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Salt Repository Project

Interactive Leach Tests of UO_2 and Spent Fuel with Waste Package Components in Salt Brine

J. O. Barner
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September 1986

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

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

Spent fuel is being considered as a waste form for disposal in a repository located in salt. To adequately model spent fuel performance as a waste form that may be contacted by brine in a repository, it is necessary to describe the leach (dissolution) behavior of spent fuel and the chemical interactions of the released radionuclides with their environment. To this end, leach tests were conducted on:

- UO_2 in Permian Basin salt brine or deionized water at test temperatures of 25, 75, and 150°C. Some tests were done in the presence of ductile cast iron, which is a representative overpack material, and/or oxidized Zircaloy, which is the usual fuel cladding material.
- spent fuel (H. B. Robinson) in Permian Basin salt brine at 25 and 75°C. Some of the tests were conducted in the presence of ductile cast iron.

The release values for leach periods up to 60 days were determined for systems utilizing both UO_2 and spent fuel. This report is based upon data obtained during 1982 and 1983.

The larger temperature dependence of the leach behavior that was observed for deionized water than was observed for brine is difficult to interpret on the basis of our present knowledge. Differences in UO_2 alteration products may be involved. For example, the existence of sodium uranates is likely in the brines but not possible with deionized water. Differences in ionic strength may also play a role.

Observations derived from the leach tests performed in brine include: 1) the presence of iron coupons had no effect on total release of uranium from either spent fuel or UO_2 but did reduce solution concentrations, and 2) 100 to 200 times more uranium was released from spent fuel than from UO_2 per unit of geometric surface area.

ACKNOWLEDGMENTS

These studies are part of the Waste Package Program sponsored by the Salt Repository Project Office/Office of Nuclear Waste Isolation, which is part of the U.S. Department of Energy sponsored Geologic Repository Deployment Program. The leach tests were performed by W. C. Richey, C. O. Harvey, and H. J. Cartrnell.

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1.0 INTRODUCTION

Spent fuel is being considered as a waste form for disposal in a repository located in salt. To adequately model the behavior of this waste form if it is contacted by brine in a repository, it is necessary to describe: 1) the release of radionuclides from the spent fuel, 2) the extent to which the release is affected by the interaction between the waste form and other waste package components, 3) the distribution of the released radionuclides, and 4) the solubility limits of radionuclides in their environment. The experiments described here are intended to provide initial data concerning the needed information for at least the first three items. Uranium release information was determined using unirradiated UO_2 in combination with some of the expected waste package components, i.e., Zircaloy and iron, over the temperature range from 25 to 150°C. Information on other radionuclides including uranium was also determined using spent fuel in combination with some of the expected waste package components at 25 and 75°C. Unirradiated UO_2 was used because spent fuel is primarily UO_2 and, therefore, an understanding of this material should aid in understanding spent fuel. Because of the lower radioactivity, UO_2 does not have to be remotely handled in a hot cell, thus, investigation is easier and more analytical equipment can be used to gather data for understanding the interactions with aqueous environments. These studies are part of the Waste Package Program (WPP) sponsored by the Salt Repository Project Office (SRPO)/Office of Nuclear Waste Isolation (ONWI), which is part of the Department of Energy (DOE) sponsored Geologic Repository Deployment Program.

2.0 CONCLUSIONS

As a result of this study, the following conclusions were made:

- The presence of ductile cast iron has no measurable effect on total release of uranium from spent fuel or UO_2 but greatly reduces solution concentrations.
- Ductile cast iron has no measurable effect on total release of plutonium, technetium, or cesium from spent fuel, but greatly reduces plutonium and technetium solution concentrations.
- Greater than 100 times more uranium leaches from spent fuel than from UO_2 under similar conditions per unit geometric surface area.
- Uranium leaching from both UO_2 and spent fuel is not strongly temperature dependent.
- Oxidized Zircaloy has no effect on UO_2 leaching.
- The fission products technetium and cesium are released in greater abundance and plutonium is released in lesser abundance from spent fuel than is uranium, when compared to congruent leaching.

3.0 EXPERIMENTAL PROCEDURE

Experimental test matrices and testing materials, test equipment and procedures, analytical techniques, and leachants are described in the following subsections.

3.1 TEST MATRICES AND TESTING MATERIALS

Three separate test matrices were used based on the material to be tested, the equipment required and/or available, and the testing temperature. The first matrix was for unirradiated UO_2 at 25 and 75°C. The second matrix was for unirradiated UO_2 at 150°C. The third was for spent fuel at 25 and 75°C.

3.1.1 UO_2 Leach Test Matrix at 25 and 75°C

The test matrix for the 25 and 75°C tests is shown in Table 3.1. Unirradiated UO_2 pellets were tested according to a procedure that was based on the Materials Characterization Center (MCC) procedure "MCC-1, Static Leach Test Method" (DOE/TIC-11400). Testing temperatures were 25 and 75°C with primary leach periods of 2, 5, 14, 28, and 60 days. The primary leachant was synthetic Permian Basin salt brine, which is described later. Deionized water was used as the leachant in a few tests to permit better comparison with results from other studies. Ductile cast iron coupons were included in some tests to represent overpack material; oxidized Zircaloy was included in other tests to simulate fuel cladding; and some tests included both iron and oxidized Zircaloy. Finally, a few spot checks were included in the matrix to determine the effect of oxidation of iron prior to leaching, and to explore the releaching of pellets using fresh leachant.

The UO_2 used in the investigation was depleted in ^{235}U; as-fabricated characteristics of the pellets are listed in Table 3.2. An average geometric surface area of 360 mm^2 per pellet was used for calculational purposes for most of the tests even though the lengths of the pellets varied by ±5%. For a few retests where the leach data was questionable, pellets of the geometry listed in Table 3.2 were unavailable and pellets with a geometric surface area of

TABLE 3.1. UO_2 Leach Test Matrix at 25 and 75°C

Days	No. of Tests Run Under Each Condition			
	UO_2	UO_2-Fe	UO_2-Zr	UO_2-Fe-Zr
25 and 75°C in Brine				
2	1	1	1	1
5	1	1	1	1
14	1	1	1	1
28	1	1	1	1
60	3	3	3	3
Additional Specimen Releached After 60-Day Exposure, 25 and 75°C				
14	-	-	-	1
Additional Specimen in Brine with Oxidized Iron, 75°C Only				
14	-	1	-	-
Additional Specimens in Deionized Water, 25 and 75°C				
7	-	-	1	1
14	1	1	1	1

TABLE 3.2. Characteristics of UO_2 Pellets

Characteristic	Value
Density	94% T.D.
Diarneter	8.3 mm
Length	9 to 10 mm
Oxygen/Uranium	2.001
Grain Size at Surface	~ 3.0 μm
Surface Condition	Centerless ground on cylindrical surface, ends were as-sintered
Total Impurities	~ 700 ppm

585 mm^2 were substituted. The pellets were used in the as-received condition (i.e., the cylindrical surface had been ground and cleaned using deionized water immediately after grinding).

The iron used in the study had the chemical composition range shown in Table 3.3. The nominal surface area of the iron specimens was the same as the

Table 3.3. Composition Range for Iron Used in Leach Tests

Element	Weight Percent
C	3.53 - 3.90
Mn	0.27 - 0.31
Si	2.43 - 2.51
P	0.05 - 0.08
S	0.004 - 0.005
Fe	Balance

UO_2 pellets, 360 mm^2. The surface of the single specimen of oxidized iron was prepared by exposing the sample to air at 200°C for one week, followed by a week-long exposure to Permian Basin salt brine at 75°C.

The Zircaloy-4 used in the study was standard PWR-type tubing. A chemical analysis for the material was not available. The nominal specimen surface area was 368 mm^2, essentially the same as the UO_2 pellets. The Zircaloy-4 specimens were oxidized by autoclaving at 1500 psig in steam at 400°C for 189 h. The surfaces were oxidized to simulate an average expected cladding condition of irradiated fuel rods.

The surfaces of both the iron and Zircaloy-4 specimens were cleaned prior to testing by: 1) three sequential 5 min rinses in ethanol in an ultrasonic cleaner, 2) three sequential 5-min rinses in deionized water in an ultrasonic cleaner, and 3) drying at 110° C for one-half hour in air.

The specimen releached after its initial 60-day exposure was from one of the UO_2-Fe-Zr tests. It was releached for 14 days using fresh brine and also fresh iron and Zircaloy-4 coupons.

The synthetic Permian Basin salt brine was prepared by adding the reagents listed in Table 3.4 to deionized water and diluting the resultant solution to a volume of one liter. This simulates the saturated solution obtained by dissolving actual salt core from the G. Friemel Hole No. 1 in Deaf Smith County, Texas, which is located in the Permian Basin, and thus it simulated a Permian

TABLE 3.4. Reagents for Synthetic Permian Basin Brine

Reagents	Weight, g
NaCl	310.05
Na_2SO_4	4.729
$CaCl_2 \cdot 2H_2O$	5.733
$MgCl_2 \cdot 6H_2O$	0.524
KC1	0.0745
$SrCl_2$	0.0634
$NaHCO_3$	0.0420
NaBr	0.0412
$ZnCl_2$	0.0162
NaF	0.0024

Basin intrusion brine. The ratio of UO_2 surface area to leachant volume (SA/V) was 10 m^{-1} for all tests except the few retests with the larger pellets. For the latter, SA/V was 16 m^{-1}.

3.1.2 UO_2 Leach Test Matrix at 150°C

Tests were conducted at 150°C; the static-test matrix is given in Table 3.5. Nearly equivalent matrices were completed using brine and deionized water. In addition to the static tests, two rocking autoclave tests at 150°C, one each with brine and deionized water, with sampling times between 4 and 58 days were conducted.

The UO_2, iron, Zircaloy, and leachants used for the 150°C tests were identical to those in the 25 and 75°C tests. SA/V was 10 m^{-1} for all tests.

3.1.3 Spent Fuel Leach Test Matrix

Tests conducted on spent fuel in a hot cell necessitate specialized, expensive equipment; therefore, the static leach test matrix for the unclad spent fuel (Table 3.6) was limited to single tests for each time-temperature condition, and only spent fuel and spent fuel plus iron were studied. Leach periods ranged from 2 to 60 days at 25°C and only 5 and 28 days for the 75°C tests. A single check point was conducted with spent fuel at 25°C using real

TABLE 3.5. UO_2 Static Leach Test Matrix at 150°C in Brine and Deionized Water

No. Days	No. of Tests Run Under Each Condition		
	UO_2	UO_2-Fe	UO_2-Zr
Brine - Static Tests			
7	1	1	1
14	1(a)	1	1
28	2	2	2
60	2	1(b)	1
Deionized Water - Static Tests			
7	1	1	1
14	1(a)	1	1
28	2	2	2
60	2	1	1

(a) Test period actually was 15 days.
(b) Test period actually was 62 days.

TABLE 3.6. Spent Fuel Leach Test Matrix

No. Days	No. of Tests Run Under Each Condition	
	Spent Fuel	Spent Fuel-Fe
Synthetic Brine - 25°C		
2	1	1
5	1	1
14	1	1
28	1	1
60	1	1
Synthetic Brine - 75°C		
5	1	1
28	1	1
Real Brine(a) - 25°C		
14	1	-

(a) Obtained by preparing a saturated solution of salt cored from the Permian Basin.

brine (obtained by preparing a saturated solution of actual Permian Basin salt). All other tests were conducted in synthetic Permian Basin salt brine.

The spent fuel used in the study was from fuel bundle BO-5, which was discharged from the H. B. Robinson II reactor on May 6, 1974. The average burnup of the fuel was 28 MWd/kgM. Fuel was removed from the fuel rod at the Battelle-Columbus Division Hot Laboratory. Unclad fuel fragments from more than one rod were combined and shipped to PNL for testing. Representative samples of fuel were dissolved and chemically analyzed for uranium, plutonium and two important fission products; results are listed in Table 3.7.

Each leach specimen consisted of three fuel fragments with a surface area of about 2 cm^2. This area was estimated from photographs that were taken from three different angles of each specimen together with a millimeter scale. The SA/V was 10 m^{-1} for all tests.

The iron specimens were prepared to have a surface area equivalent to the fuel fragments with which they were leached. The composition of the iron is listed in Table 3.3.

3.2 TEST EQUIPMENT AND PROCEDURES

The testing equipment varied for the three types of tests and is described in the following subsections. Leach containers and the associated specimen-contacting equipment were cleaned according to "MCC-1, Static Leach Test Method" requirements (DOE/TIC-11400).

TABLE 3.7. Selected Chemical Analysis for the Spent Fuel

Species	Value(a)
Uranium	8.4 x 10^5 µg/g
^{239}Pu and ^{240}Pu	2.47 x 10^7 Bq/g
^{99}Tc	4.55 x 10^5 Bq/g
^{137}Cs	2.37 x 10^9 Bq/g

(a) Total uranium concentration was measured whereas only the activities of the radioisotopes listed were determined for the other elements.

3.2.1 UO_2 Leach Tests at 25 and 75°C

The 25 and 75°C static leach tests using UO_2 were conducted in clean screwtop Teflon® leach containers containing the appropriate leachant, either brine or deionized water. The residual atmosphere in the container was air. The 25°C tests were conducted by exposure of the sealed containers to ambient laboratory temperature. The 75°C tests were conducted in an oven maintained at temperature to within ±1°C. The specimens (i.e., UO_2 pellets, iron and Zircaloy-4 coupons) rested on a perforated Teflon specimen holder during the leach period to minimize contact between the specimens and the container and maxiniize the surface of the specimens exposed to the leachant.

Following the leach period, the containers were cooled to room temperature and the specimen(s) removed. The leachant was separated into aliquots for a pH measurement(a) and chemical analyses. The uranium chemical analysis consisted of direct fluorometric measurements on: 1) acidified solutions of both the original leachate and a filtrate produced by passing the leachate through a 1.8 nm filter, 2) a plateout solution produced by acid stripping the inner surface of the leach container and specimen holder with 5M HNO_3, and 3) solutions produced by acid stripping either the iron with 3M HCl or the Zircaloy-4 with 5M HNO_3.

3.2.2 UO_2 Leach Tests at 150°C

The 150°C static leach tests with UO_2 were conducted in clean Teflon-lined digestion vessels containing either brine or deionized water. The residual atmosphere in the leach vessel was air. The tests were conducted in an oven maintained at a temperature of 150 ± 1°C. The specimens rested on a perforated Teflon specimen holder during the leach period.

Following the leach period, the specimens were cooled to room temperature and the pH(a) and uranium analyses conducted. The types of UO_2 samples and

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(a) Note that, for brine, measured pH values are somewhat unreliable and can be in error by as much as one unit, or sometimes slightly more than one unit.

analyses were identical to those for the 25 and 75°C static leach tests with UO_2, except two pellets were used in each test instead of one and the amount of leachate was, therefore, also doubled.

