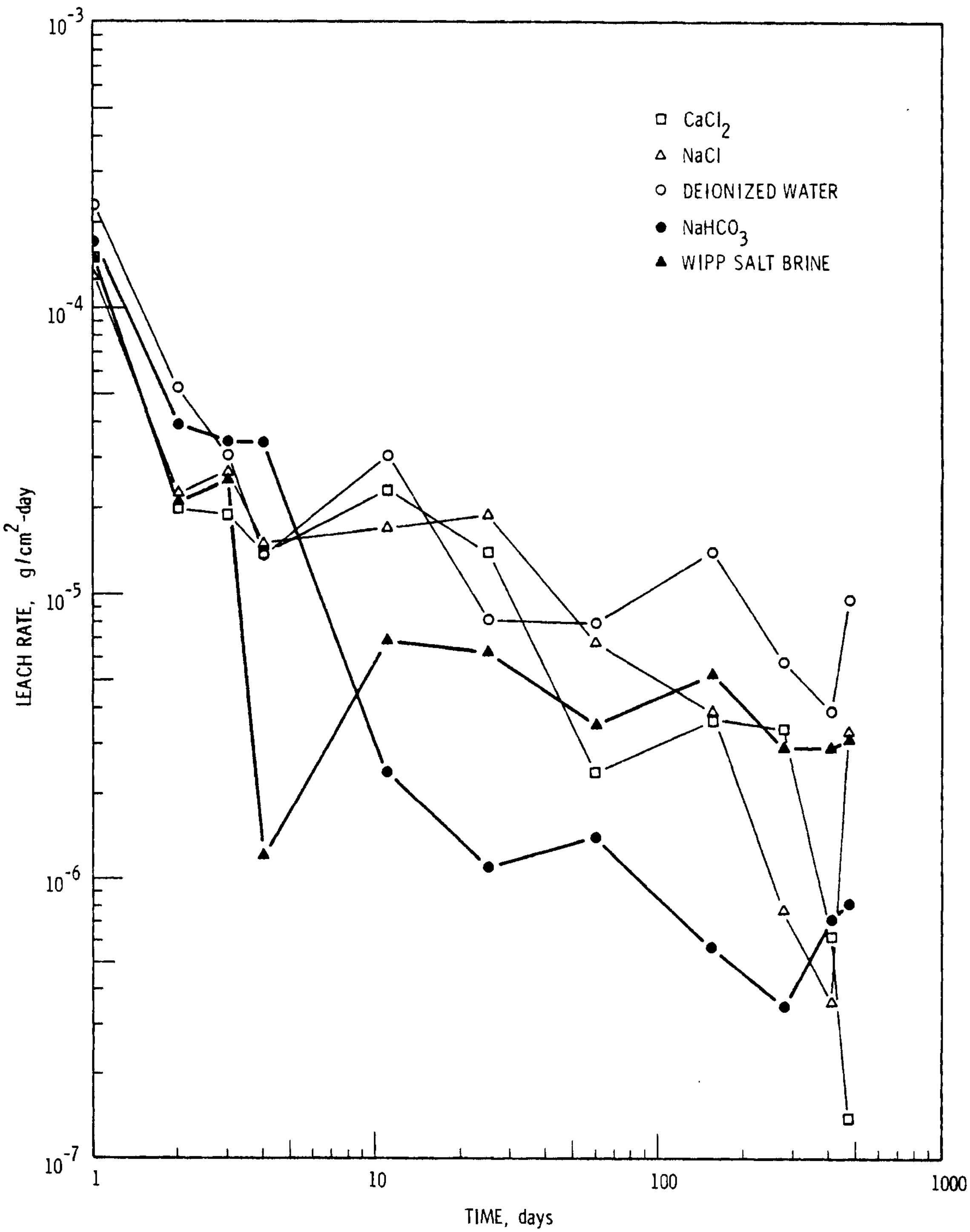

FIGURE 13. Leach Rate of 28,000 MWd/MTU Spent Fuel at 25°C Based on Release of ^{90}Sr + ^{90}Y

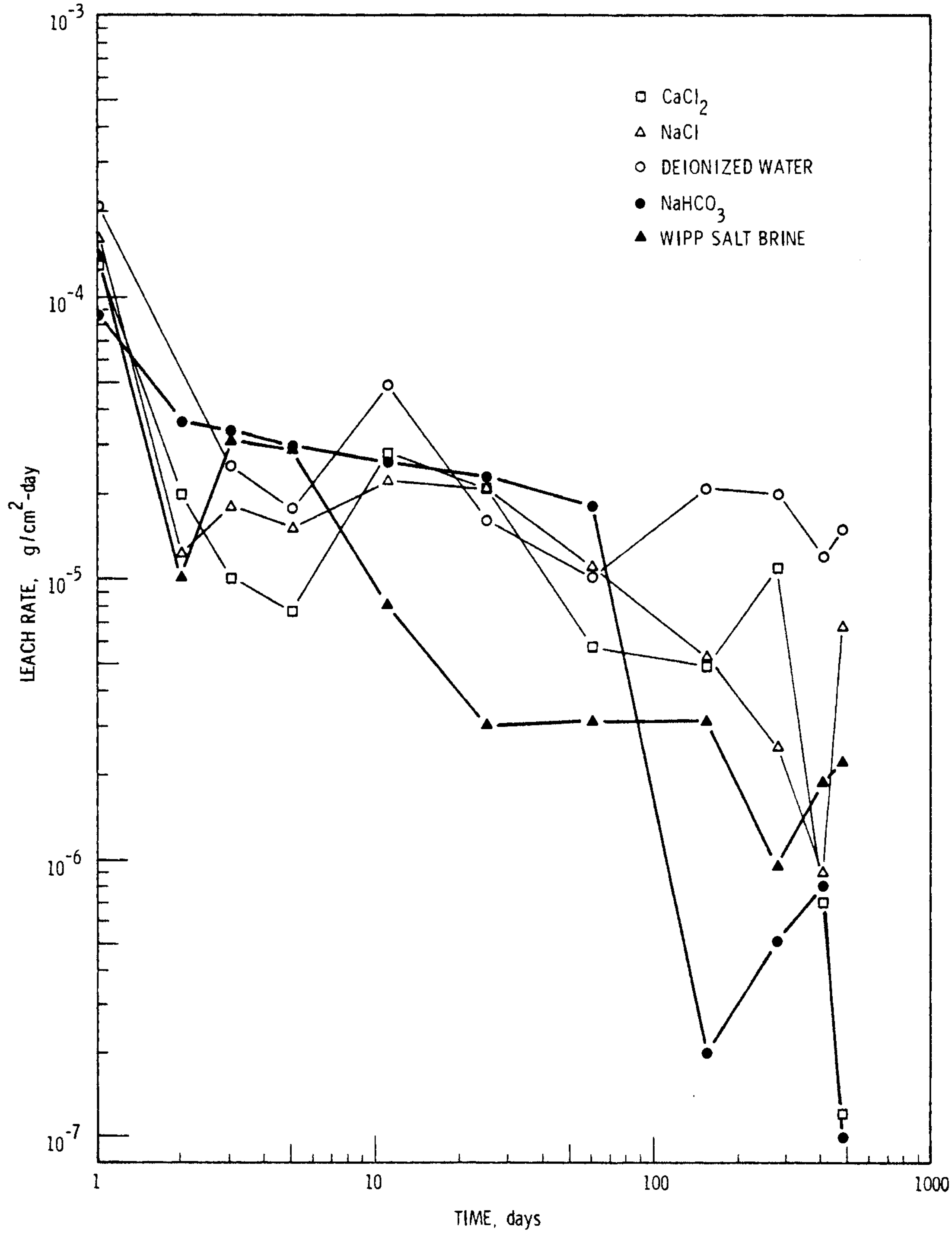

FIGURE 14. Leach Rate of 28,000 MWd/MTU Spent Fuel at 25°C Based on Release of ^{154}Eu