The 150°C tests using UO_2 that were conducted in a rocking autoclave exposed the pellets to originally 230 ml of leachant in a gold bag within the autoclave. Approximately 5 ml of leachate were removed at each specified time, passed through a 1.8 nm filter, and the filtrate was analyzed for uranium.

3.2.3 Spent Fuel Leach Tests

The 25 and 75°C spent fuel leach tests were conducted in clean quartz leach containers. The residual atmosphere in the container was air. The lower temperature tests were conducted under ambient hot-cell conditions, which maintained the temperature at 25 & 1°C. Quartz lids were sealed to the ground surfaces of the leach containers using a silicone vacuum grease for the 25°C tests. The 75°C tests were conducted in a water bath and maintained at 75 ± 1°C. Silicone "O"-rings were used to seal the lids of leach containers for the 75°C tests. The spent fuel and iron specimens were placed in perforated quartz baskets to minimize contact between the specimens and the container, and to maximize the surfaces of the specimens exposed to the leachant.

Following the leach period, the containers were cooled to ambient hot-cell temperature, weighed to assure no leachant had been lost, and the leachate pH was measured. The spent fuel and iron specimens were removed and the leachate was passed through a 1.8 nm filter. The filtrate was used to rinse the container and specimen basket and then passed through the same filter again. An analytical sample was taken from the filtrate and acidified with 0.1 ml of concentrated nitric acid to assure that the uranium and plutonium remained in solution. A 5M HNO_3, 0.05M HF solution was placed in each leach container with its associated specimen basket in volumes equal to the original leachate volume and allowed to stand 24 hours at ambient temperature to dissolve any material plated out on the container walls or on the basket. In addition, each filter was placed in the same type of solution and allowed to stand for 24 hours at ambient temperature and then heated to near boiling for 30 minutes. Aliquots of the plateout and filter solutions were taken for analysis. Note, that for the spent fuel experiments, the analyses were done on the filtrate and on a

solution produced by cleaning up the filter, rather than on the leachate and filtrate, as was done for the UO_2 tests. For comparative purposes, a leachate value was calculated for the spent fuel experiments by summing the filtrate and filter-solution results.

Material plated out on the iron coupons was removed by placing each coupon in a series of three fresh solutions of 6M HCl; the coupons were left in each successive solution for a few minutes. Then the solutions were combined and diluted to a known volume. Measurement of the radioactivity of a few of the iron coupons before and after the acid treatments demonstrated that this procedure was adequate for removal of the deposited radionuclides.

A portion of each solution was taken for uranium analysis and a second portion for plutonium, cesium and technetium analyses. The amount of uranium was determined by making two direct fluorometric measurements on each solution; once by itself and a second time after spiking with a known amount of uranium to reduce matrix effects. The amount of plutonium was determined by evaporating the solution to dryness, dissolving in 1M HCl, extracting the plutonium into thenoyltrifluoroacetone, evaporating onto a plate and alpha-counting the residue. The amount of cesium was determined by direct gamma counting each solution. The amount of technetium was determined by: removing most other cationic radioactivity from the solution in a cation exchange column; adding tetraphenylarsonium chloride to the solution to form tetraphenylarsonium pertectnetate; extracting the latter from the aqueous solution with hexone; evaporating the hexone from a known amount of the extract; and counting the residue with a beta proportional counter.

4.0 RESULTS AND DISCUSSION

4.1 BASIS FOR COMPARISON OF RESULTS

Results are presented in terms of normalized mass loss, which is the actual mass loss divided by the fraction of the given element present in the specimen, and divided by the surface area of the specimen. This procedure allows a direct comparison of leach values for specimens of different size and composition. It should be noted, in particular, that the normalized mass loss will have the same value for each element provided that the specimen leaches congruently. Thus, unequal normalized mass loss is indicative of preferential (incongruent) leaching. For uranium release from UO_2 or spent fuel, results are presented in terms of the normalized uranium mass loss:

$$(NL)_u = \frac{M_u}{f_u\,(SA)} \tag{1}$$

where $(NL)_u$ = normalized uranium mass loss, g/m^2,
M_u = mass of uranium in a solution = mass per unit volume times solution volume, g,
f_u = mass fraction of uranium in the unleached specimen = 0.88, and
SA = surface area of the leach specimen, m^2.

Similarly, the results for the more radioactive species that were analyzed by activity are presented in terms of the normalized elemental mass loss:

$$(NL)_i = \frac{a_i}{a_o}\,\frac{W_o}{SA} \tag{2}$$

where $(NL)_i$ = normalized elemental mass loss, g/m^2,
a_i = activity of the element in the solution, Rq,
a_o = activity of the element in the spent fuel fragment, Bq,
W_o = mass of the specimen, g, and
SA = surface area of the leach specimen, m^2.

In the following sections, the results are presented in terms of location of the released radioactive species; Sections 4.2 through 4.5 contain discussions of the data that are presented in tabular and graphical form in Section 4.6. The sum of species in the leachate, plateout on the Teflon or quartz, and the plateouts on the iron and Zircaloy-4, as applicable, represents the total released during a particular test. The amount in the filtrate represents the amount of the species that passed through the 1.8 nm filter. Any particle smaller than 1.8 nm would be very nearly molecular in size. Therefore, the filtrate can be considered to contain species that are in true solution. The difference between the leachate value and the filtrate value represents that which was suspended, possibly in colloidal form, in the leachate. For each group of tests, the results are presented in tabular form, in figures that summarize each leach system, and in figures that compare the location of the released species. Note that "Fe Plateout" is plotted in the summary figures whereas "Total Plateout" is plotted on one of the comparative figures in each data set. Inspection of the data tables shows that most of the plateout occurs on the iron with only a small amount on the Teflon and almost none on the Zircaloy-4. Thus, plotting "Fe Plateout" on one set of figures and "Total Plateout" on the other allows a clear presentation of the data.

4.2 UO_2 TESTS AT 25 AND 75°C

Table 4.1 (Page 4.10) and Figure 4.1 show the location of uranium released from the four different systems with UO_2 in brine at 25°C. The results are compared by location of the uranium in the leach systems in Figures 4.2 through 4.5. In addition, one of the UO_2 specimens originally leached for 60 days with iron and Zircaloy coupons was placed in fresh brine with fresh iron and Zircaloy coupons and leached for an additional 14 days. Results from this test are shown in Table 4.1 and Figures 4.2 through 4.5 as "Releach UO_2-Fe-Zr". In all cases, the total uranium released in the four systems that were tested at 25°C tended to increase with leaching time up to 60 days. However, significant fractions of the uranium, ranging from 32% for the UO_2-Fe system to 79% for the UO_2-Zr system, were released in the first two days. The total uranium released in the four systems does not appear to be significantly different.

The major difference in leach behavior in the four systems was the location of the uranium after release from the UO_2. In the UO_2 and UO_2-Zr systems, essentially all of the released uranium was dissolved in the leachate (Figures 4.1, 4.3 and 4.5). The plateout from solution in these systems was very small, <5%, and remained essentially constant over 60 days (Figure 4.4). For the UO_2-Zr system, essentially no plateout occurred on the oxidized Zircaloy-4.

On the other hand, most of the uranium released in the UO_2-Fe and UO_2-Fe-Zr systems plated out on the iron (Figure 4.4 and Table 4.1). Essentially no plateout occurred on the oxidized Zircaloy-4 in the UO_2-Fe-Zr system, and plateout on the teflon in both systems with iron was similar to that in the UO_2 and UO_2-Zr system. The concentration of uranium in the leachates for the systems with iron decreased with time up to 60 days, and significant quantities of the uranium that remained in the leachates at the longer times were filterable (Table 4.1).

For the single specimen in the UO_2-Fe-Zr system that was releached for 14 days in fresh brine after originally being leached for 60 days, the leach behavior was apparently very similar to the leaching of a fresh pellet.

The results for the 75°C leach tests in brine are presented in Table 4.2 (Page 4.14) and Figures 4.6 through 4.10 in a manner analogous to the way in which the 25°C results were presented. The uranium released at 75°C was about the same as at 25°C, and essentially all was released in the first five days. Within the scatter in the data, there appears to be no significant difference in uranium released in the four systems, just as in the 25°C tests.

Again, the major difference in leach behavior in the four systems was the location of the uranium after release from the UO_2. In the UO_2 and UO_2-Zr systems respectively, about 50% and 80% of the released uranium was dissolved in the brine (Figure 4.6 or Figures 4.7 and 4.10). Essentially all of the uranium in the leachate was in solution, i.e., not filterable. The remaining uranium released in these systems was plated out on the teflon leach container.

On the other hand, the bulk of the uranium released in the UO_2-Fe and UO_2-Fe-Zr systems was plated out on the iron and teflon (Figure 4.9) or was filterable (Figures 4.7 and 4.10). Essentially no plateout occurred on the oxidized Zircaloy-4 in the UO_2-Zr and UO_2-Fe-Zr systems.

For the single specimen in the UO_2-Fe-Zr system that was releached for 14 days in fresh brine after originally being leached for 60 days, the amount of uranium released was significantly lower than for a fresh pellet, but the distribution of uranium within the system was similar to that for a fresh pellet. The behavior of the single sample that started with oxidized iron (UO_2-Fe_3O_4) was similar to the trend for the system utilizing non-oxidized iron. The actual uranium released at 14 days in the UO_2-Fe system was much higher than for the UO_2-Fe_3O_4 sample, but the former is totally out of line with the rest of the system and can probably be discounted.

Results for both the 25 and 75°C tests using UO_2 in deionized water are presented in Table 4.3 (Page 4.18) and Figure 4.11. Results from similar tests in brine are also shown in Figure 4.11 for comparison. For the UO_2 and UO_2-Zr systems, the total uranium released was four to five times higher in deionized water than in brine at 25°C and 20 to 40 times higher at 75°C. Almost all of the uranium was in the leachate with low values for plateout on the teflon and oxidized Zircaloy-4. However, unlike the brine results, approximately half the uranium in the leachate was filterable in both systems at 25°C, but little or none was filterable at 75°C.

For the UO_2-Fe and UO_2-Fe-Zr systems at both temperatures, the total uranium released in deionized water was comparable to that in brine. Most of the released uranium was plated out or was filterable in both systems, which is similar to the results in brine.

Table 4.4 (Page 4.20) shows the results of selected iron analyses of the leachates, filtrates, and plateout in some of the tests at 25 and 75°C in brine. The table includes results from tests that contained no iron for comparison and, as expected, almost none was found. In those tests that did contain iron coupons, some iron was found in the leachate but most of that was filterable (i.e., almost none was found in the filtrate after passing through a 1.8 nm filter). The Teflon plateout also contained some iron but little was found on the Zircaloy-4 plateout. This behavior is similar to that of the uranium in these tests.

The uranium concentration data for the 25 and 75°C tests using unirradiated UO_2, including pH data, are listed in Appendix A, Tables A.1, A.2, and A.3.

4.3 UO_2 TESTS AT 150°C

Table 4.5 (Page 4.21) and Figure 4.12 show the location of uranium released from the three different systems with UO_2 in brine at 150°C. The results are compared by location of the uranium in the leach systems in Figures 4.13 through 4.16. The greatest uranium release was for the UO_2-Fe system (Figure 4.13). However, almost all of the released uranium was plated out or was filterable. The plateout occurred preferentially on the iron but some uranium also plated out on the Teflon (Table 4.5). After 28 days of leaching, no uranium was detected in solution, Except for one high value at 14 days in the UO_2 tests, the leach behavior of the UO_2 and UO_2-Zr systems were similar (i.e., the total uranium released in both systems was similar, a significant fraction of the uranium in the leachate was in solution early in the test, and nearly all the uranium was filterable after 60 days of leaching).

Analogous results using deionized water are given in Table 4.6 (Page 4.24) and Figures 4.17 through 4.21. The amount of uranium released for the UO_2 system was an order of magnitude higher in deionized water than in brine. A significant fraction of the uranium in the UO_2 system was in solution (Figure 4.20) while most of the uranium in the UO_2-Fe system was plated out or filterable (Figures 4.20 and 4.21).

Results for the rocking autoclave tests are compared to the static tests for unirradiated UO_2 in brine and deionized water at 150°C in Figures 4.22 and 4.23, respectively. When brine was used as the leachant, the amount of uranium in solution after 48 days of leaching was similar in both test types, and the amount in solution decreased with time (Figure 4.22). When deionized water was the leachant, the amount of uranium in solution was consistently higher in the static system than in the rocking autoclave test (Figure 4.23). There is no obvious explanation for the difference, but it should be noted that the leach container materials were very different (i.e., Teflon vs. gold), and it may be that this somehow affected the degree of precipitation of uranium from solution.

The uranium concentration data for the 150°C tests using unirradiated UO_2 are given in Appendix A, Tables A.4, A.5, and A.6.

4.4 SPENT FUEL TESTS AT 25 AND 75°C

Results were obtained for the leaching of uranium, plutonium, technetium and cesium from spent fuel (SF) from the H. B. Robinson II reactor. The uranium released as a function of location in the leaching system for spent fuel in brine at 25 and 75°C is listed in Table 4.7 (Page 4.30) and Figure 4.24. The results are compared by location of the uranium in Figures 4.25 through 4.28. The total uranium released in the SF and SF-Fe systems at 25°C was similar and was about two orders of magnitude greater than that released from unirradiated UO_2.

The results for total uranium released in the two systems at 75°C were somewhat mixed. Results in the SF-Fe system were similar to those at 25°C, but a significant decrease with time occurred in the UO_2 systern. Uranium in the SF system was primarily in solution at 25°C but only partially in solution at 75°C. In the SF-Fe system, a large fraction of the uranium was plated out or was filterable at both test teniperatures.

The plutonium released as a function of location for the leaching of spent fuel in brine at 25 and 75°C is listed in Table 4.8 (Page 4.34) and Figure 4.29. The results are compared by location of the plutonium in Figures 4.30 through 4.33.

Trends for the plutonium leach results in brine at 25 and 75°C were similar to those for uranium except that the amount of plutonium released was only about one-tenth that of the uranium. Plutonium tended to be plated out or filterable at both temperatures in the SF-Fe system somewhat more plated out than uranium in the SF system and, again, mixed results at the two test periods at 75°C.

The technetium released as a function of location for the leaching of spent fuel in brine at 25 and 75°C is listed in Table 4.9 (Page 4.38) and Figure 4.34. The results are compared by location of the technetium in Figures 4.35 through 4.38. The release of technetium from the spent fuel in brine was four to six times greater than that of uranium at 25°C and twice as great at 75°C. Most of the technetium was in solution (i.e., not filterable or plated out) for the SF system at both test temperatures. In the SF-Fe system at 25°C, about three-fourths of the technetium was plated out on the iron. For the SF-Fe system at 75°C, about half the technetium was plated out on the iron

and a significant fraction of the technetium in the leachate was filterable. However, the amount of technetium in solution was still more than 10 times higher than that for uranium in the SF-Fe system at 75°C.