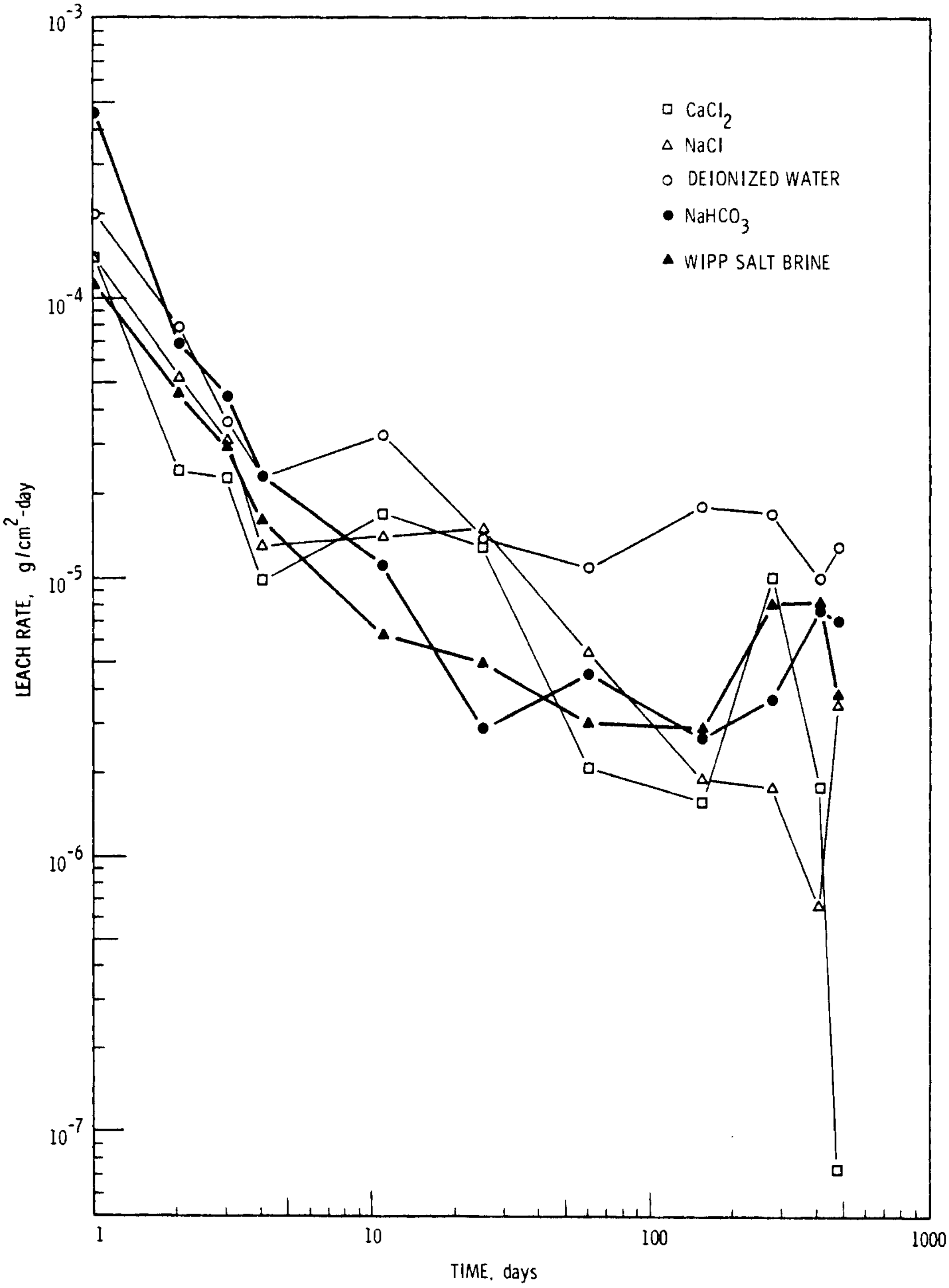

FIGURE 15. Leach Rate of 28,000 MWd/MTU Spent Fuel at 25°C Based on Release of ^{125}Sb

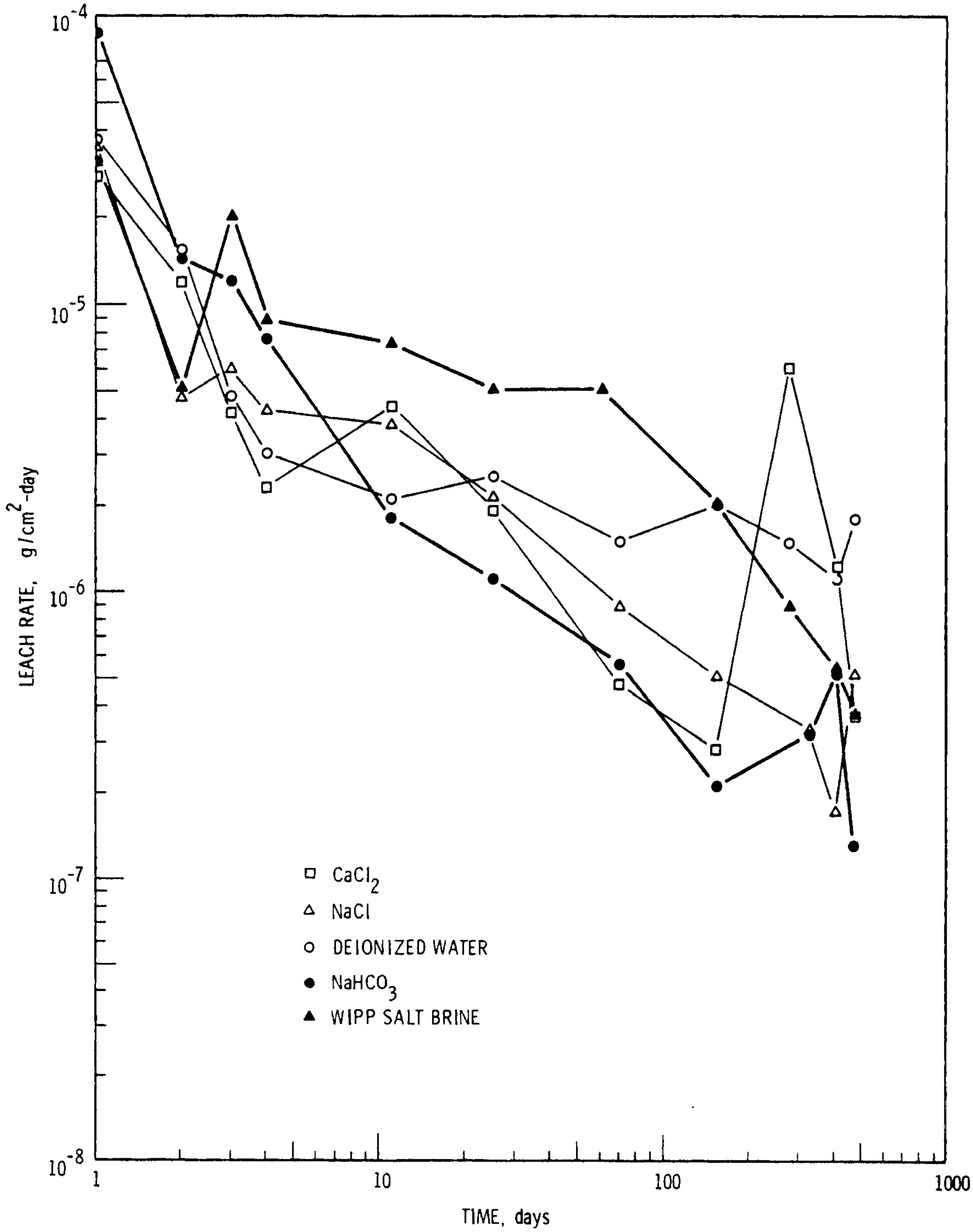

FIGURE 16. Leach Rate of 28,000 MWd/MTU Spent Fuel at 25°C Based on Release of ^{106}Ru

TABLE 4. Results of Effects of Solution Type on Each Element (Figures 8 through 16)

Element	Observed Ranking of Solutions From Highest to Lowest Element Release (467 days)
Uranium	DiW,[a] $NaHCO_3$, NaCl, WIPP, $CaCl_2$
^{137}Cs	DiW, NaCl, WIPP, $NaHCO_3$, $CaCl_2$
^{144}Ce	DiW, NaCl, WIPP, $CaCl_2$, $NaHCO_3$
$240 + {}^{239}Pu$	DiW, WIPP, NaCl, $NaHCO_3$, $CaCl_2$
^{244}Cm	DiW, NaCl, WIPP, $NaHCO_3$, $CaCl_2$
$^{90}Sr + {}^{90}Y$	DiW, NaCl, WIPP, $NaHCO_3$, $CaCl_2$
^{154}Eu	DiW, NaCl, WIPP, $CaCl_2$, $NaHCO_3$
^{125}Sb	DiW, $NaHCO_3$, WIPP, NaCl, $CaCl_2$
^{106}Ru	DiW, NaCl, WIPP, $CaCl_2$, $NaHCO_3$

(a) DiW = deionized water.

Figure 19 is a graph of leach rate curves in a saturated WIPP "B" brine solution for the nine elements measured. The spread in the leach curves at the first day of leaching covers about 1-1/2 orders of magnitude and decreases to a spread of about one order of magnitude after 467 days of cumulative leaching. Cesium has the highest initial leach rate and continues to stay near the highest value for the 467 days. Ruthenium has the lowest leach rate for the first two days and the last 63 days during the 467 days of cumulative leaching. In between these two low periods, the ruthenium leach rate increases to become the second highest (next to cesium) at 154 days of cumulative leaching.

Figure 20 is a graph of leach rate curves in a 0.03$\underline{M}$ $NaHCO_3$ solution for the nine elements measured. The spread in the leach curves at the first day of leaching covers about 1-1/3 order of magnitude and spreads to about two orders of magnitude at 467 days of cumulative leaching. Cesium has the highest initial leach rate and remains the highest until the antimony leach rate overtakes cesium at 200 days. All the other elements are grouped near the lower portion of the spread in the leach curves.

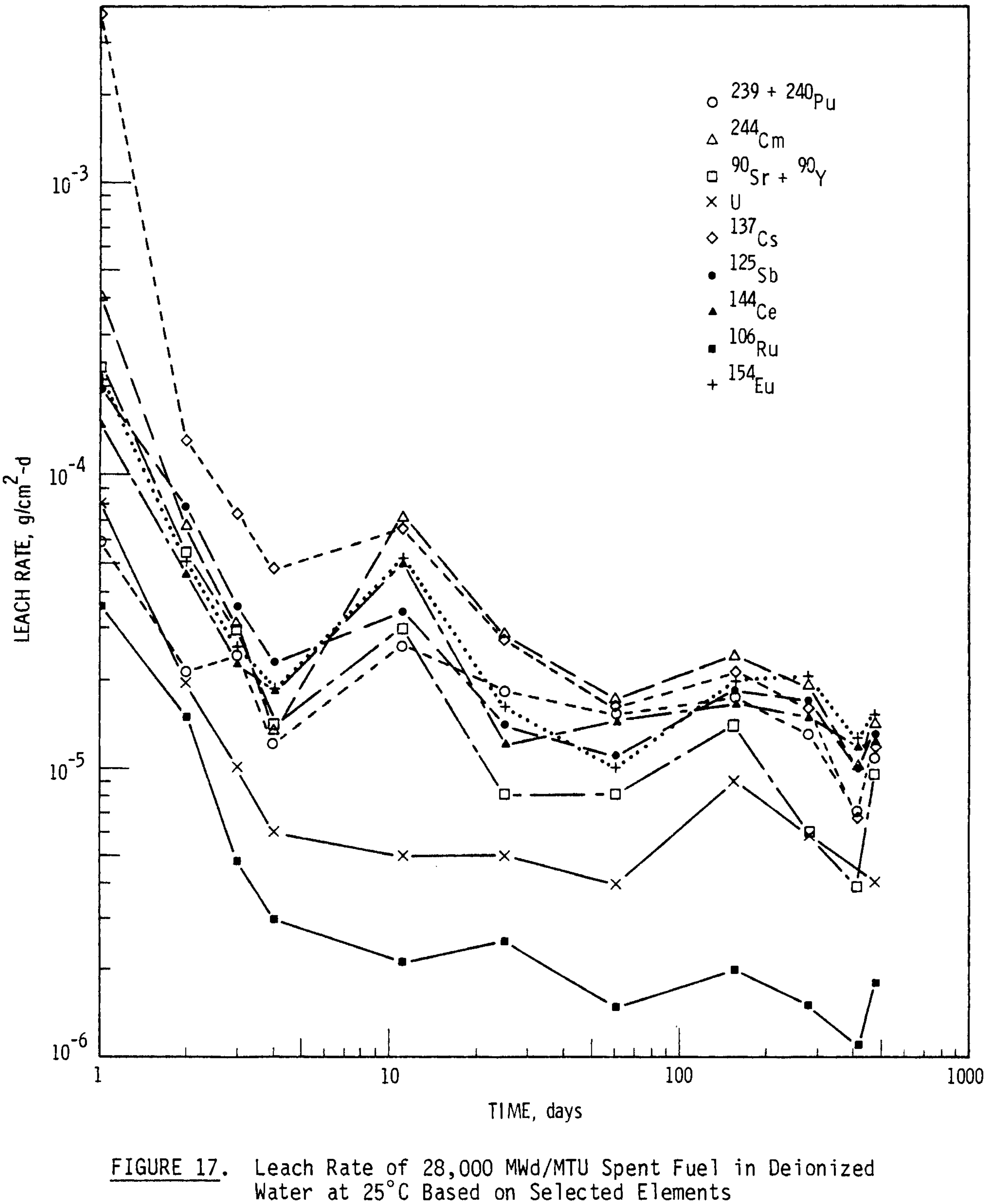

FIGURE 17. Leach Rate of 28,000 MWd/MTU Spent Fuel in Deionized Water at 25°C Based on Selected Elements

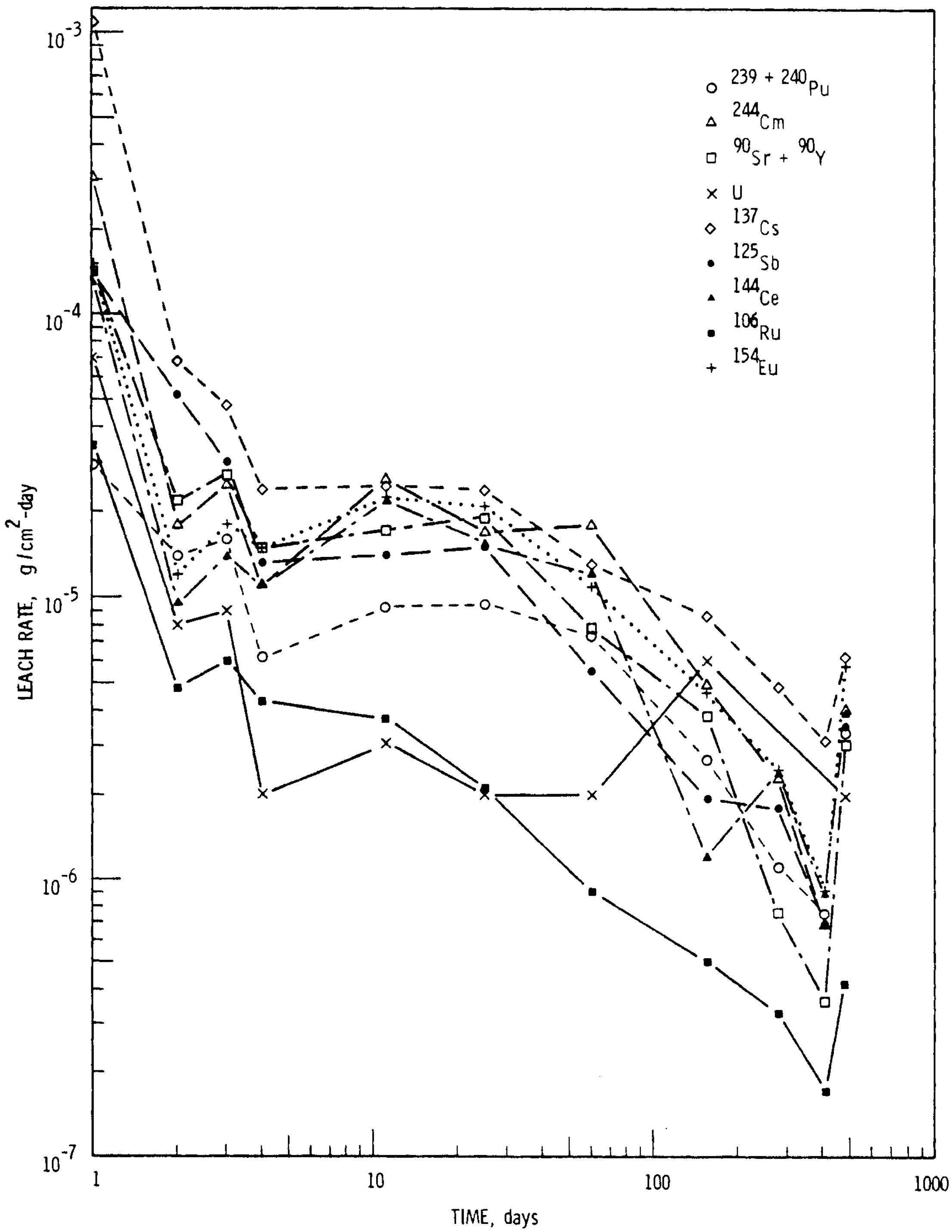

FIGURE 18. Leach Rate of 28,000 MWd/MTU Spent Fuel in 0.03M NaCl at 25°C Based on Selected Elements

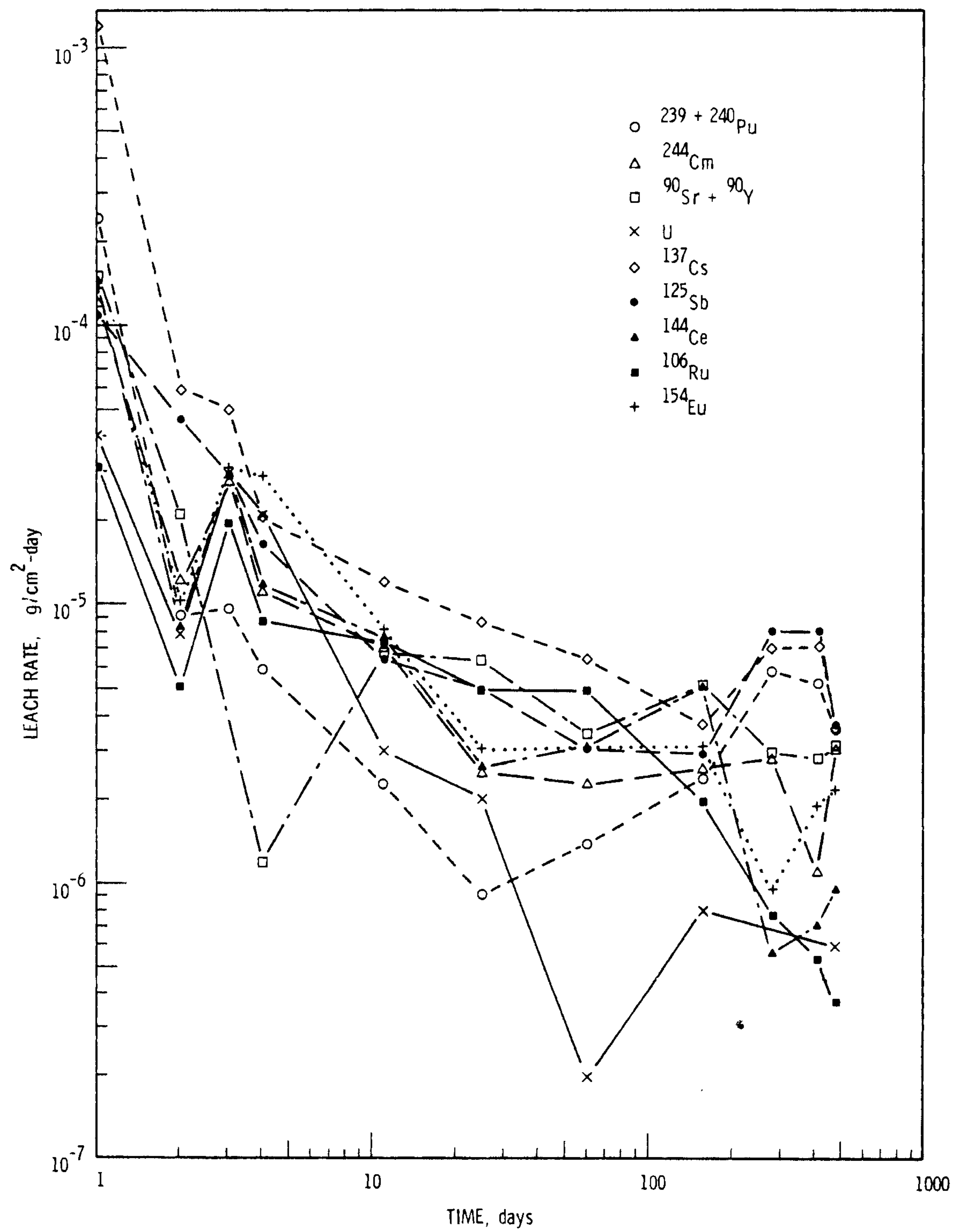

FIGURE 19. Leach Rate of 28,000 MWd/MTU Spent Fuel in WIPP "B" Saturated Brine Solution at 25°C Based on Selected Elements