The amount of cesium released as a function of location for the leaching of spent fuel in brine at 25 and 75°C is listed in Table 4.10 (Page 4.42) and Figure 4.39. The results are compared by location of the cesium in Figures 4.40 through 4.43. The leach values for cesium were comparable to or greater than those for technetium, except that at both temperatures and for both the SF and the SF-Fe systems essentially all the cesium was in solution. Leach results at 75°C were higher than at 25°C in both systems.

The uranium, plutonium, technetium, and cesium concentration data for the spent fuel tests are given in Appendix A, Tables A.7, A.8, A.9, and A.10, respectively.

4.5 COMPARISON OF LEACH RESULTS

Comparison of the results from the different spent fuel and UO_2 systems with different leachants and at different temperatures indicates several noteworthy observations.

- The leaching characteristics of uranium from both UO_2 and spent fuel in brine have a very small temperature dependence over the range studied. Little more (if any) uranium was released in brine at 150°C than at 25°C.
- The leaching characteristics of both UO_2 and spent fuel in brine are nearly independent of time. Within the scatter of the data, most of the uranium is released in the first few days. This suggests that the uranium release is restricted by solubility limitations or that there exists a surface phase that is more readily soluble than the bulk material.
- Unlike brine, leaching of UO_2 in deionized water is dependent on temperature, at least when iron was not present. Uranium release values in deionized water, in the absence of iron, are higher at

150°C than at 75°C and higher at 75°C than at 25°C. The larger temperature dependence for deionized water than for brine is difficult to interpret on the basis of our present knowledge. Differences in UO_2 alteration products may be involved. For example, the existence of sodium uranates is likely in the brines but not possible with deionized water. Differences in ionic strength may also play a role.

- Considerably more uranium leaches from UO_2 in deionized water than in brine at 150°C; the difference is less pronounced at 25°C. In the absence of iron, the difference between the uranium released in deionized water and brine is small at 25°C but increases at 75°C. This trend continues at 150°C to the point where the release in deionized water is greater than the release in brine by more than one order of magnitude. When iron is present, there is little difference in the release of uranium between deionized water and brine at 25 and 75°C, although some difference prevails at 150°C.

- Over 100 times more uranium is leached from spent fuel in brine than is leached from UO_2 per unit of geometric surface area.

- The presence of iron coupons reduces solution concentrations of uranium; the uranium plates out on the iron and container walls or precipitates as filterable particles. Iron has no effect on the total uranium released in brine, but iron does cause a substantial reduction in solution concentration of uranium probably because it lowers the oxidation potential, thereby lowering uranium solubility. An alternative explanation could be that iron selectively sorbs multivalent ions. The same trend is true for plutonium and technetium in the spent fuel tests. Cesium, the only other element analyzed in the spent fuel tests, was not affected by iron.

  Oxidized Zircaloy coupons have no effect on UO_2 leaching characteristics.

- Final measured pH values listed in Appendix A, Tables A.1 through A.7, with few exceptions, are in the range 5.0 to 7.5. This is a

change of no more than about one pH unit from the starting values of both brine and deionized water. However, it should be noted for brine that the measured pH values are somewhat unreliable and can be in error by as much as one unit or sometimes slightly more than one unit.

4.6 ORGANIZATION OF TABLES AND FIGURES

The data that were discussed in Sections 4.1 through 4.5, with a couple of exceptions, are presented in this section in both tabular and graphical form. The tables and their corresponding figures, are listed below. This format was chosen to allow uninterrupted reading of the data analysis. This also places tables and related figures in closer proximity to each other, which allows the reader a better opportunity for data comparison.

Tables	Figures
4.1	4.1 - 4.5
4.2	4.6 - 4.10
4.3	4.11
4.4	none
4.5	4.12 - 4.16
4.6	4.17 - 4.21
none	4.22 - 4.23
4.7	4.24 - 4.28
4.8	4.29 - 4.33
4.9	4.34 - 4.38
4.10	4.39 - 4.43

TABLE 4.1. Uranium Released During UO_2 Tests at 25°C in Brine

Sample Type	No. Days	Uranium Released, mg/m^2					
			Plateout on				
		Leachate	Teflon	Iron	Zircaloy-4	Total	Filtrate
UO_2	2	12.5	<0.2	-	-	12.5	14.5
	5	10.5	1.9	-	-	12.4	10.0
	14	15.9	1.0	-	-	16.9	13.6
	28	19.4	1.0	-	-	20.4	19.4
	60	25.1	0.9	-	-	26.0	25.1
	60	21.7	0.9	-	-	22.6	20.5
	60	22.8	1.4	-	-	24.2	22.8
	60(avg)	23.2	1.1	-	-	24.3	22.8
UO_2-Fe	2	5.1	0.3	1.6	-	7.0	5.1
	5	3.8	1.4	7.9	-	13.1	3.8
	14	2.7	1.0	1.7	-	5.4	1.5
	28	2.4	1.0	17.3	-	20.7	1.9
	60	0.5	2.2	18.9	-	21.6	0.2
	60	1.4	0.8	23.7	-	25.9	0.3
	60	0.8	1.6	16.6	-	19.0	0.2
	60(avg)	0.9	1.5	19.7	-	22.1	0.2
UO_2-Zr	2	15.0	0.5	-	<0.1	15.5	11.0
	5	17.0	0.2	-	<0.1	17.2	17.0
	14	17.1	0.2	-	ND	17.3	17.1
	28	19.4	0.3	-	ND	19.7	19.4
	60	20.5	0.4	-	<0.1	20.9	20.5
	60	18.2	0.5	-	<0.1	18.7	18.2
	60	19.4	<0.5	-	<0.1	19.4	19.4
	60(avg)	19.4	0.3	-	<0.1	19.7	19.4
UO_2-Fe-Zr	2	5.4	<0.2	3.8	<0.1	9.2	5.0
	5	5.0	0.3	9.0	<0.1	14.3	4.4
	14	1.8	0.7	9.9	ND	12.4	1.6
	28	1.6	0.7	14.7	ND	17.0	0.8
	60	1.3	1.6	35.2	<0.1	38.1	0.7
	60	1.1	1.1	27.2	<0.1	29.4	0.7
	60	0.3	0.7	13.4	<0.1	14.4	0.3
	60(avg)	0.9	1.1	25.3	<0.1	27.3	0.6
Releach UO_2-Fe-Zr	14	1.2	<0.1	3.9	0.5	5.6	1.2

ND - not detected

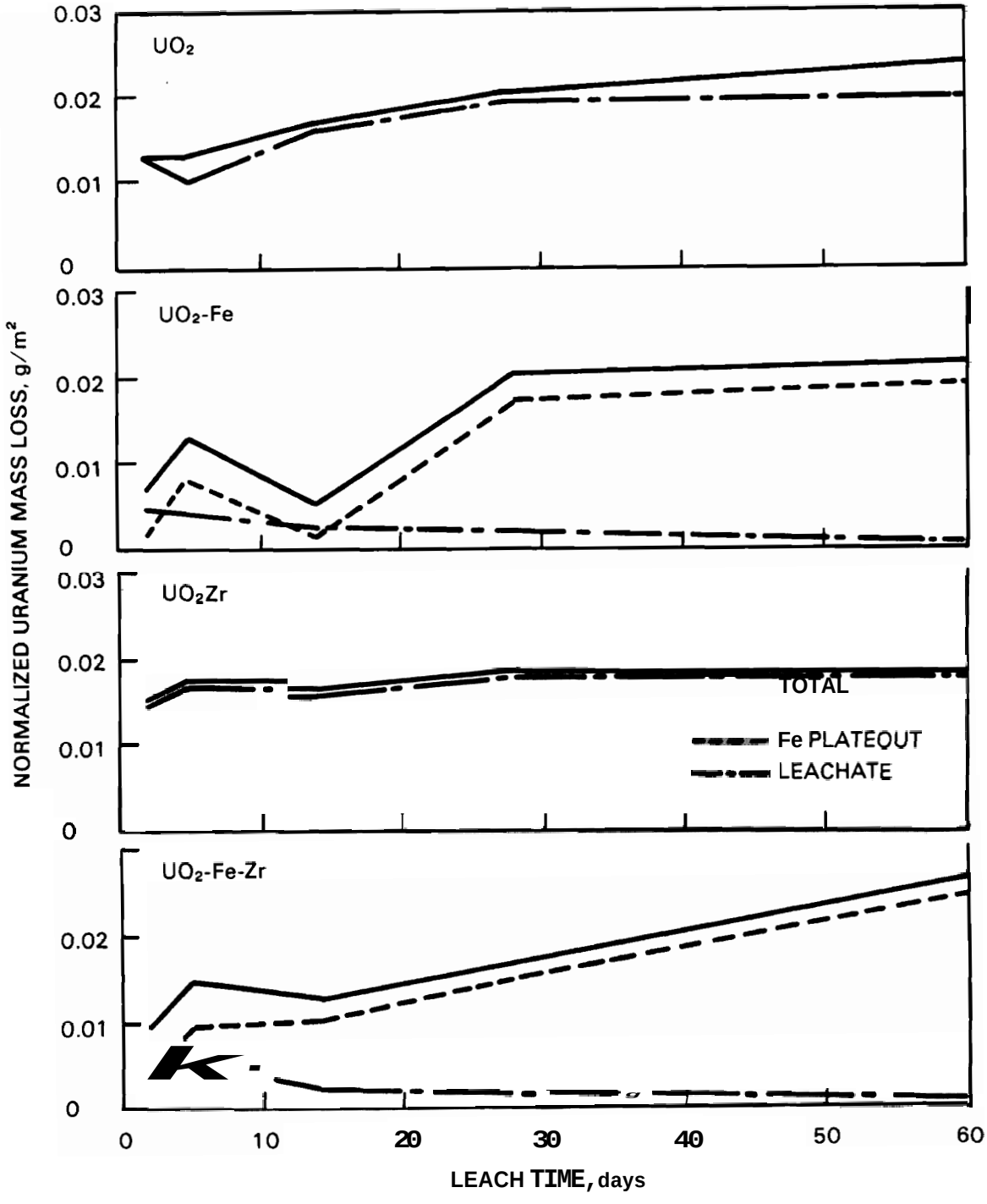

FIGURE 4.1. Comparison of Results Between Leach Systems Incorporating UO_2 in Brine at 25°C