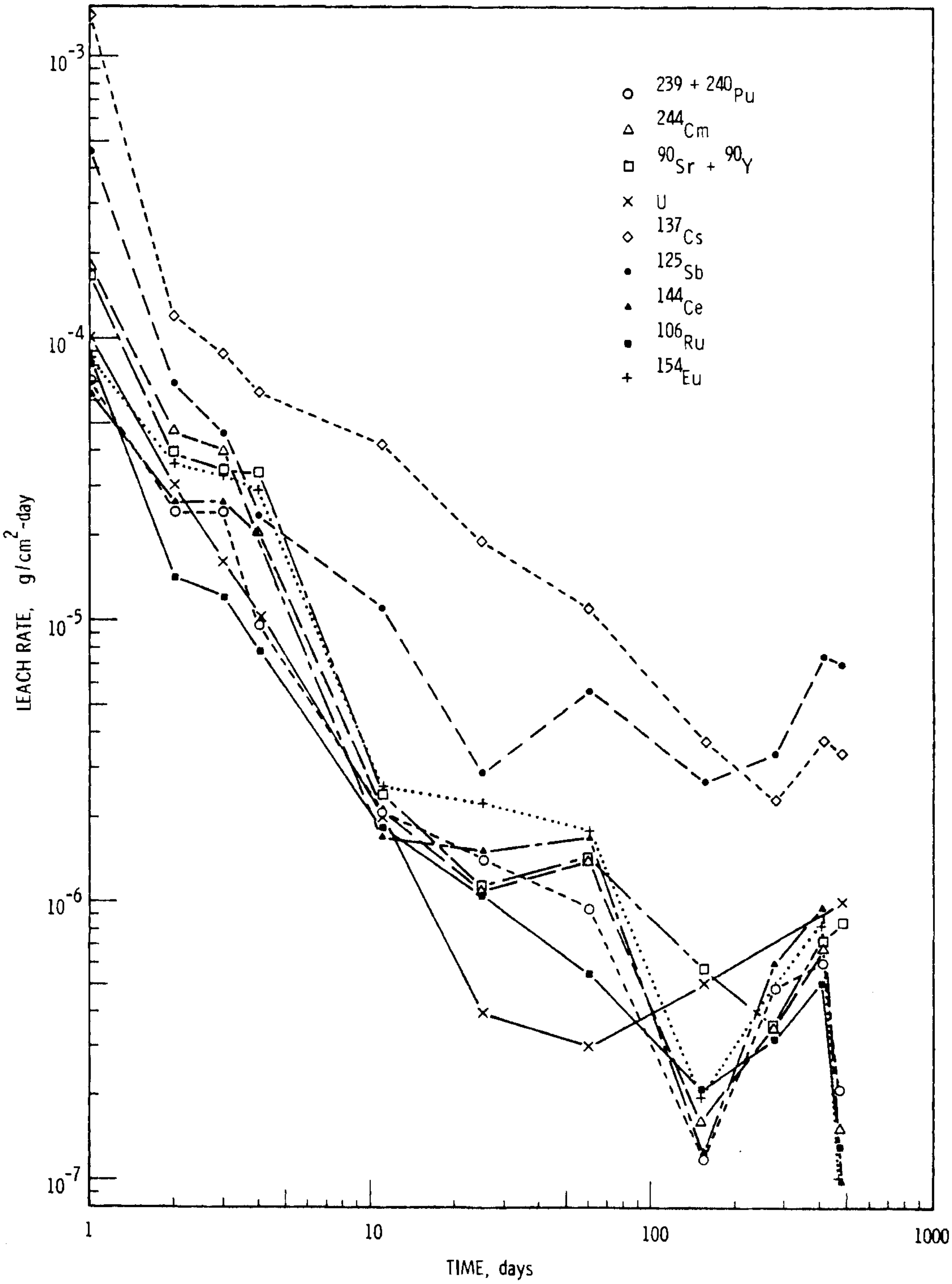

FIGURE 20. Leach Rate of 28,000 MWd/MTU Spent Fuel in 0.03M $NaHCO_3$ at 25°C Based on Selected Elements

Figure 21 is a graph of leach rate curves in a 0.015$\underline{M}$ $CaCl_2$ solution for the nine elements measured. The spread in the leach curves at the first day of leaching covers about 1-1/2 orders of magnitude, and after 467 days of cumulative leaching the spread is still about 1-1/2 orders of magnitude. After the first week of leaching, the uranium and ruthenium leach curves tend to follow the lower bounds of the spread in the leach curves and the other elements tend to follow the upper portion of the spread.

The elemental leach rate curves for the five leach solutions start at similar values, but the general trends with cumulative time of leaching are lower for the NaCl, WIPP "B" brine. $NaHCO_3$ and $CaCl_2$ solutions than for deionized water. For comparison purposes, if we choose the midpoint of the spread in elemental leach curves after 467 days of cumulative leaching as the average leaching value for that solution, then the relative leachability of the spent fuel in the five leach solutions can be calculated. Table 5 lists the midpoint of the spread in leach curves after 467 days, where the relative leachability value is given with deionized water as a reference of 1.00. In WIPP "B" brine solution the leach rate of spent fuel is only 20% of the value in deionized water. The leachability of spent fuel in 0.015$\underline{M}$ $CaCl_2$ solution is only 4% of the rate in deionized water.

In Table 6 the observed ranking of element release from highest to lowest is tabulated based on the incremental leach rate status at 467 days. Cesium is the element with the highest release rate in WIPP "B" brine, 0.015$\underline{M}$ $CaCl_2$ and 0.03$\underline{M}$ NaCl. It has the second highest release rate in 0.03$\underline{M}$ $NaHCO_3$ and ranks fifth in deionized water. Ruthenium is the lowest for all leachants. Uranium and cerium are also low-release elements in all the leachants.

Spent-fuel leach rates measured by the Paige procedure were found to be comparable to the first-generation borosilicate glasses vitrified during the Waste Solidification Engineering Prototype demonstration at PNL (Katayama 1976). Comparisons of spent-fuel and glass leach rates using the IAEA procedures are tabulated in Table 7. The ^{244}Cm and $^{239+240}$Pu based glass leach rates are for a second-generation borosilicate glass coded 76-68 (Bradley, Harvey and Turcotte 1979). In the reference, deionized water leach solution, the spent-fuel leach rates for the actinides plutonium and curium were 50 to

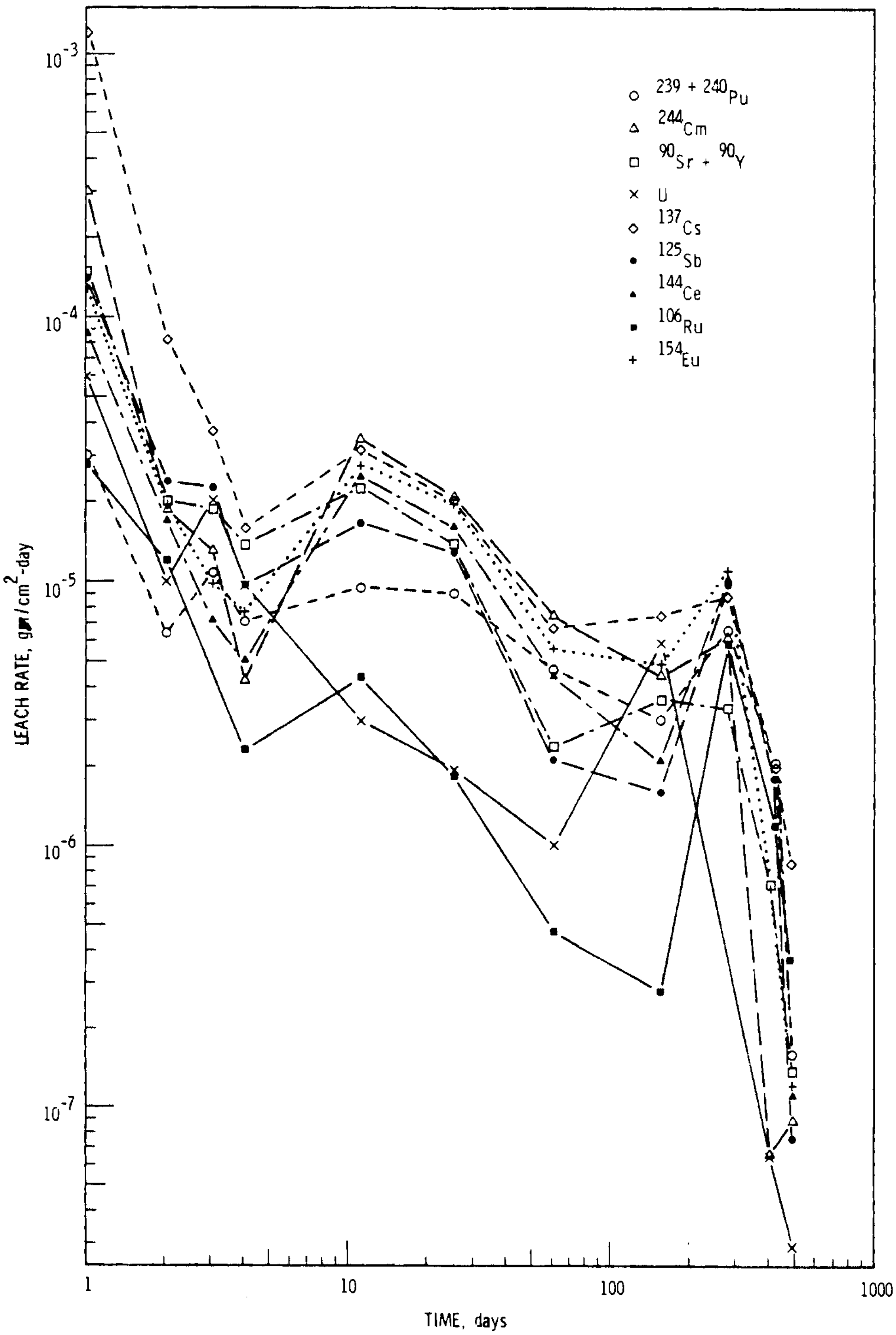

FIGURE 21. Leach Rate of 28,000 MWd/MTU Spent Fuel in 0.015M $CaCl_2$ Solution at 25°C Based on Selected Elements