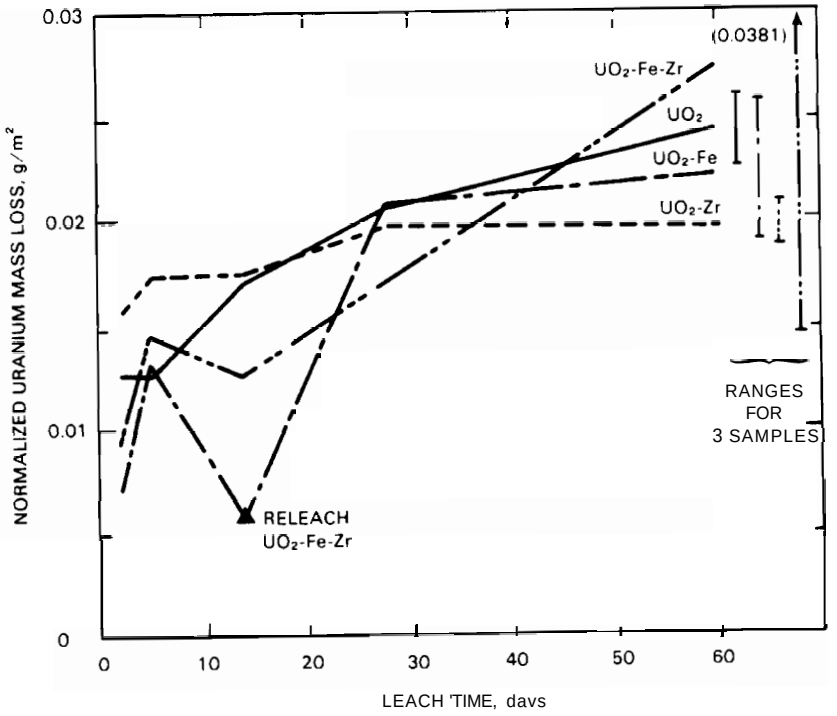

FIGURE 4.2. Total Uranium Released in UO_2 Systems at 25°C in Brine

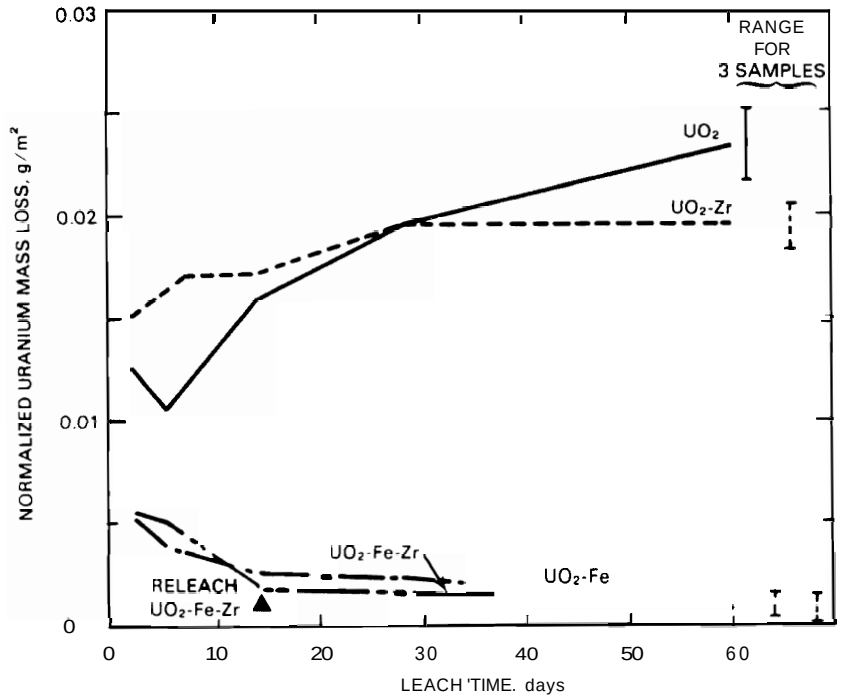

FIGURE 4.3. Uranium in Leachate in UO_2 Systems at 25°C in Brine

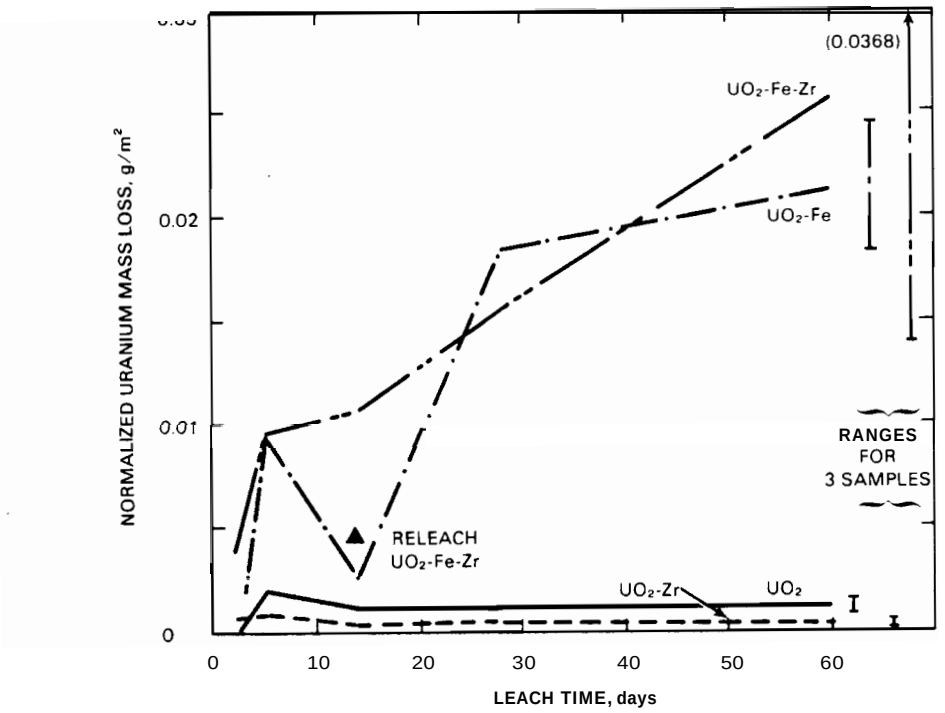

FIGURE **4.4.** Total Uranium Plateout in UO_2 Systems at 25°C in Brine

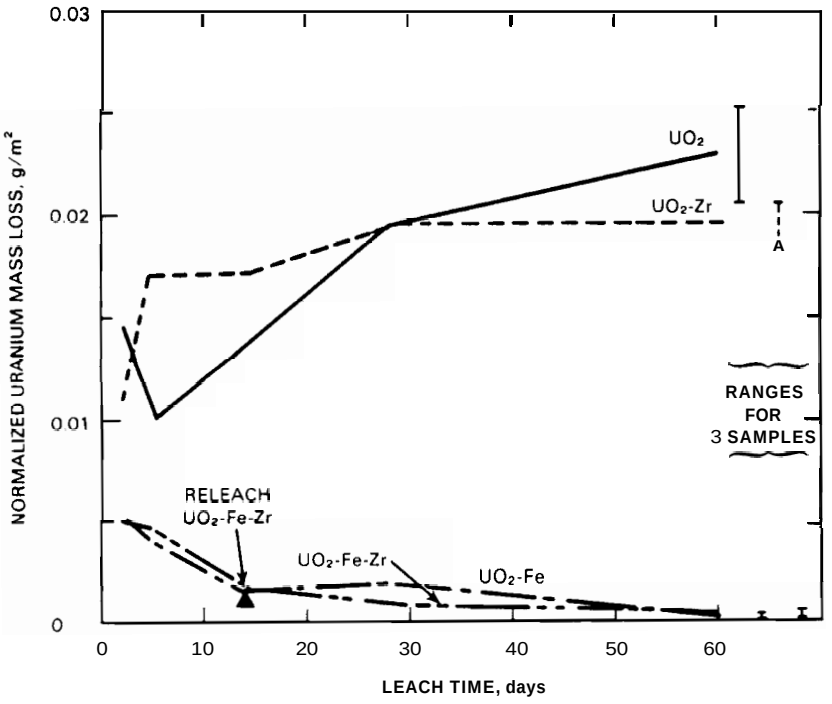

FIGURE 4.5. Uranium in Filtrate in UO_2 Systems at 25°C in Brine

TABLE 4.2. Uranium Released During UO_2 Tests at 75°C in Brine

Sample Type	No. Days	Uranium Released, mg/m2					
			Plateout on				
		Leachate	Teflon	Iron	Zircaloy-4	Total	Filtrate
UO_2	2	7.5	1.2	-	-	8.7	7.2
	5	10.5	9.3	-	-	19.8	9.4
	14	7.3	13.9	-	-	21.2	6.2
	28	6.6	15.5	-	-	22.1	6.4
	60	7.3	12.8	-	-	20.1	6.2
	60	7.8	9.3	-	-	17.1	7.5
	60	8.7	6.0	-	-	14.7	8.7
	60(avg)	7.9	9.4	-	-	17.3	7.5
UO_2-Fe	2	4.7	4.9	2.1	-	11.7	2.9
	5	1.8	20.5	4.4	-	26.7	0.4
	14	19.3	33.3	10.7	-	63.3	4.7
	28	4.3	10.6	2.7	-	17.6	<0.3
	60	3.8	10.7	6.4	-	20.9	<3
	60	16.0	13.3	12.8	-	42.1	1.1
	60	2.5	9.2	3.8	-	15.5	<0.3
	60(avg)	7.4	11.1	7.7	-	26.2	0.4
UO_2-Zr	2	15.0	0.5	-	<0.1	15.5	15.0
	5	11.0	0.9	-	0.1	12.0	11.0
	14	9.6	1.7	-	ND	11.3	9.7
	28	7.6	2.2	-	0.4	10.2	7.4
	60	8.3	1.7	-	0.2	10.2	8.3
	60	7.8	2.2	-	0.3	10.3	8.6
	60	10.4	1.0	-	0.3	11.7	9.3
	60(avg)	8.8	1.6	-	0.3	10.7	8.7
UO_2-Fe-Zr	2	3.5	2.7	2.1	<0.1	8.3	1.9
	5	5.9	3.3	2.7	<0.1	11.9	0.9
	14	5.0	7.7	5.1	ND	17.8	ND
	28	8.6	8.7	3.5	ND	20.8	ND
	60	2.6	3.2	9.0	<0.1	14.5	<0.3
	60	2.2	2.8	7.0	<0.1	12.0	<0.3
	60	4.1	3.0	6.1	<0.1	13.2	<0.3
	60(avg)	3.0	3.0	7.4	<0.3	13.2	<0.3
Releach UO_2-Fe-Zr	14	2.9	4.0	0.6	<0.1	7.5	0.4
UO_2-Oxidized Iron	14	4.0	6.0	0.2	-	10.2	ND
	14	4.7	9.2	0.2	-	14.1	ND

ND - not detected

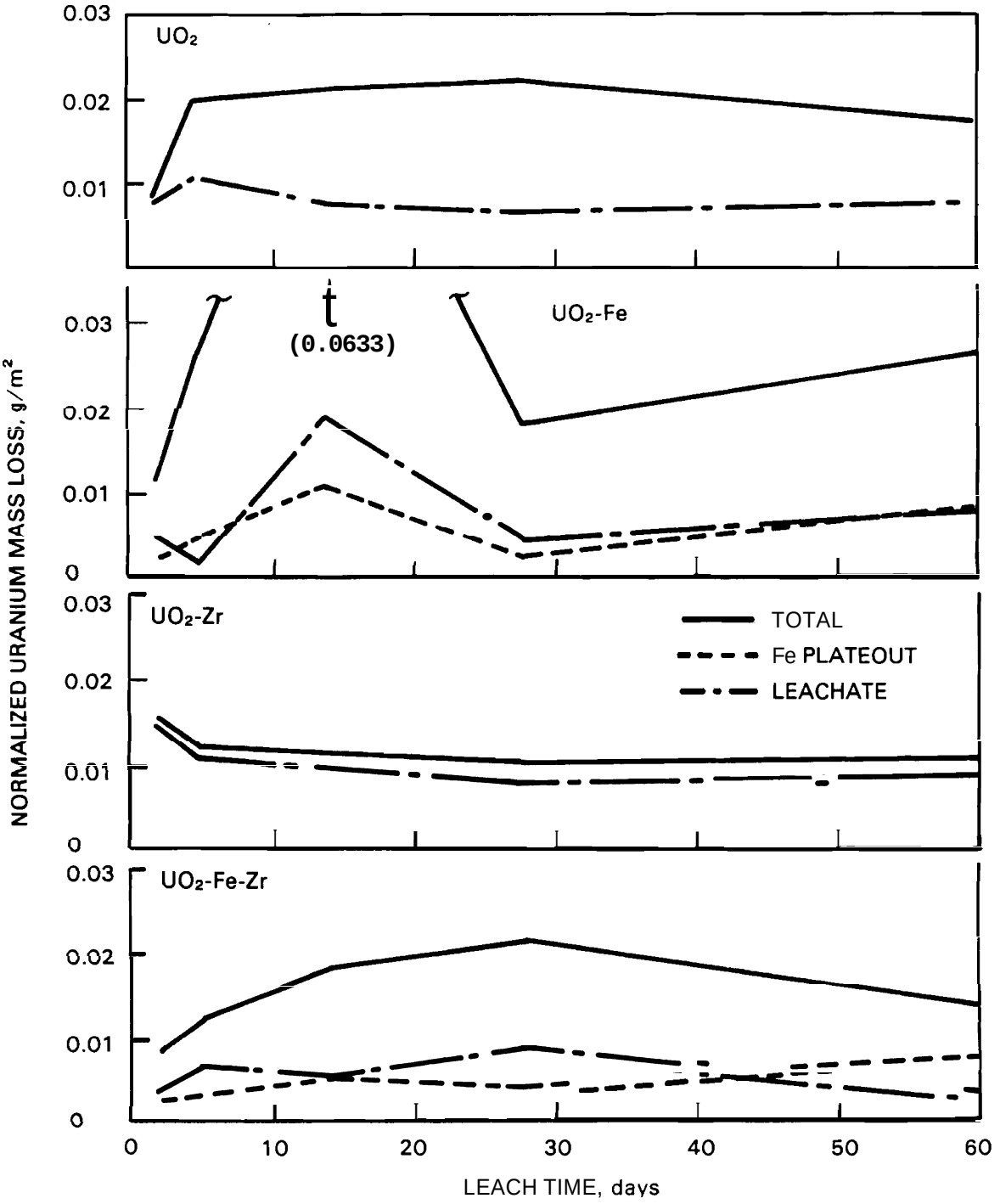

FIGURE 4.6. Comparison of Results Between Leach Systems Incorporating UO_2 at 75°C in Brine