TABLE 5. Relative Leachability of Spent Fuel in Various Leach Solutions Based on Average Trend of Leach Curves After 467 Days of Cumulative Leaching

Solution	Midpoint of Leach Curves at 467 Days, g/cm^2-day	Relative Leachability
Deionized Water	5×10^{-6}	1.00
0.03M NaCl	2×10^{-6}	0.40
Saturated WIPP "B"	1×10^{-6}	0.20
0.03M $NaHCO_3$	8×10^{-7}	0.16
0.015M $CaCl_2$	2×10^{-7}	0.04

TABLE 6. Results of Comparing Incremental Leach Rates of Elements for Each Solution Type (Figures 17 through 21)

Solution	Observed Ranking of Element Release From Highest to Lowest (467 Days)
Deionized Water	Eu, Cm, Sb, Ce, Cs, Pu, Sr+Y, U, Ru
WIPP "B"	Cs, Sb, Pu, Sr+Y, Cm, Eu, Ce, U, Ru
$NaHCO_3$	Sb, Cs, U, Sr+Y, Pu, Cm, Ru, Eu, Ce
NaCl	Cs, Eu, Cm, Ce, Sb, Pu, Sr+Y, U, Ru
$CaCl_2$	Cs, Ru, Pu, Sr+Y, Eu, Ce, Cm, Sb, U

TABLE 7. Comparison of IAEA Leach Rates at 25°C

Solution	$^{240+239}Pu$ Leach Rates(a), g/cm^2-day		^{244}Cm Leach Rates(b), g/cm^2-day	
	76-68 Glass	Spent Fuel	76-68 Glass	Spent Fuel
Deionized Water	5×10^{-8}	2×10^{-5}	4×10^{-7}	2×10^{-5}
WIPP "B" Brine	2 x 10-8	2×10^{-6}	1×10^{-8}	3×10^{-6}
NaCl	7×10^{-8}	3×10^{-6}	2×10^{-7}	4×10^{-6}
$CaCl_2$	2×10^{-8}	3×10^{-6}	1×10^{-7}	9×10^{-8}
$NaHCO_3$	2×10^{-7}	1×10^{-7}	2×10^{-7}	2×10^{-7}

(a) 151 days.
(b) 454 days.

400 times higher than that of glass. The differences decrease as tests progress from deionized water to the WIPP "B" Brine to the 0.03M NaCl solution to the 0.015M $CaCl_2$ solution, and nearly disappear in 0.03M $NaHCO_3$ solution.

The IAEA leach rates are higher than the published Paige-test leach rates (Katayama 1979) for the same spent fuel after 467 days of cumulative leaching. This comparison is given in Table 8. The IAEA test results are about one order in magnitude higher than the Paige-test results. In WIPP "B" brine (287 g/L NaCl) and sodium chloride groundwater (1.76 g/L NaCl), the IAEA cesium-based leach rates are 4×10^{-6} and 6×10^{-6} g/cm^2-day, respectively, whereas the Paige cesium-based leach rate in sea brine (28 g/L NaCl) is 6×10^{-7} g/cm^2-day. These differences may be due to radionuclide plate-out on the Paige apparatus, which can not be measured.

The pH of the deionized water used in the experiment averaged 6.6, and the pH at the end of the incremental leach period averaged 4.3. The net result was a decrease in the pH of the deionized water during the incremental leach test period which averaged 2.3 pH units. This decrease in the pH value is the opposite of that reported by Grandstaff (1976) for the leaching of uraninite. Such a difference may be due to the difference of chemistry between the spent LWR fuel and the uraninite. Decreases in pH were also found in the WIPP "B", 0.015M $CaCl_2$ and 0.03M NaCl solutions. Actinide solid-induced radiolysis of similar synthetic groundwater solutions have been observed to cause similar pH drops (Rai et al. in press). However, increases in pH were found for the 0.03M $NaHCO_3$ solution.

There is a subtask in the WRIT program at PNL entitled "Spent-Fuel Special Studies." The major thrust of this task is directed toward understanding the mechanisms and kinetics for spent fuel release in aqueous solutions. The non-uniform release of uranium to the leach solution (see Figure 11), the effects of oxygen, and solubility constraints are being investigated. The progress of these experiments will be presented in the next status report.

The leach rate data presented in this report show that uranium has a lower leach rate than most of the elements studied. This indicates the absence of congruent dissolution. Preliminary results from our electrochemical experiments (part of the Spent-Fuel Special Studies Subtask within WRIT) with fused,

TABLE 8. Comparison of IAEA Leach Rates to Paige-Test Leach Rates (467 days)

Solution	Element	Leach Rates (g/cm^2-day) at 25°C Paige	IAEA
Deionized water	^{137}Cs	2×10^{-6}	1×10^{-5}
Deionized water	$^{239+240}Pu$	9×10^{-7}	1×10^{-5}
Deionized water	Uranium	7×10^{-7}	4×10^{-6}
Deionized water	^{244}Cm	1×10^{-7}	4×10^{-6}
NaCl groundwater	^{137}Cs	--	6×10^{-6}
Sea brine	^{137}Cs	6×10^{-7}	--
WIPP "B"	^{137}Cs	--	4×10^{-6}

single crystal UO_2 indicates that a film forms on the surface of the UO_2 in the standard WRIT leach solutions (those used in this report). If such a hydrolyzed, gelatinous, uranium oxide film forms on the surface of the spent fuel, the uranium release to the leach solution may be lower than the other radionuclides that may pass through this film. The fluctuations in the uranium leach rates presented in this report do not relate to the leach rate trends of the other elements studied. This incongruency in the leach rates lends support to the possibility of a hydrolyzed uranium oxide film forming on our spent-fuel samples. Thus, the observed uranium leach rate may be dependent on the formation, dissolution and partial spallation of such a film.

CONCLUSIONS

The leach rates of antimony, cerium, cesium, curium, europium, plutonium, ruthenium, strontium + yttrium, and uranium were determined from LWR spent fuel (unclad) with a burnup of 28,000 MWd/MTU in five different leach solutions. These solutions were deionized water, 0.03M sodium chloride solutions, 0.03M sodium bicarbonate solution, 0.015M calcium chloride solution and a saturated WIPP "B" brine solution. On the average, of the five solutions studied deionized water produced the highest elemental leach rate. The leach rates in the bicarbonate solution were second lowest, with the $CaCl_2$ solution exhibiting the lowest leach rates. The two brines were intermediate.

Cesium had the highest leach rate in three solutions (WIPP "B" brine, 0.03M NaCl and 1.015M $CaCl_2$). Europium had the highest leach rate in deionized water and antimony the highest in 0.03M $NaHCO_3$. Ruthenium had the lowest leach rate in three solutions (deionized water, 0.03M NaCl and WIPP "B" brine). Cerium had the lowest elemental leach rate in 0.015M $NaHCO_3$ and uranium the lowest in 0.03M $CaCl_2$ solution.

Based on the release of plutonium and curium, spent fuel has a leach rate comparable to 76-68 glass in sodium bicarbonate solution and is 50 to 400 times less leach resistant than 76-68 glass in deionized water and WIPP "B" brine solution.

The spent-fuel leach rates calculated from the IAEA procedure are one order of magnitude higher than the values reported for the Paige procedure. The discrepancy may be caused by nuclide plate-out on the walls of the Paige apparatus, which can not be measured during testing. Thus, to be conservative one should perform leaching tests by the modified IAEA procedure rather than the Paige procedure.

The leach rate data in this status report show that spent fuel may not leach congruently with uranium.

REFERENCES

Bradley, D. J., C. O. Harvey and R. P. Turcotte. 1979. Leaching of Actinides and Technetium from Simulated High-Level Waste Glass. PNL-3152, Battelle, Pacific Northwest Laboratory, Richland, Washington.

Centanni, Ross A. M. and M. A. DeSesa. 1956. "Fluorimetric Determination of Uranium." Anal. Cheml. 28:1651-1657.

Dosch, R. G., and A. W. Lynch. 1978. Interaction of Radionuclides with Geomedia Associated with the Waste Isolation Pilot Plant (WIPP) Site in New Mexico. SAND 78-0297, Sandia Laboratory, Albuquerque, New Mexico.

Finston, H. L. 1961. "The Radiochemistry of Cesium." Nuclear Science Series. No. 5-NA3085.

Grandstaff, D. E. 1976. "A Kinetic Study of the Dissolution of Uraninite." Econ. Geol. 71:1493-1506.

Katayama, Y. B. 1976. Leaching of Irradiated LWR Fuel Pellets in Deionized and Typical Groundwater. BNWL-2057, Pacific Northwest Laboratories, Richland, Washington.

Katayama, Y. B. 1979. Spent LWR Fuel Leach Test. PNL-2982, Battelle, Pacific Northwest Laboratory, Richland, Washington.

Koltoff, I. M., and R. J. Elving. 1966. Treatise of Anaytical Chemistry, Part I, Vol. 4. Interscience Publishers, Inc., New York, New York.