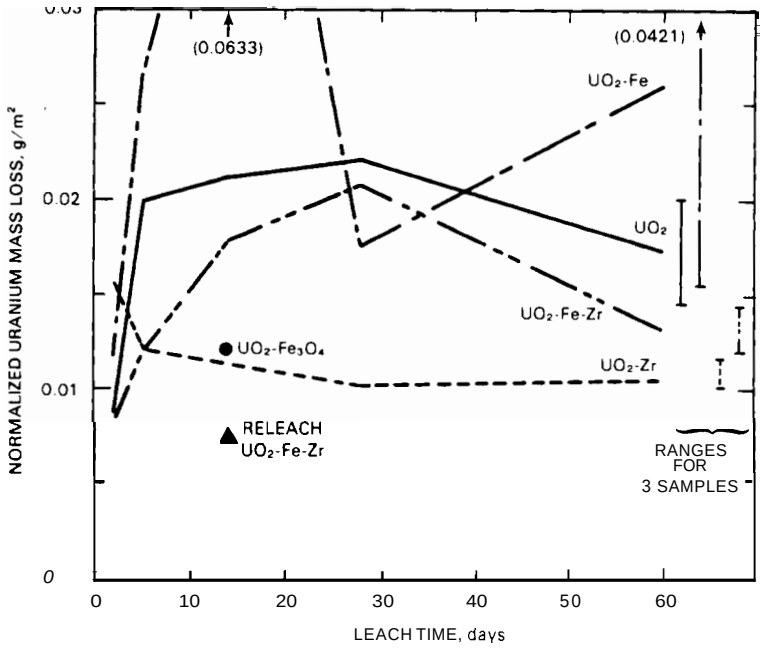

FIGURE 4.7. Total Uranium Released in UO_2 Systems at 75°C in Brine

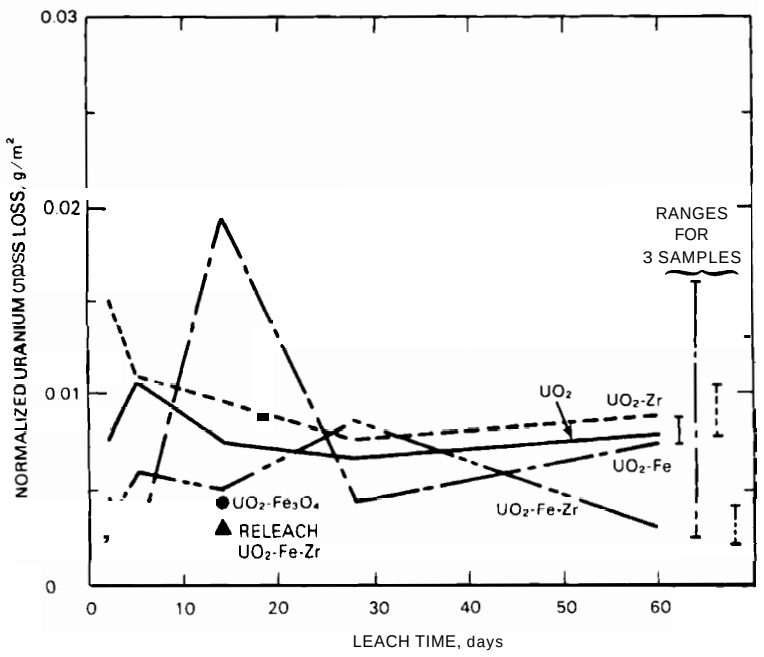

FIGURE 4.8. Uranium in Leachate in UO_2 Systems at 75°C in Brine

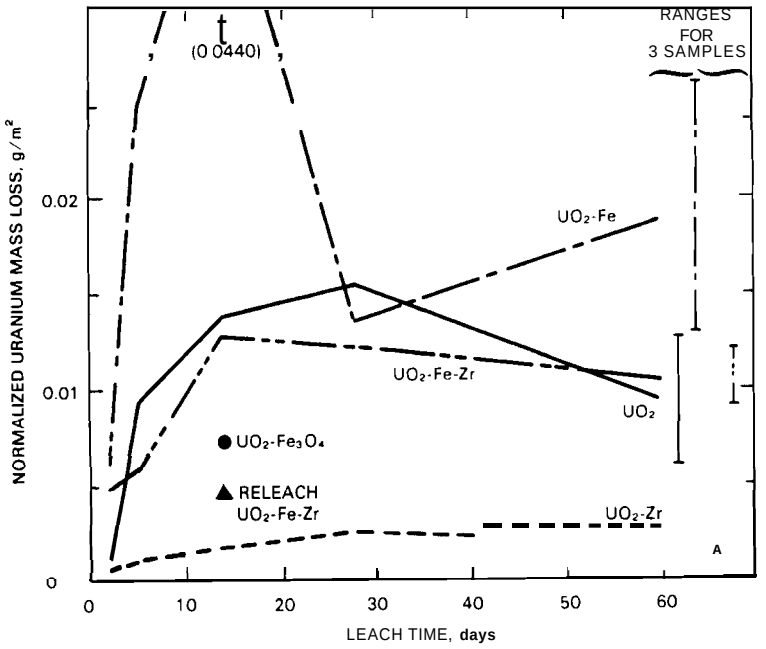

FIGURE 4.9. Total Uranium Plateout in UO_2 Systems at 75°C in Brine

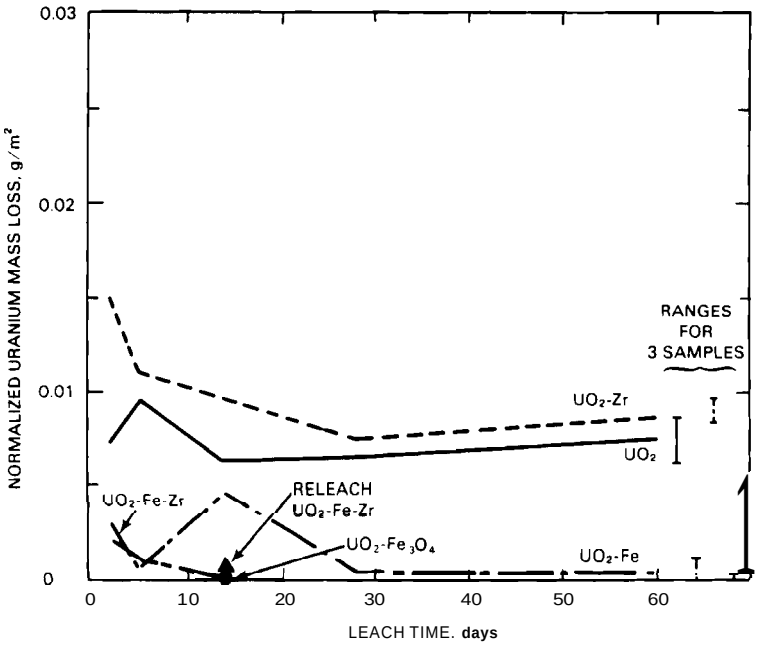

FIGURE 4.10. Uranium in Filtrate in UO_2 Systems at 75°C in Brine

TABLE 4.3. Uranium Released During UO_2 Tests in Deionized Water

Sample Type	No. Days	Uranium Released, mg/m2					
			Plateout on				
		Leachate	Teflon	Iron	Zircaloy-4	Total	Filtrate
25°C Tests							
UO_2	14	67.0	5.1	-	-	72.1	38.6
UO_2-Fe	14	4.1	4.4	10.1	-	18.6	0.2
UO_2-Zr	7	96.9	4.1	-	0.7	101.7	53.6
	14	94.6	2.8	-	1.4	98.8	63.8
UO_2-Fe-Zr	7	8.8	6.3	0.1	0.4	15.6	0.8
	14	18.2	2.7	1.4	0.3	22.6	0.1
75°C Tests							
UO_2	14	363.6	43.1	-	-	406.7	227.3
UO_2-Fe	14	9.4	41.7	9.6	-	60.7	0.3
UO_2-Zr	7	285.0	14.5	-	2.3	301.8	216.6
	14	501.6	9.6	-	2.4	513.6	558.6
UO_2-Fe-Zr	7	8.7	37.9	2.0	0.4	49.0	1.5
	14	(a)	137.9	13.4	1.3	-	0.8

(a) Sample precipitated.

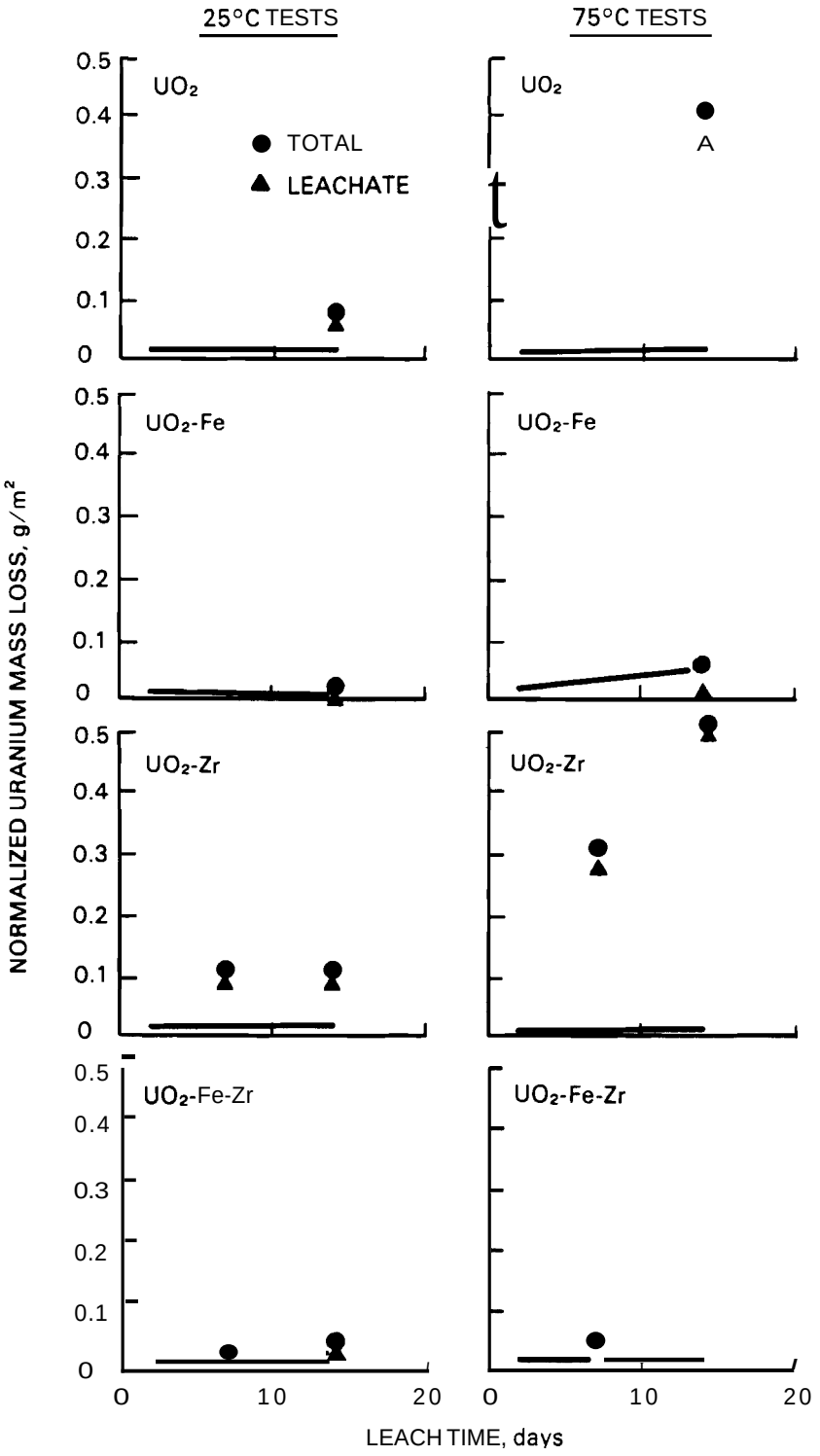

FIGURE 4.11. Comparison of Results Between Leach Systems Incorporating UO_2 at 25 and 75°C in Deionized Water (lines represent total released in brine; points are deionized water results)

TABLE 4.4. Results of Selected Iron Analyses from UO_2 Tests in Brine at 25 and 75°C

Test Type	Temperature, °C	Time, d	Iron Content, mg/L			
			Leachate	Filtrate	Teflon Plateout	Zircaloy-4 Plateout
UO_2	25	28	0.1	0.1	-	-
	75	28	<0.1	0.6	-	-
UO_2-Zr	25	14	0.1	0.2	-	0.38
	75	14	0.1	0.2	-	0.05
UO_2-Fe	25	5	-	-	4	-
		14	-	-	7	-
		28	8	<0.1	10	-
UO_2-Fe	75	5	-	-	30	-
		14	-	-	33	-
		28	19	0.6	18	-
UO_2-Fe-Zr	25	14	11	<0.1	-	0.55
	75	14	42	0.2	-	0.23
UO_2-Fe_3O_4	75	14	37	0.1	-	-

TABLE 4.5. Uranium Released from UO_2 during Tests at 150°C in Brine

Sample Type	No. Days	Uranium Released, mg/m^2 (a)					
			Plateout on				
		Leachate	Teflon	Iron	Zircaloy-4	Total	Filtrate
UO_2	7	4.5	17.0	-	-	21.5	4.5
	15	4.5	43.0	-	-	47.5	3.4
	28	3.4	(107.0)	-	-	(110.4)	2.3
	28	10.2	3.4	-	-	13.6	4.5
	60	2.3	0.9	-	-	3.2	ND
	60	(193.0)	1.0	-	-	(194.0)	11.3
UO_2-Fe	7	9.0	0.3	30.0	-	39.3	3.4
	14	4.5	ND	24.0	-	28.5	2.2
	28	0.5	ND	18.	-	18.5	<0.1
	28	1.0	65.0	4.0	-	70.0	<0.1
	62	2.0	7.0	22.0	-	31.0	<0.1
UO_2-Zr	7	3.4	10.2	-	3.5	I7.1	3.4
	14	6.8	2.2	-	0.4	9.4	4.5
	28	0.9	.3	-	-	1.2	0.8
	28	15.0	0.5	-	-	15.5	9.1
	60	1.0	ND	-	-	1.0	1.0

(a) Values in parentheses are apparently in error and have been deleted from data plots.
ND - not detected

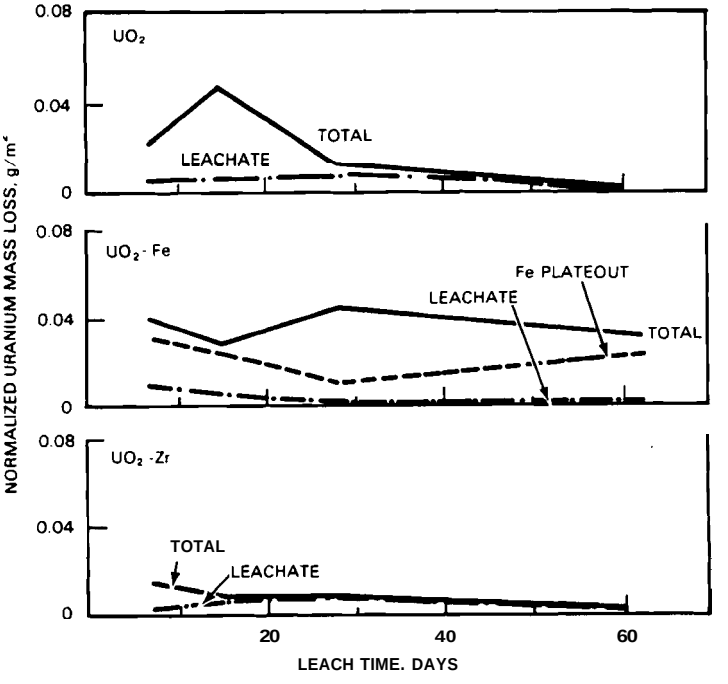

FIGURE 4.12. Comparison of Results Between Leach Systems Incorporating UO_2 at 150°C in Brine