Mendel, J. E. 1973. A Review of Leaching Test Methods and the Leachability of Various Solids Media Containing Radioactive Wastes. BNWL-1765, Pacific Northwest Laboratories, Richland, Washington.

Moore, K. L., and J. E. Hudgens, Jr. 1957. "Separation and Determination of Plutonium by Liquid-Liquid Extraction." Anal. Chem. 29:1767.

Price, G. R., G. R. Perretti and S. Swaitz. 1953. "Fluorophotometric Determination of Uranium." Anal. Chem. 25(2):322-331.

Rai, Dhanpat et al., in press. "Solubility of Plutonium Compounds and Their Behavior in Soils," Soil Sci. Soc. Ann. J.

APPENDIX

LEACH DATA

IAEA LEACH RATE BASED ON ^{244}Cm (g/cm^2-day)
FOR 28,000 MWd/MTU SPENT FUEL AT 25°C

Series	Cumulative Days	WIPP Brine	$CaCl_2$ Solution	NaCl Solution	$NaHCO_3$ Solution	Deionized Water
1	1	1.2 E-4	3.0 E-4	3.0 E-4	1.8 E-4	4.1 E-4
2	2	1.2 E-5	1.9 E-5	1.8 E-5	4.7 E-5	6.7 E-5
3	3	2.6 E-5	1.3 E-5	2.5 E-5	4.0 E-5	3.1 E-5
4	4	1.1 E-5	4.3 E-6	1.1 E-5	2.0 E-5	1.4 E-5
5	11	7.1 E-6	3.5 E-5	2.6 E-5	2.1 E-6	7.2 E-5
7	25	2.5 E-6	2.1 E-5	1.7 E-5	1.1 E-6	2.8 E-5
12	60	2.3 E-6	7.6 E-6	1.8 E-5	1.4 E-6	1.7 E-5
15	154	2.6 E-6	4.5 E-6	4.9 E-6	1.6 E-7	2.4 E-5
19	277	2.8 E-6	6.2 E-6	2.3 E-6	3.5 E-7	1.9 E-5
23	404	1.1 E-6	6.7 E-8	6.8 E-7	6.8 E-7	1.0 E-5
25	467	3.1 E-6	9.1 E-8	4.0 E-6	1.5 E-7	1.5 E-5

IAEA LEACH RATE BASED ON U (g/cm^2-day)
FOR 28,000 MWd/MTU SPENT FUEL AT 25°C

Series	Cumulative Days	WIPP Brine	$CaCl_2$ Solution	NaCl Solution	$NaHCO_3$ Solution	Deionized Water
1	1	4E-5	6E-5	7E-5	1E-4	8E-5
2	2	8E-6	1E-5	8E-6	3E-5	2E-5
3	3	3E-5	2E-5	9E-6	4E-5	1E-5
4	4	2E-5	1E-5	2E-6	1E-5	6E-6
5	11	3E-6	3E-6	3E-6	2E-6	5E-6
7	25	2E-6	2E-6	2E-6	4E-7	4E-6
12	60	2E-7	1E-6	2E-6	3E-7	4E-6
15	154	8E-7	6E-6	5E-6	5E-7	9E-6
19	277	(a)	(a)	(a)	(a)	(a)
23	404	(a)	(a)	(a)	(a)	(a)
25	467	6E-7	3E-8	2E-6	1E-6	4E-6

(a) analysis not availble at time of reporting.

IAEA LEACH RATE BASED ON ^{125}Sb (g/cm^2-day)
FOR 28,000 MWd/MTU SPENT FUEL AT 25°C

Series	Cumulative Days	WIPP Brine	$CaCl_2$ Solution	NaCl Solution	$NaHCO_3$ Solution	Deionized Water
1	1	1.1 E-4	1.4 E-4	1.4 E-4	4.6 E-4	2.0 E-4
2	2	4.6 E-5	2.4 E-5	5.1 E-5	6.9 E-5	7.9 E-5
3	3	2.9 E-5	2.3 E-5	3.0 E-5	4.5 E-5	3.6 E-5
4	4	1.6 E-5	9.8 E-6	1.3 E-5	2.3 E-5	2.3 E-5
5	11	6.2 E-6	1.7 E-5	1.4 E-5	1.1 E-5	3.4 E-5
7	25	5.0 E-6	1.3 E-5	1.5 E-5	2.9 E-6	1.4 E-5
12	60	3.0 E-6	2.1 E-6	5.4 E-6	5.6 E-6	1.1 E-5
15	154	2.9 E-6	1.6 E-6	1.9 E-6	2.7 E-6	1.8 E-5
19	277	8.1 E-6	1.0 E-5	1.8 E-6	3.7 E-6	1.7 E-5
23	404	8.2 E-6	1.8 E-6	6.7 E-7	7.7 E-6	1.0 E-5
25	467	3.8 E-6	7.6 E-8	3.5 E-6	7.0 E-6	1.3 E-5

IAEA LEACH RATE BASED ON ^{144}Ce (g/cm^2-day)
FOR 28,000 MWd/MTU SPENT FUEL AT 25°C

Series	Cumulative Days	WIPP Brine	$CaCl_2$ Solution	NaCl Solution	$NaHCO_3$ Solution	Deionized Water
1	1	1.5 E-4	8.8 E-5	1.3 E-4	6.3 E-5	1.5 E-4
2	2	8.2 E-6	1.7 E-5	9.6 E-6	2.6 E-5	4.6 E-5
3	3	2.9 E-5	7.1 E-6	1.4 E-5	2.6 E-5	2.3 E-5
4	4	1.1 E-5	4.9 E-6	1.1 E-5	2.0 E-5	1.8 E-5
5	11	7.7 E-6	2.5 E-5	2.2 E-5	1.7 E-6	5.0 E-5
7	25	2.6 E-6	1.6 E-5	1.5 E-5	1.5 E-6	1.2 E-5
12	60	3.2 E-6	4.4 E-6	1.2 E-5	1.7 E-6	1.5 E-5
15	154	5.1 E-6	2.1 E-6	1.2 E-6	1.2 E-7	1.7 E-5
19	277	5.6 E-7	1.0 E-5	2.4 E-6	6.0 E-7	1.5 E-5
23	404	7.0 E-7	1.8 E-6	8.4 E-7	9.7 E-7	1.2 E-5
25	467	9.4 E-7	1.1 E-7	3.5 E-6	1.0 E-7	1.2 E-5

IAEA LEACH RATE BASED ON $^{90}Sr + ^{90}Y$ (g/cm^2-day)
FOR 28,000 MWd/MTU SPENT FUEL AT 25°C

Series	Cumulative Days	WIPP Brine	$CaCl_2$ Solution	NaCl Solution	$NaHCO_3$ Solution	Deionized Water
1	1	1.5 E-4	1.5 E-4	1.4 E-4	1.7 E-4	2.3 E-4
2	2	2.1 E-5	2.0 E-5	2.2 E-5	3.9 E-5	5.4 E-5
3	3	2.5 E-5	1.9 E-5	2.7 E-5	3.4 E-5	3.1 E-5
4	4	1.2 E-6	1.4 E-5	1.5 E-5	3.4 E-5	1.4 E-5
5	11	6.8 E-6	2.3 E-5	1.7 E-5	2.4 E-6	3.1 E-5
7	25	6.3 E-6	1.4 E-5	1.9 E-5	1.1 E-6	8.1 E-6
12	60	3.5 E-6	2.4 E-6	6.8 E-6	1.4 E-6	8.1 E-6
15	154	5.2 E-6	3.6 E-6	3.9 E-6	5.7 E-7	1.4 E-5
19	277	2.8 E-6	3.4 E-6	7.6 E-7	3.5 E-7	5.8 E-6
23	404	2.8 E-6	7.2 E-7	3.6 E-7	7.1 E-7	3.9 E-6
25	467	3.1 E-6	1.4 E-7	3.3 E-6	8.2 E-7	9.6 E-6

IAEA LEACH RATE BASED ON ^{137}Cs (g/cm^2-day)
FOR 28,000 MWd/MTU SPENT FUEL AT 25°C

Series	Cumulative Days	WIPP Brine	$CaCl_2$ Solution	NaCl Solution	$NaHCO_3$ Solution	Deionized Water
1	1	1.2 E-3	1.2 E-3	1.1 E-3	1.4 E-3	3.8 E-3
2	2	5.9 E-5	8.1 E-5	6.8 E-5	1.2 E-4	1.3 E-4
3	3	5.1 E-5	3.8 E-5	4.8 E-5	8.9 E-5	7.5 E-5
4	4	2.0 E-5	1.6 E-5	2.4 E-5	6.4 E-5	4.8 E-5
5	11	1.2 E-5	3.2 E-5	2.5 E-5	4.2 E-5	6.6 E-5
7	25	8.9 E-6	2.1 E-5	2.4 E-5	1.9 E-5	2.8 E-5
12	60	6.4 E-6	6.7 E-6	1.3 E-5	1.1 E-5	1.6 E-5
15	154	3.8 E-6	7.5 E-6	8.8 E-6	3.7 E-6	2.1 E-5
19	277	7.0 E-6	8.8 E-6	4.8 E-6	2.3 E-6	1.6 E-5
23	404	7.1 E-6	2.0 E-6	3.1 E-6	3.8 E-6	6.8 E-6
25	467	3.8 E-6	8.9 E-7	6.1 E-6	3.3 E-6	1.2 E-5

IAEA LEACH RATE BASED ON ^{154}Eu (g/cm^2-day)
FOR 28,000 MWd/MTU SPENT FUEL AT 25°C

Series	Cumulative Days	WIPP Brine	$CaCl_2$ Solution	NaCl Solution	$NaHCO_3$ Solution	Deionized Water
1	1	1.4 E-4	1.3 E-4	1.6 E-4	8.7 E-5	2.1 E-4
2	2	1.0 E-5	2.0 E-5	1.2 E-5	3.6 E-5	5.1 E-5
3	3	3.1 E-5	1.0 E-5	1.8 E-5	3.4 E-5	2.5 E-5
4	4	2.9 E-5	7.8 E-6	1.5 E-5	2.9 E-5	1.8 E-5
5	11	8.0 E-6	2.8 E-5	2.2 E-5	2.6 E-6	4.9 E-5
7	25	3.0 E-6	2.1 E-5	2.1 E-5	2.3 E-6	1.6 E-5
12	60	3.1 E-6	5.7 E-6	1.1 E-5	1.8 E-6	1.0 E-5
15	154	3.1 E-6	4.9 E-6	5.3 E-6	2.0 E-7	2.1 E-5
19	277	9.5 E-7	1.1 E-5	2.5 E-6	5.1 E-7	2.0 E-5
23	404	1.9 E-6	7.0 E-7	9.0 E-7	8.2 E-7	1.2 E-5
25	467	2.2 E-6	1.2 E-7	6.9 E-6	9.9 E-6	1.5 E-5

IAEA LEACH RATE BASED ON ^{239}Pu + ^{240}Pu (g/cm^2-day)
FOR 28,000 MWd/MTU SPENT FUEL AT 25°C

Series	Cumulative Days	WIPP Brine	$CaCl_2$ Solution	NaCl Solution	$NaHCO_3$ Solution	Deionized Water
1	1	2.4 E-5	3.0 E-5	2.9 E-5	7.1 E-5	5.9 E-5
2	2	9.2 E-6	6.5 E-6	1.4 E-5	2.4 E-5	2.1 E-5
3	3	9.7 E-6	1.1 E-5	1.6 E-5	2.4 E-5	2.4 E-5
4	4	5.9 E-6	7.1 E-6	6.1 E-6	9.7 E-6	1.2 E-5
5	11	2.3 E-6	9.7 E-6	9.2 E-6	2.1 E-6	2.6 E-5
7	25	9.2 E-7	9.1 E-6	9.5 E-6	1.4 E-6	2.3 E-5
12	60	1.4 E-6	4.7 E-6	7.4 E-6	9.5 E-7	1.5 E-5
15	154	2.4 E-6	3.1 E-6	2.7 E-6	1.2 E-7	1.7 E-5
19	277	5.8 E-6	6.7 E-6	1.1 E-6	4.8 E-7	1.3 E-5
23	404	5.3 E-6	2.1 E-6	7.5 E-7	6.0 E-7	8.1 E-6
25	467	3.7 E-6	1.6 E-7	3.0 E-6	2.1 E-7	1.1 E-5

IAEA LEACH RATE BASED ON ^{106}Ru (g/cm^2-day)
FOR 28,000 MWd/MTU SPENT FUEL AT 25°C

Series	Cumulative Days	WIPP Brine	$CaCl_2$ Solution	NaCl Solution	$NaHCO_3$ Solution	Deionized Water
1	1	3.1 E-5	2.8 E-5	3.5 E-5	8.7 E-5	3.7 E-5
2	2	5.1 E-6	1.2 E-5	4.8 E-6	1.4 E-5	1.5 E-5
3	3	2.0 E-5	4.2 E-6	6.0 E-6	1.2 E-5	4.8 E-6
4	4	8.9 E-6	2.3 E-6	4.3 E-6	7.6 E-6	3.0 E-6
5	11	7.3 E-6	4.4 E-6	3.8 E-6	1.8 E-6	2.1 E-6
7	25	5.0 E-6	1.9 E-6	2.1 E-6	1.1 E-6	2.5 E-6
12	60	5.0 E-6	4.8 E-7	8.9 E-7	5.5 E-7	1.5 E-6
15	154	2.0 E-6	2.8 E-7	5.0 E-7	2.1 E-7	2.0 E-6
19	277	7.9 E-7	6.0 E-6	3.3 E-7	3.2 E-7	1.5 E-6
23	404	5.4 E-7	1.2 E-6	1.7 E-7	5.1 E-7	1.1 E-6
25	467	3.8 E-7	3.7 E-7	5.1 E-7	1.3 E-7	1.8 E-6

DISTRIBUTION

No. of
Copies

OFFSITE

A. A. Churm
DOE Chicago Patent Division
9800 South Cass Avenue
Argonne, IL 60439

27 DOE Technical Information Center

B. Adams
Corning Glass Works
Technical Staffs Division
Corning, NY 14830

Allied Chemical Corporation
(File Copy)
550 2nd Street
Idaho Falls, ID 83401

J. M. Batch
Battelle Memorial Institute
505 King Avenue
Columbus, OH 43201

Battelle Memorial Institute
Office of Nuclear Waste
Isolation
Attn: Beverly Rawles
505 King Avenue
Columbus, OH 43201

J. R. Berreth
Allied Chemical Corporation
550 2nd Street
Idaho Falls, ID 83401

R. E. Blanco
Union Carbide Corporation (ORNL)
Chemical Technology Division
P.O. Bpx Y
Oak Ridge, TN 37830

J. O. Blomeke
Union Carbide Corporation (ORNL)
P.O. Box Y
Oak Ridge, TN 37830

No. of
Copies

Brookhaven National Laboratory
Reference Section
Information Division
Upton, NY 11973

R. A. Buckham
Allied-General Nuclear Service
P.O. Box 847
Barnwell, SC 29812

Wayne Carbiener
Office of Nuclear Waste
Isolation
505 King Avenue
Columbus, OH 43201

T. C. Chee
DOE Office of Nuclear Waste
Management
Washington DC 20545

R. B. Chitwood
DOE Division of Nuclear Power
Development
Washington DC 20545

5 D. Coles
Lawrence Livermore Laboratory
P.O. Box 808
Livermore, CA 94550

P. Colombo
Department of Applied Science
Brookhaven National Laboratory
Upton, NY 11973

C. R. Cooley
DOE Office of Nuclear Waste
Management
Washington DC 20545

J. L. Crandall
E. I. duPont DeNemours and
Company
Savannah River Laboratory
Aiken, SC 29801

No. of
Copies

R. E. Cunningham
Deputy Director for Fuels and Materials
Nuclear Regulatory Commission
Silver Springs, MD 20910

H. Diamond
Argonne National Laboratory
9700 South Cass Avenue
Argonne, IL 60439

J. P. Duckworth
Plant Manager
Nuclear Fuel Services, Inc.
P.O. Box 124
West Valley, NY 14171

J. O. Duguid
Office of Nuclear Waste Isolation
505 King Avenue
Columbus, OH 43201

Environmental Protection Agency
Technology Assessment Division (AW-559)
Office of Radiation Programs
Washington DC 20460

Keith Flynn
Argonne National Laboratory
9700 South Cass Avenue
Argonne, IL 60439

A. Friedman
Argonne National Laboratory
9700 South Cass Avenue
Argonne, IL 60439

R. G. Garvin
E. I. duPont DeNemours and Company
Savannah River Laboratory
Aiken, SC 29801

No. of
Copies

H. W. Godbee
Union Carbide Corporation (ORNL)
Chemical Technology Division
P.O. Box Y
Oak Ridge, TN 37830

E. S. Goldberg
DOE Savannah River Operations Office
P.O. Box A
Aiken, SC 29801

Mark Harwell
P.O. Box 667
Cannon Beach, OR 97110

C. A. Heath
DOE Office of Nuclear Waste Management
Washington DC 20545

R. E. Heineman
Office of Nuclear Waste Isolation
505 King Avenue
Columbus, OH 43201

L. L. Hench
Department of Materials Science and Engineering
University of Florida
Gainsville, FL 32611

H. Henning
Electric Power Research Institute
3412 Hillview Avnue
P.O. Box 10412
Palo Alto, CA 94301

Peter Hoffman
Office of Nuclear Waste Isolation
505 King Avenue
Columbus, OH 43201

No. of
Copies

J. L. Jardine
Argonne National Laboratory
9700 South Cass Avenue
Argonne, IL 60439

J. K. Johnstone
Sandia Laboratories
Albuquerque, NM 87107

J. A. Kelley
E. I. dePont DeNemours and
Company
Savannah River Laboratory
Aiken, SC 29801

C. J. Kershner
Monsanto Research Corporation
Mound Laboratory
P.O. Box 32
Miamisburg, OH 45342

J. F. Kircher
Office of Nuclear Waste
Isolation
Battelle Memorial Institute
505 King Avenue
Columbus, OH 43201

D. A. Knecht
Allied Chemical Corporation
550 2nd Street
Idaho Falls, ID 83401

E. H. Kobish
Solid State Division
Oak Ridge National Laboratory
Oak Ridge, TN 37830

D. Lam
Argonne National Laboratory
9700 South Cass Avenue
Argonne, IL 60439

Paul W. Levy
Brookhaven National Laboratory
Upton, NY 11973

No. of
Copies

S. E. Logan
Los Alamos Technical
Associates, Inc.
P.O. Box 410
Los Alamos, NM 87544

Los Alamos Scientific
Laboratory (DOE)
P.O. Box 1663
Los Alamos, NM 87544

J. B. Martin
United States Nuclear Regulatory
Commission
Washington DC 20555

W. C. McClain
RE/SPEC Inc.
P.O. Box 725
Rapid City, SD 57701

M. D. McCormack
E.G. & G. Idaho, Inc.
P.O. Box 1625
Idaho Falls, ID 83401

Sheldon Meyers
DOE Office of Nuclear Waste
Management
Washington DC 20545

Don Moak
Office of Nuclear Waste
Isolation
505 King Avenue
Columbus, OH 43201

M. A. Molecke
Sandia Laboratories
Albuquerque, MN 87107

W. E. Mott
DOE Division of Environmental
Control Technology
Washington DC 20545

No. of
Copies

J. Neff, Program Manager
Department of Energy
Columbus Program Office
505 King Avenue
Columbus, OH 43201

R. M. Neilson, Jr.
Department of Applied Science
Brookhaven National Laboratory
Upton, NY 11973

Ed Norris
Los Alamos Scientific Laboratory
CNC-DO Mailstop 760
Los Alamos, NM 87545