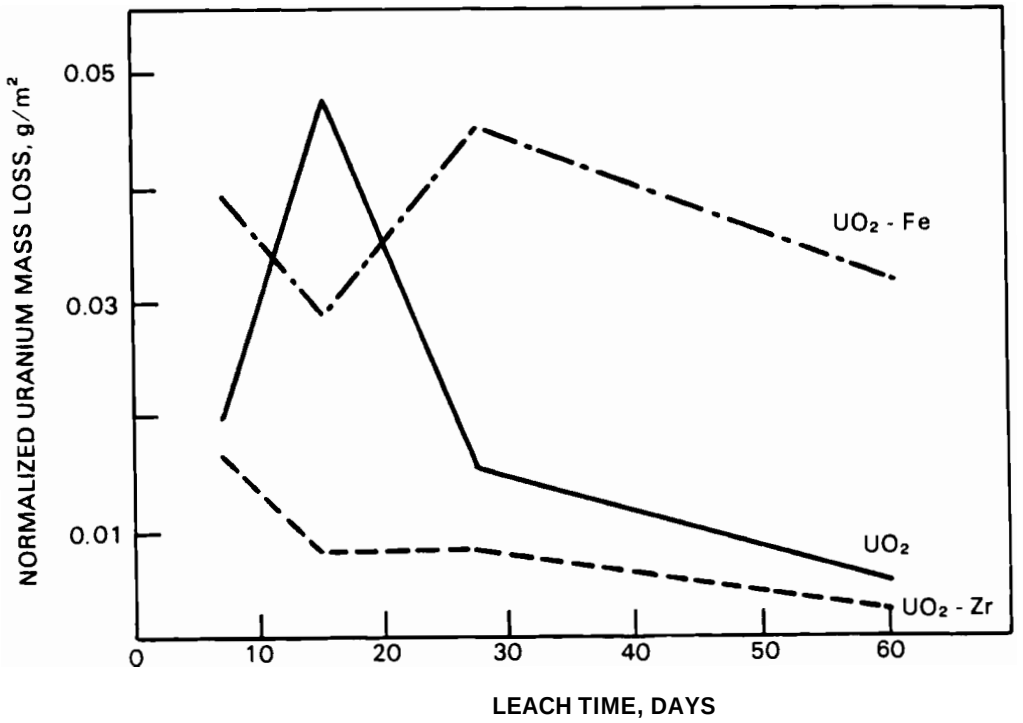

FIGURE 4.13. Total Uranium Released in UO_2 Systems at 150°C in Brine

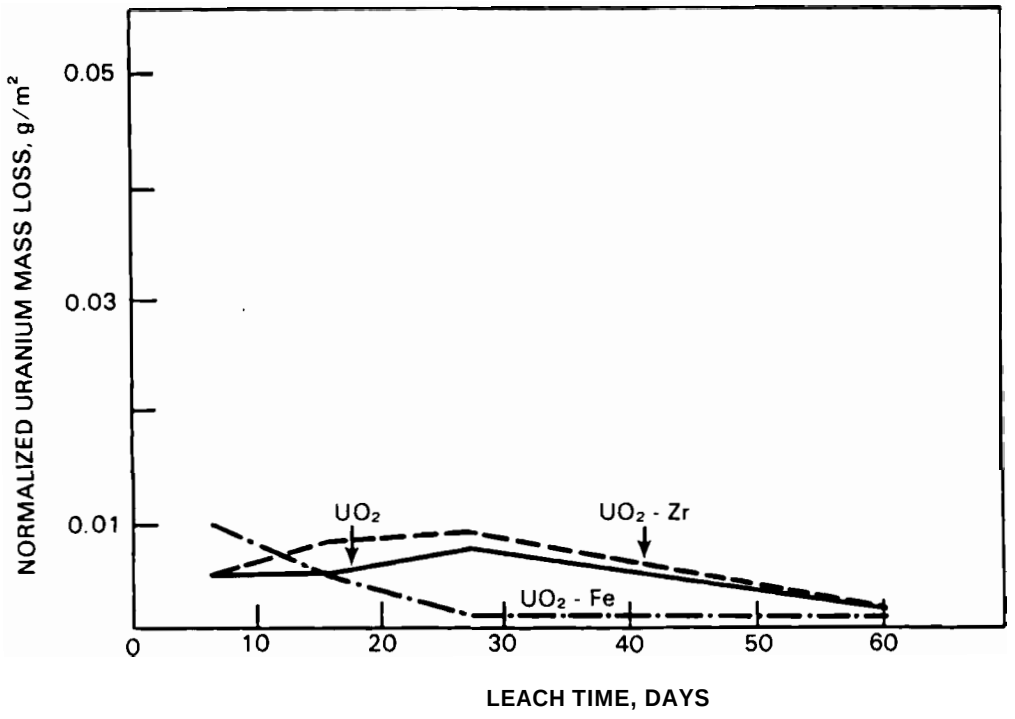

FIGURE 4.14. Uranium in Leachate in UO_2 Systems at 150°C in Brine

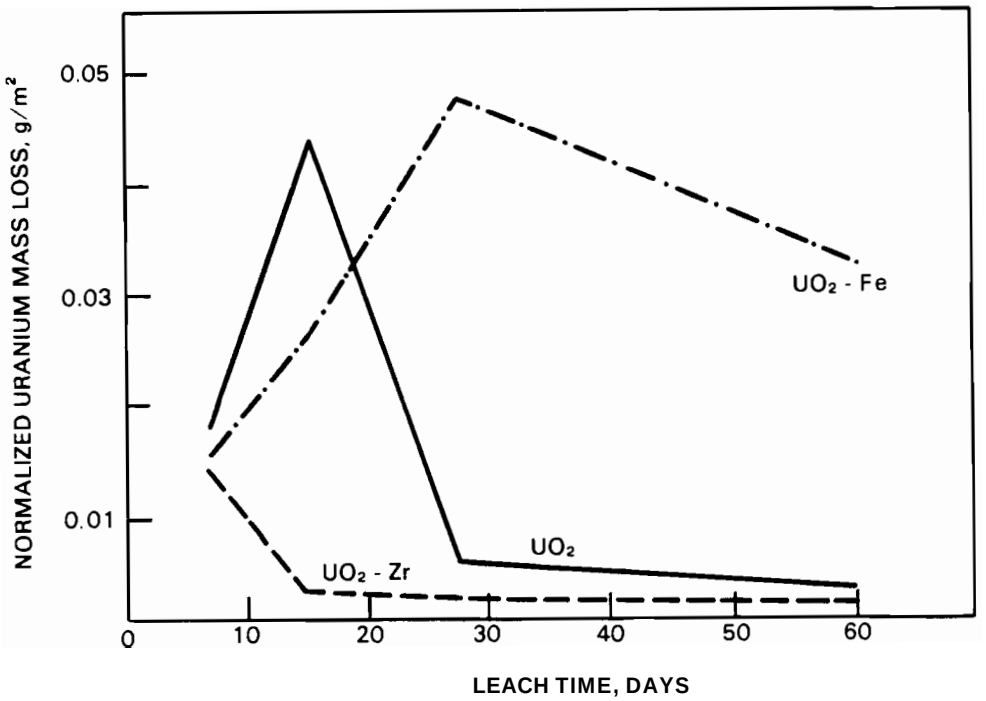

FIGURE 4.15. Total Uranium Plateout in UO_2 Systems at 150°C in Brine

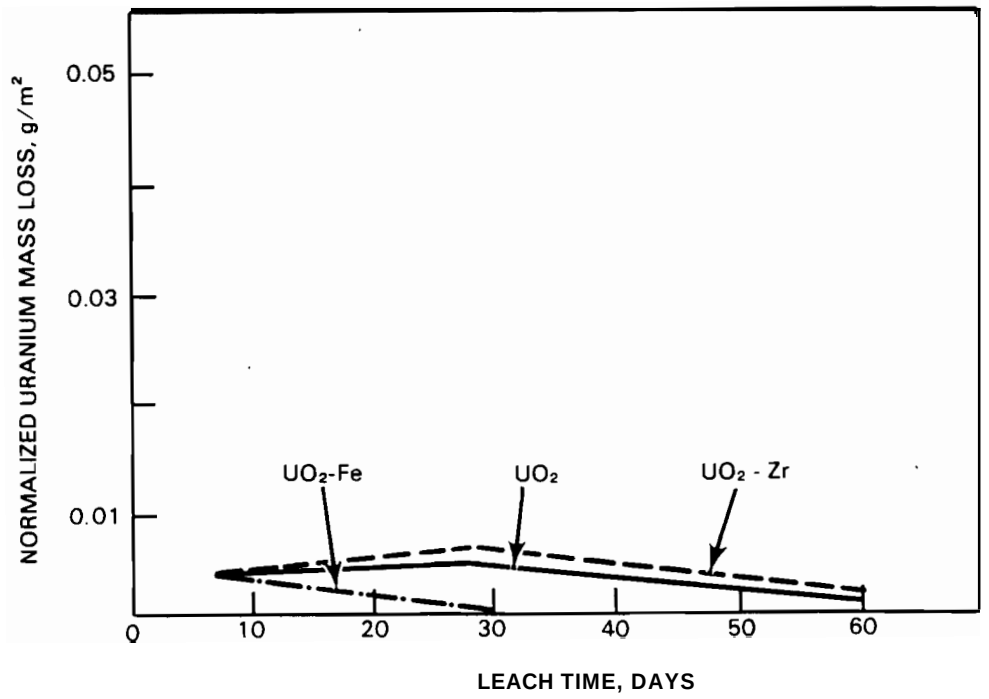

FIGURE 4.16. Uranium in Filtrate in UO_2 Systems at 150°C in Brine

TABLE 4.6. Uranium Released from UO_2 During Tests at 150°C in Deionized Water

Sample Type	No. Days	Uranium Released, mg/m^2					
			Plateout on				
		Leachate	Teflon	Iron	Zircaloy-4	Total	Filtrate
UO_2	7	580	670	-	-	1250	341
	15	227	1364	-	-	1591	102
	28	318	102	-	-	420	159
	28	136	523	-	-	659	91
	60	216	102	-	-	318	114
	60	330	91	-	-	421	227
UO_2-Fe	7	5	2	26	-	33	0.3
	14	40	6	94	-	140	1.4
	28	2	9	190	-	201	0.2
	28	102	9	56	-	167	0.1
	60	11	20	54	-	85	0.1
UO_2-Zr	(Data apparently not reliable, See Appendix A).						

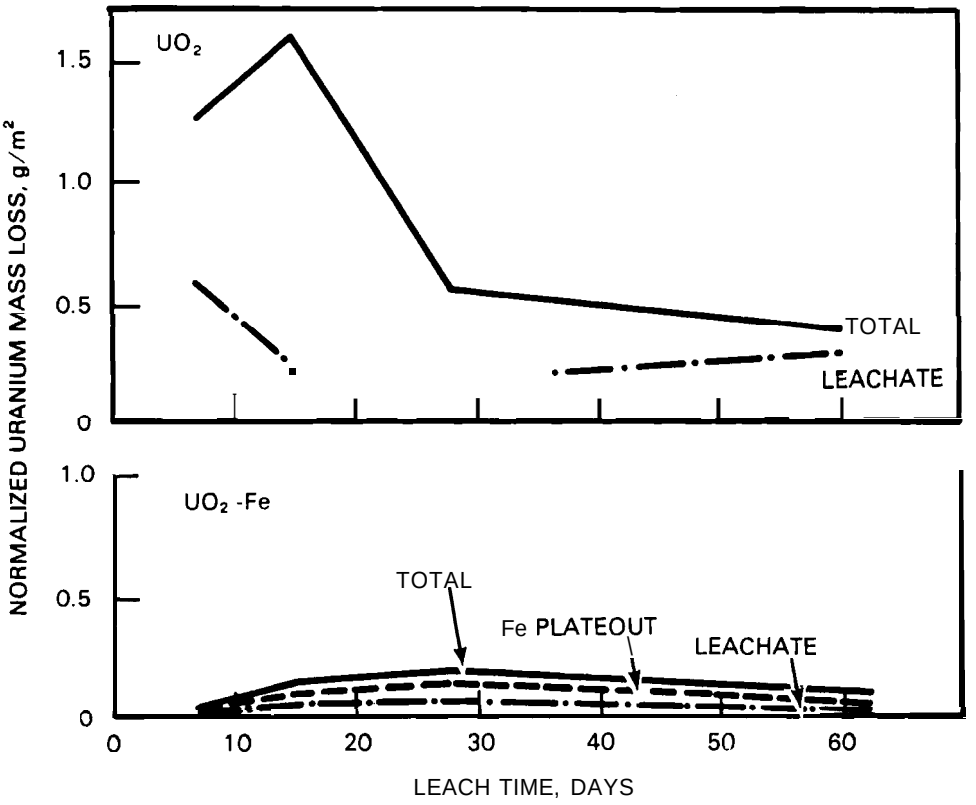

FIGURE 4.17. Comparison of Results Between Leach Systems Incorporating UO_2 at 150°C in Deionized Water

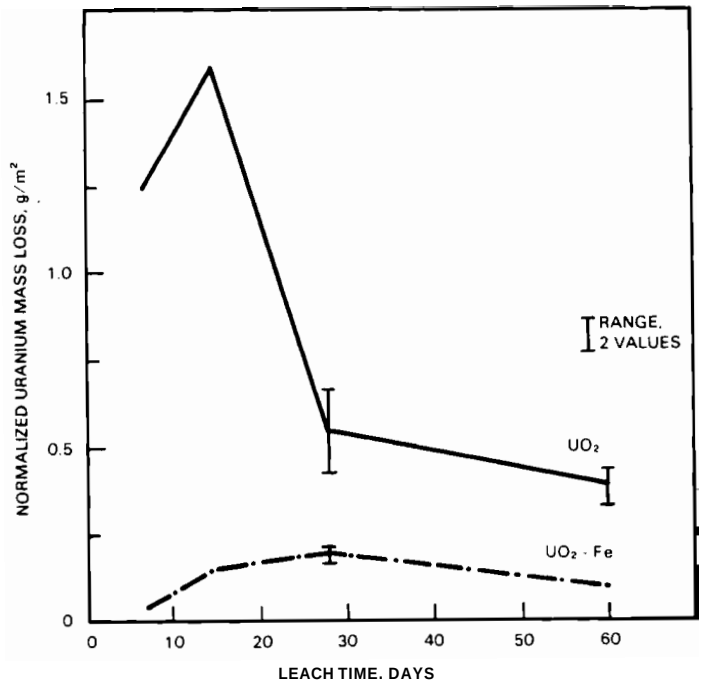

FIGURE 4.18. Total Uranium Released in UO_2 Systems at 150°C in Deionized Water

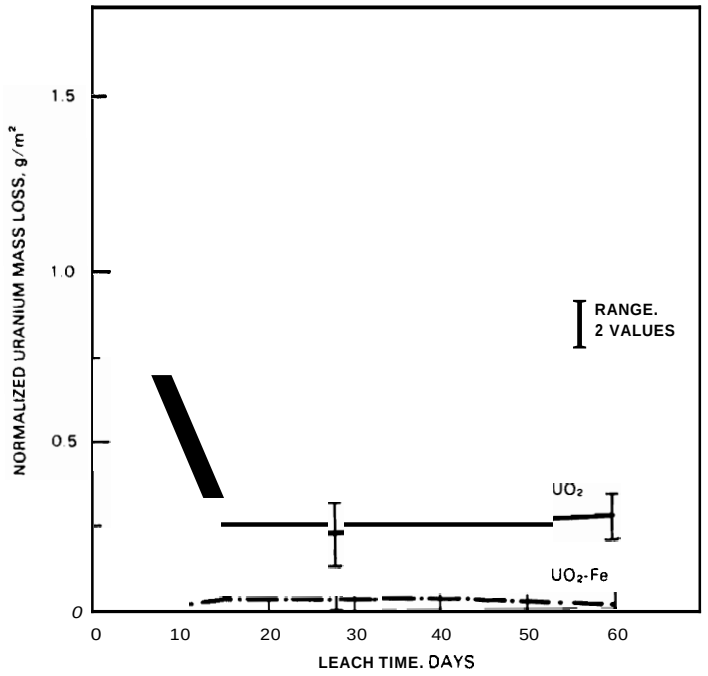

FIGURE 4.19. Uranium in Leachate in UO_2 Systems at 150°C in Deionized Water

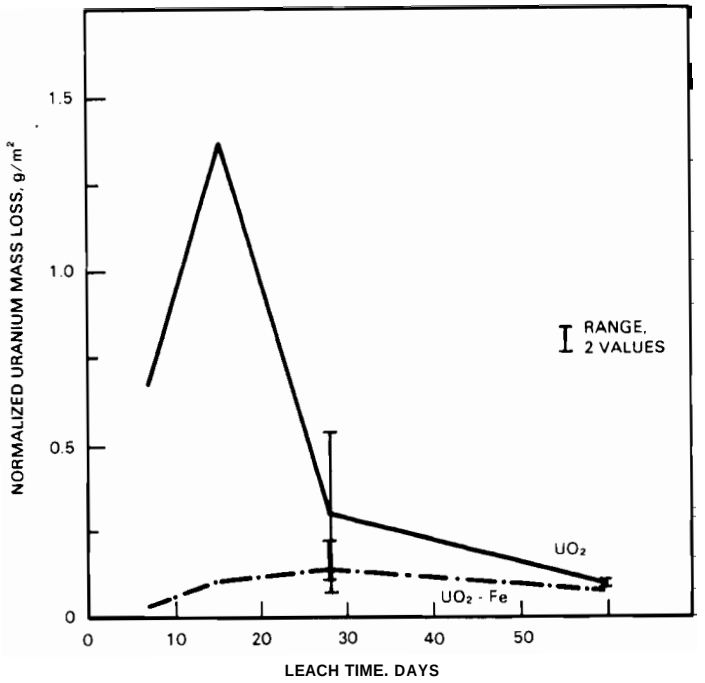

FIGURE 4.20. Total Uranium Plateout in UO_2 Systems at 150°C in Deionized Water

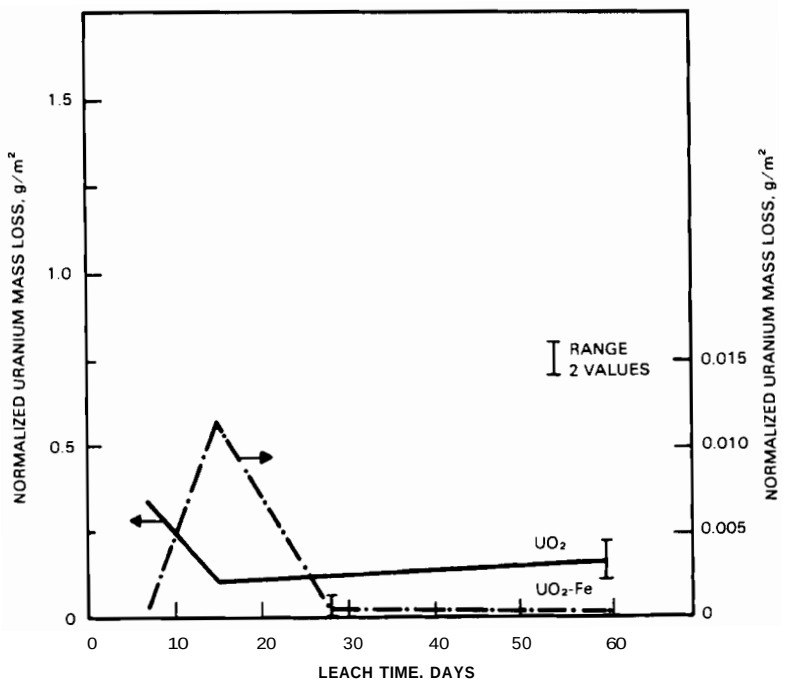

FIGURE 4.21. Uranium in Filtrate in UO_2 Systems at 150°C in Deionized Water. Note the different left and right scales which apply to the two different curves.