2 Oak Ridge National Laboratory (DOE)
Central Research Library
Document Reference Library
P.O. Box X
Oak Ridge, TN 37830

G. Oertel
DOE Office of Nuclear Waste Management
Washington DC 20545

Al Ogle
Los Alamos Scientific Laboratory
CNC-CO Mailstop 760
Los Alamos, NM 87545

A. F. Perge
DOE Office of Nuclear Waste Management
Washington DC 20545

M. S. Plodinec
E. I. duPont DeNemours and Company
Savannah river Laboratory
Aiken, SC 29801

No. of
Copies

John Pomeroy
Technical Secretary
National Academy of Sciences
Committee of Radioactive Waste Management
National Research Council
2101 Constitution Avenue
Washington DC 20418

R. G. Post
College of Engineering
University of Arizona
Tucson, AZ 85721

Professor Guna Salvaduray
Materials Engineering
San Jose State University
San Jose, CA 95192

L. Ramspott
Lawrence Livermore Laboratory
P.O. Box 808
Livermore, CA 94550

D. M. Rohrer
United States Nuclear Regulatory Commission
Washington DC 20555

R. G. Romatowski
DOE Office of Nuclear Waste Management
Washington DC 20545

B. Scheetz
210 Materials Research Laboratory
Pennsylvania State University
University Park, PA 16802

T. Scott
Ames Laboratory
Iowa State University
Ames, IA 50011

No. of
Copies

W. C. Seymour
E.G. & G. Idaho, Inc.
P.O. Box 1625
Idaho Falls, ID 83401

M. J. Steindler
Argonne National Laboratory
9700 South Cass Avenue
Argonne, IL 60439

D. B. Stewart
U.S. Department of Interior
959 National Center
Geological Survey
Reston, Virginia 22092

J. Tewhey
Lawrence Livermore Laboratory
P.O. Box 808
Livermore, CA 94550

John Van Cleve
DOE Oak Ridge Operation Office
P.O. Box X
Oak Ridge, TN 37830

E. Vejovoda, Director
Chemical Operations
Rockwell International
Rocky Flats Plant
P.O. Box 464
Golden, CO 80401

D. L. Vieth
DOE Office of Nuclear Waste Management
Washington DC 20545

J. W. Voss
Office of Nuclear Waste Management
Battelle Memorial Institute
505 King Avenue
Columbus, OH 43201

No. of
Copies

R. D. Walton
DOE Office of Nuclear Waste Management
Washington DC 20545

W. Weart
Sandia Laboratories
Albuquerque, NM 87107

H. Weed
Lawrence Livermore Laboratory
P.O. Box 808
Livermore, CA 94550

W. White
210 Materials Research Laboratory
Pennsylvania State University
University Park, PA 16802

J. B. Whitsett
DOE Idaho Operations Office
550 2nd Street
Idaho Falls, ID 83401

A. Williams
Allied-General Nuclear Service
P.O. Box 847
Barnwell, SC 29812

FOREIGN

Dr. P. G. Alfredson
Chief, Chemical Technology Division
Australian Atomic Energy Commission
Research Establishment
Lucas Heights, New South Wales, 2232

Rene Amavis
EURATOM
Health Physics Division
29, Rue Aldringer
Luxembourg
BELGIUM

No. of
Copies

R. Bonniaud
Center de Marcoule
B.P. 170
30200 Baguols-Sur-Ceze
FRANCE

Bundesministerium fur Forschung
und Technologie
Stressemannstrasse 2
5300 Bonn
WEST GERMANY

Center for Atomic Energy
Documentation (ZAED)
Attn: Dr. Mrs. Bell
P.O. Box 3640
7500 Karlsruhe
WEST GERMANY

D. W. Clelland
United Kingdom Atomic Energy
Authority
Risley
ENGLAND

F. Gera
CHEN
CSN Cassaccia L.I.S.
C.P. 24,00 00100
Rome
ITALY

K. B. Harvey
Atomic Energy of Canada, Ltd.
W.N.R.E. Pinawa, Manitoba
ROE 1LO
CANADA

Ake Hultgren
Studsvik Energiteknik AB
S-611 82 Nykoping
SWEDEN

International Atomic Energy
Agency
Kartner Ring 11
P.O. Box 590
A-1011, Vienna
AUSTRIA

No. of
Copies

K. D. B. Johnson
Atomic Energy Research
Establishment,
Harwell, Didcot,
Berks
ENGLAND

H. Krause
Kernforschungszentrum Karlsruhe
GmbH (KfK)
Postfach 3640
D7500 Karlsruhe
WEST GERMANY

F. Laude
Centre de Marcoule
B.P. 170
30200 Baguols-Sur-Ceze
FRANCE

Hans W. Levi
Hahn-Meitner Institut
1 Berlin 39
Glienickerstr. 100
WEST GERMANY

Library
Studsvik Energiteknik AB
S-611 01Nykoping
SWEDEN

Dr. Werner Lutze
Hahn-Meitner
Institut fur Kernforschung
Berlin, GmbH
Glienicker Strasse
100, B-1000
Berlin, 39
GERMANY

J. A. C. Marples
Atomic Energy REsearch
Establishment
Harwell, Didcot,
Berks
ENGLAND

No. of
Copies

E. R. Merz
Institut fur Chemische Technologie
Kernforschungsanloge Julich GmbH
D517 Julich
Postfach 365
Federal Republic
WEST GERMANY

Tonis Papp
Swedish Nuclear Supply Co.
SKBF/KBS
Box 5864
S-102 48 Stockholm
SWEDEN

N. S. Aunder Rajan
Bhabha Atomic Research Centre
Goverment of India
Hall No. 5
Trombay
Bombay 8S
INDIA

P. J. Regnaut
Centre d'Etudes Nucleaires de Fontenay-aux Roses
Boite Postale 6
92 - Fontenay-aux Roses
FRANCE

A. Rice
Department of Ocean Engineering
University of Newcastle upon Tyne
Newcastle upon Tyne, NE I 7 RU
ENGLAND

Dr. Piero Risoluti,
AGIP NUCLEARE
c/o COMB Casaccia
c.p. 2400
Rome
ITALY

No. of
Copies

Rolf Sjoblom
Studsvik Energiteknik AB
S-611 82 Nykoping
SWEDEN

C. Sombret
Centre de Marcoule
B.P. 170
30200 Baguols-Sur-Ceze
FRANCE

G. G. Strathdee
Atomic Energy of Canada, Ltd.
W.N.R.E. Pinawa, Manitoba
ROE 1LO
CANADA

S. Tashiro
Japan Atomic Energy Research Institute
Environmental Safety research Laboratory
1-1-13, Shibashi
Minatopku, Tokyo
JAPAN

M. Tomlinson
Director of Chemistry and Materials Science division
Atomic Energy of Canada Ltd.
Whiteshell Nuclear Research Establishment
Pinawa, Manitoba
CANADA

T. T. Vandergraaf
Atomic Energy of Canada, Ltd.
W.N.R.E. Pinawa, Manitoba
ROE 1LO
CANADA

No. of
Copies

ONSITE

5 DOE Richland Operations Office

P. A. Craig
H. E. Ransom
J. J. Schreiber
M. W. Shupe
M. J. Zamorski

10 Rockwell Hanford Operations

J. Babad
M. Bensky
R. A. Deju
R. J. Gimera
J. D. Kaser
M. J. Kupfer
W. W. Schultz
M. J. Smith
D. D. Wodrich
File Copy

Exxon Nuclear Company

S. J. Beard

Joint Center for Graduate Study

J. Cooper

2 United Nuclear Industries, Inc.

T. E. Dabrowski
A. E. Engler

4 Westinghouse Hanford Company

A. G. Blasewitz
D. A. Cantley
R. L. Fish
A. C. Leaf

109 Pacific Northwest Laboratory

S. M. Barnes
G. L. Benson

No. of
Copies

W. J. Bjorklund
H. T. Blair
W. F. Bonner
D. J. Bradley (10)
A. Brandstetter
R. A. Brouns
J. B. Brown, Jr.
J. L. Buelt
R. L. Bunnell
L. A. Chick
T. D. Chikalla
D. B. Cearlock
M. O. Cloninger
G. W. Dawson
W. J. Deutsch
R. D. Dierks
F. H. Dove
J. W. Finnigan
W. J. Gray
M. S. Hanson
F. T. Hara
C. O. Harvey (2)
M. H. Henry
O. F. Hill
L. K. Holton
B. Howes
J. H. Jarrett
Y. B. Katayama
W. S. Kelly
R. S. Kemper
D. E. Knowlton
K. M. Krupka
M. R. Kreiter
W. L. Kuhn
D. E. Larson
R. P. Marshall
J. L. McElroy
G. L. McVay (10)
G. B. Mellinger
J. E. Mendel
R. D. Nelson
W. R. Nelson
R. E. Nightingale
D. E. Olesen
C. R. Palmer
R. D. Peters
L. R. Pederson

No. of
Copies

A. M. Platt
D. Rai
J. F. Relyea
F. P. Roberts
W. A. Ross
J. M. Rusin
J. L. Ryan
R. J. Serne
J. W. Shade
D. H. Siemens
D. J. Silviera
S. C. Slate
R. G. Strickert
J. L. Swanson

No. of
Copies

M. T. Thomas
R. T. Treat
R. P. Turcotte
H. H. Van Tuyl
J. W. Wald
W. E. Weber
R. A. Walter
R. Wang
J. H. Westsik, Jr.
L. D. Williams
Technical Information (5)
Publishing Coordination SH/B(2)
Water and Land Resource
Library (10)