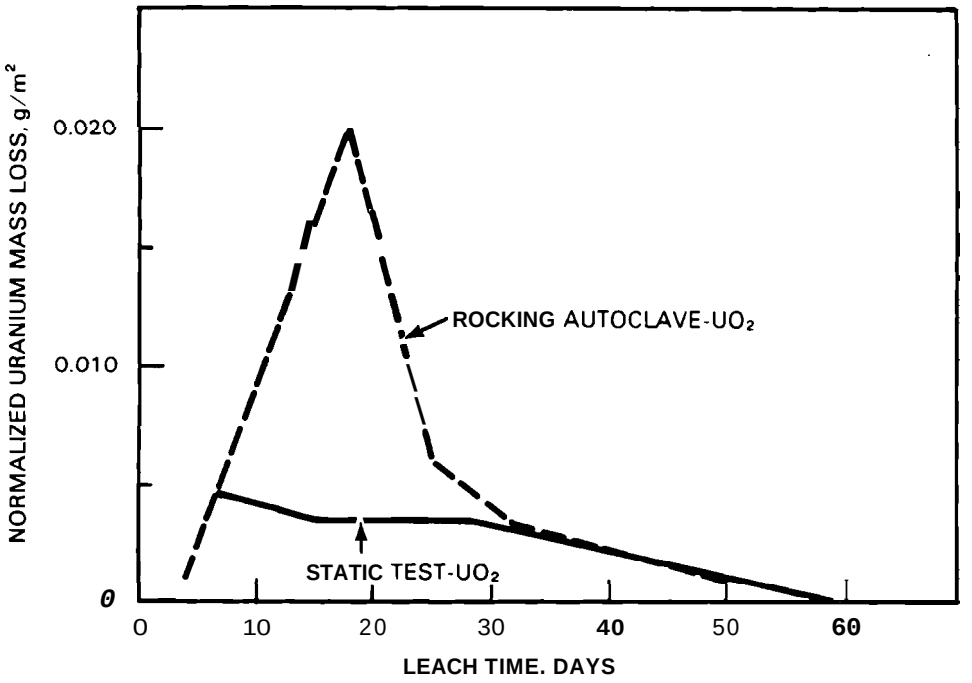

FIGURE 4.22. Comparison of Uranium in Filtrate for Static and Rocking Autoclave Tests at 150°C in Brine

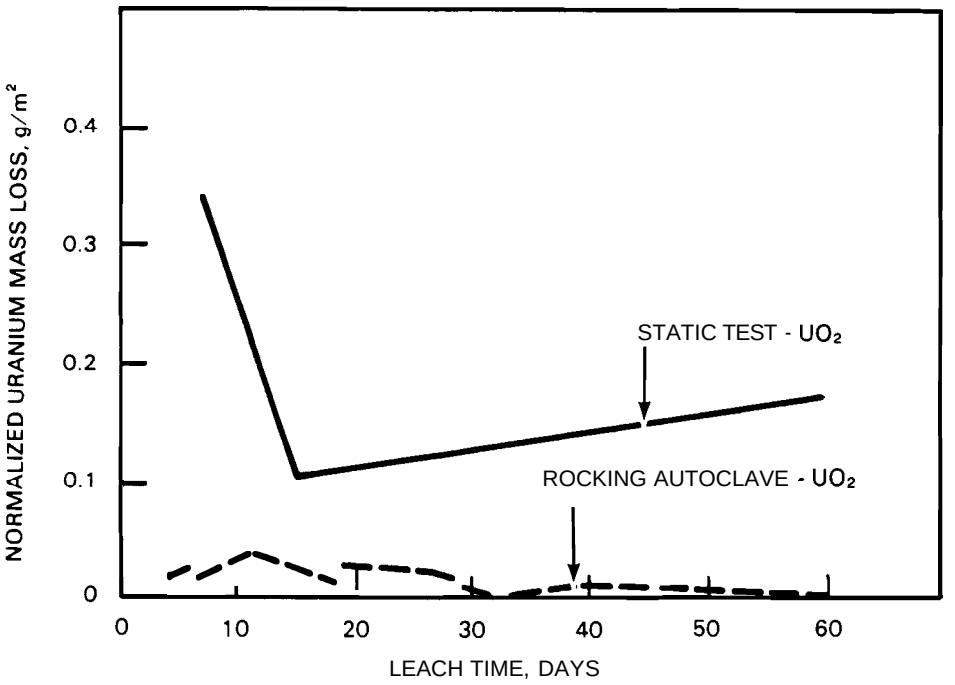

FIGURE 4.23. Comparison of Uranium in Filtrate for Static and Rocking Autoclave Tests at 150°C in Deionized Water

TABLE 4.7. Uranium Released from Spent Fuel (SF) in Synthetic and Real Brine

Sample Type	No. Days	Uranium Released, g/m^2				
		Leachate	Plateout on Quartz	Plateout on Iron	Total	Filtrate
----------25°C Tests----------						
Synthetic Brine						
	2	1.66	0.226	-	1.89	1.57
	5	1.88	0.131	-	2.01	1.70
	14	2.04	0.298	-	2.34	1.85
	28	3.29	0.262	-	3.55	3.08
	60	3.13	0.488	-	3.62	2.79
SF-Fe	2	1.56	3.96	0.88	6.40	0.095
	5	1.51	0.595	0.928	3.03	1.06
	14	1.53	0.167	1.70	3.40	0.298
	28	1.06	0.226	2.51	3.80	0.214
	60	1.54	0.619	5.69	7.85	0.107
Real Brine[a]						
SF	14	2.41	0.702	-	3.11	1.96
----------75°C Tests----------						
Synthetic Brine						
SF	5	3.19	1.32	-	4.5	2.55
	28	0.365	0.845	-	1.2	0.036
SF-Fe	5	1.42	3.92	0.299	5.6	0.083
	28	1.78	2.17	0.383	4.3	0.115

(a) Obtained by preparing a saturated solution of salt cored from the Permian Basin.

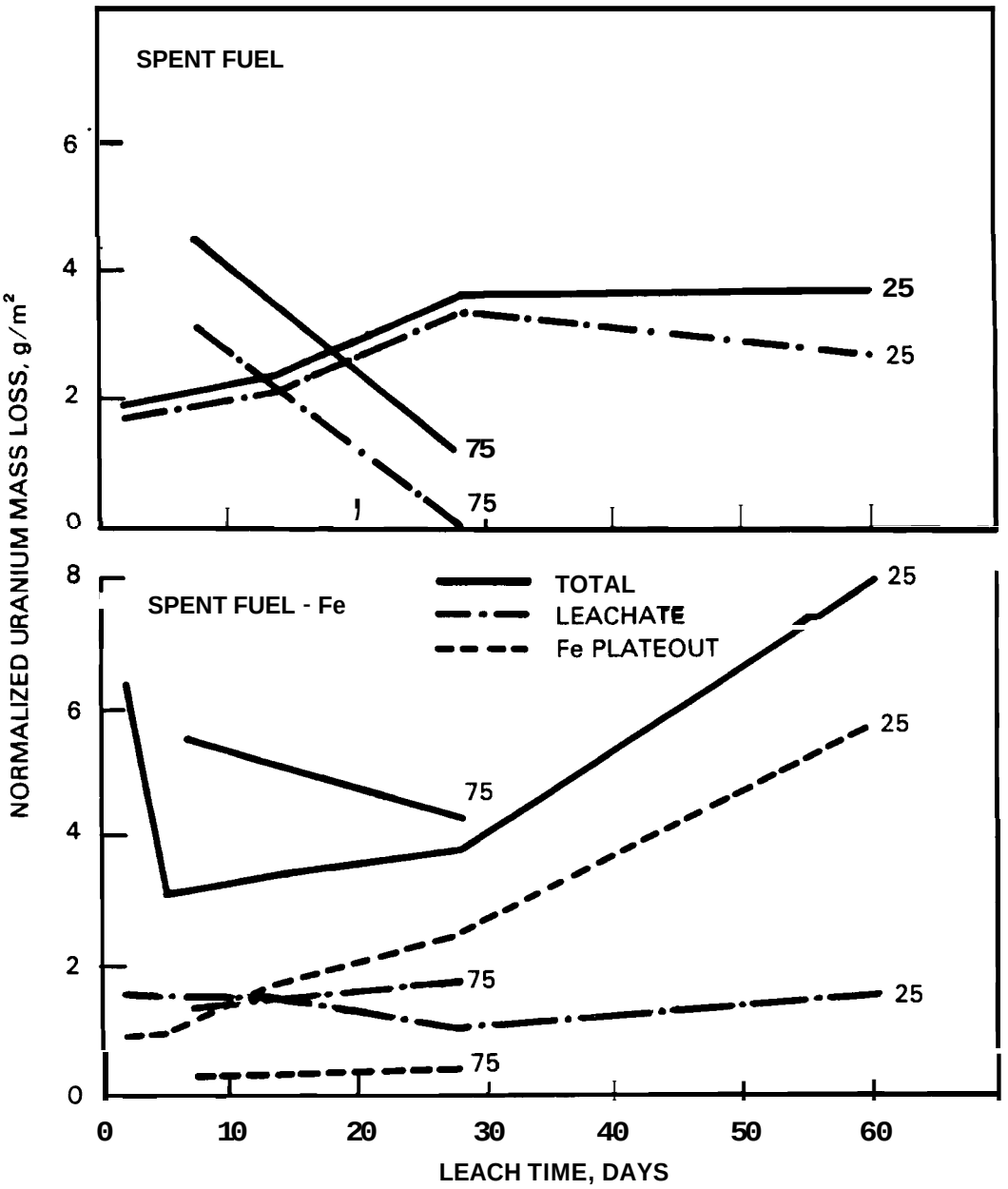

FIGURE 4.24. Comparison of Uranium Released in Systems Incorporating Spent Fuel and Brine at 25 and 75°C

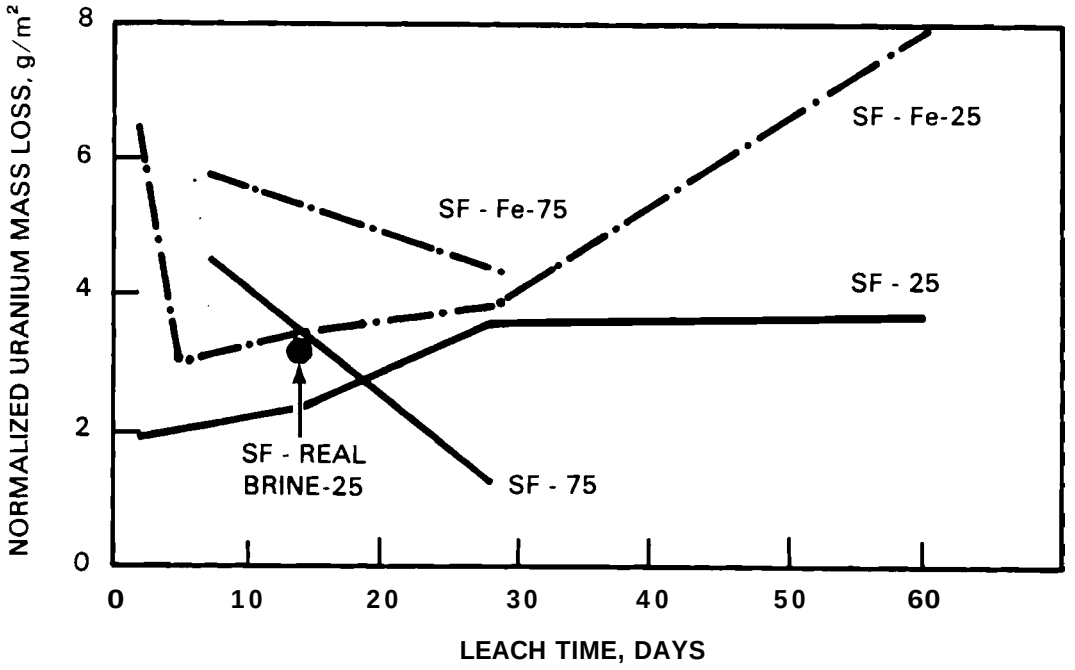

FIGURE 4.25. Total Uranium Released in Spent Fuel Systems at 25 and 75°C in Synthetic and Real Brine (obtained by preparing a saturated solution of salt cored from the Permian Basin)

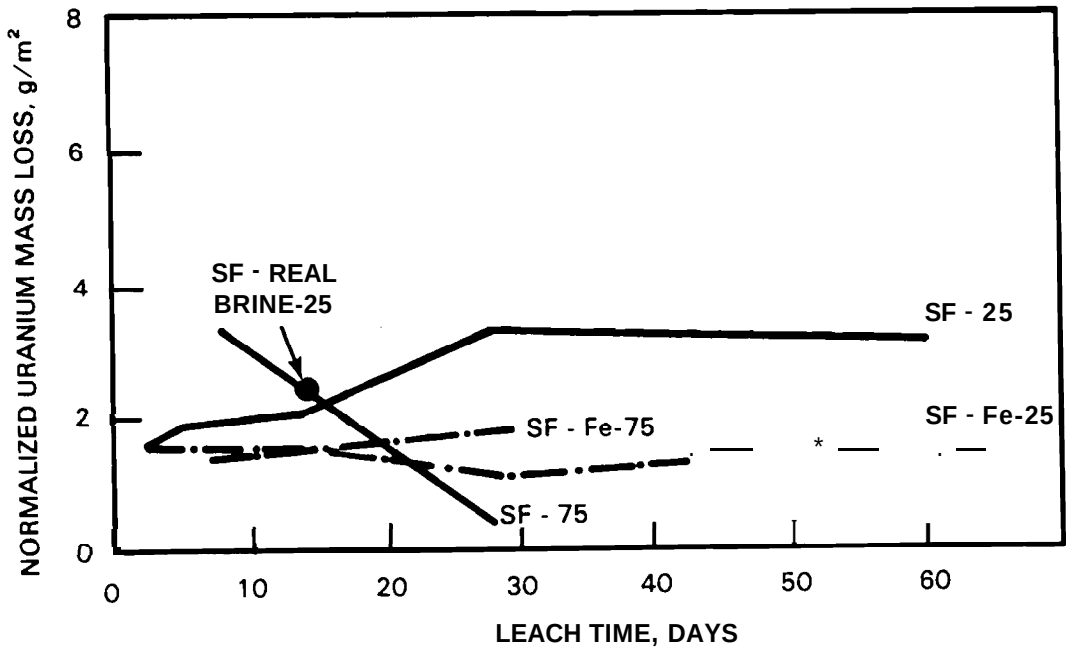

FIGURE 4.26. Uranium in Leachate in Spent Fuel Systems at 25 and 75°C in Synthetic and Real Brine (obtained by preparing a saturated solution of salt cored from the Permian Basin)

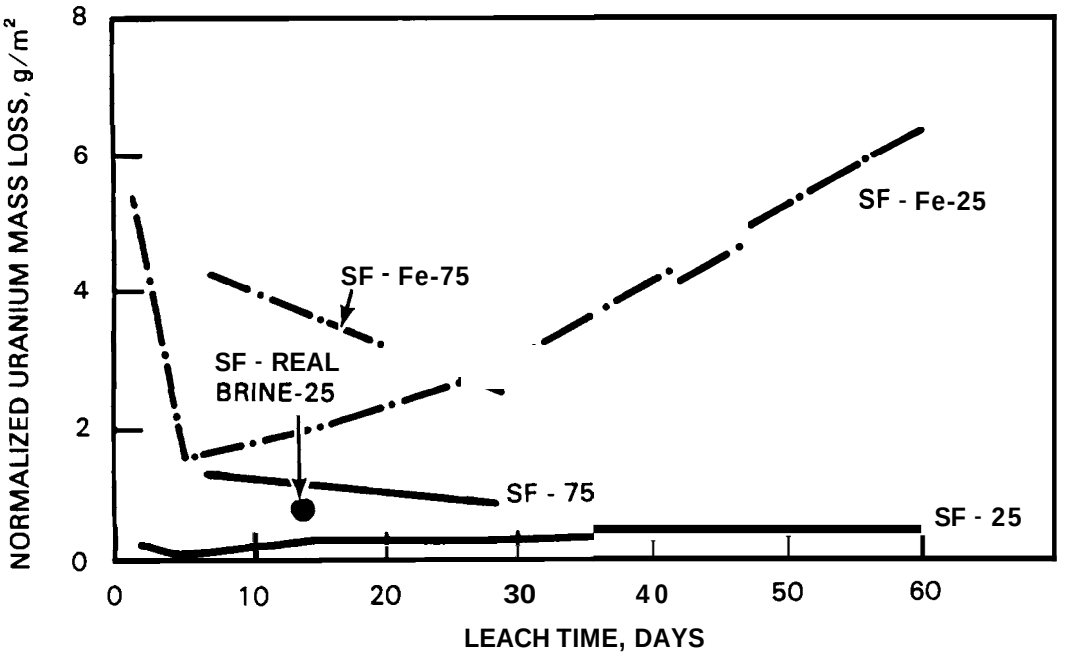

FIGURE 4.27. Total Uranium Plateout in Spent Fuel Systems at 25 and 75°C in Synthetic and Real Brine (obtained by preparing a saturated solution of salt cored from the Permian Basin)

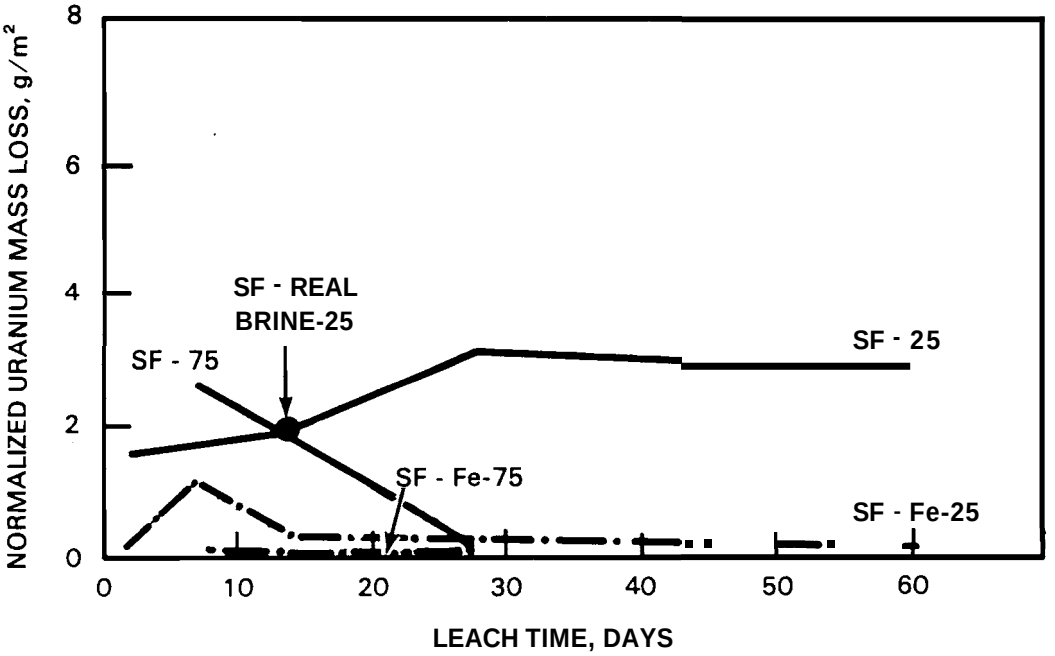

FIGURE 4.28. Uranium in Filtrate in Spent Fuel Systems at 25 and 75°C in Synthetic and Real Brine (obtained by preparing a saturated solution of salt cored from the Permian Basin)

TABLE 4.8. Plutonium Released from Spent Fuel (SF) in Synthetic and Real Brine

Sample Type	No. Days	Plutonium Released, g/m^2				
		Leachate	Plateout on Quartz	Plateout on Iron	Total	Filtrate
25°C Tests						
Synthetic Brine						
SF	2	0.1510	0.0201	-	0.171	0.1074
	5	0.0781	0.0631	-	0.141	0.0275
	14	0.1407	0.0275	-	0.168	0.0416
	28	0.1430	0.235	-	0.378	0.043
	60	0.1200	0.168	-	0.288	0.0872
SF-Fe	2	0.0692	0.094	0.016	0.179	0.0035
	5	0.112	0.262	0.01	0.384	0.002
	14	0.166	0.060	0.012	0.238	0.002
	28	0.1475	0.0805	0.024	0.252	0.0015
	60	0.631	0.443	0.419	1.493	0.002
Real Brine[a]						
SF	14	0.1567	0.356	-	0.513	0.0631
75°C Tests						
Synthetic Brine						
SF	5	0.43	0.154	-	0.6	0.362
	28	0.100	0.416	-	0.5	0.011
SF-Fe	5	0.273	NA	0.021	NA	0.003
	28	0.310	0.235	0.033	0.6	0.004

(a) Obtained by preparing a saturated solution of salt cored from the Permian Basin.

NA - not analyzed

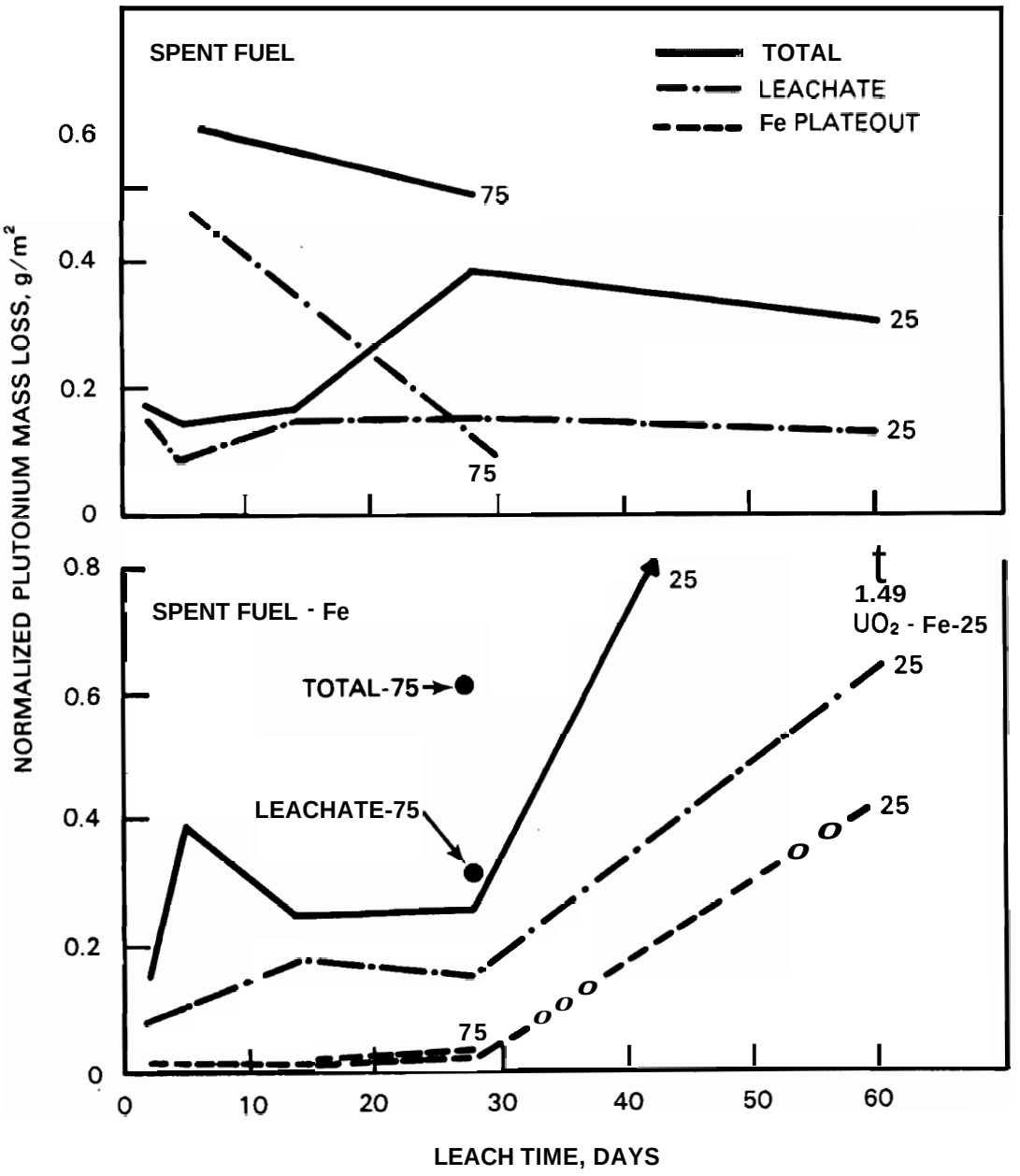

FIGURE 4.29. Comparison of Plutonium Released in Systems Incorporating Spent Fuel and Brine at 25 and 75°C

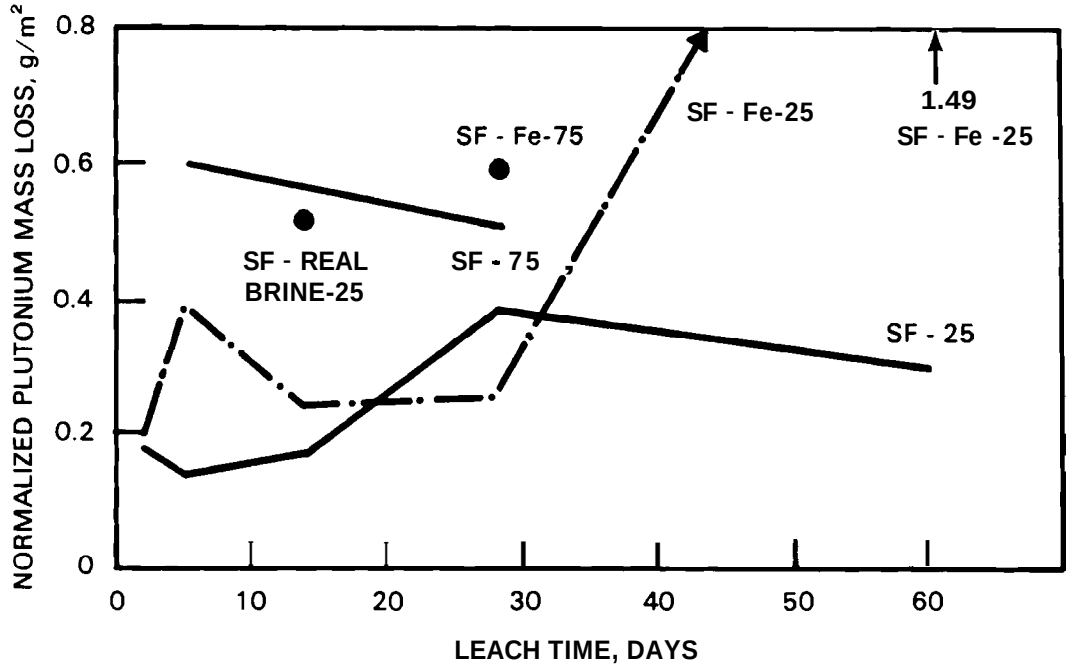

FIGURE 4.30. Total Plutonium Released in Spent Fuel Systems at 25 and 75°C in Synthetic and Real Brine (obtained by preparing a saturated solution of salt cored from the Permian Basin)

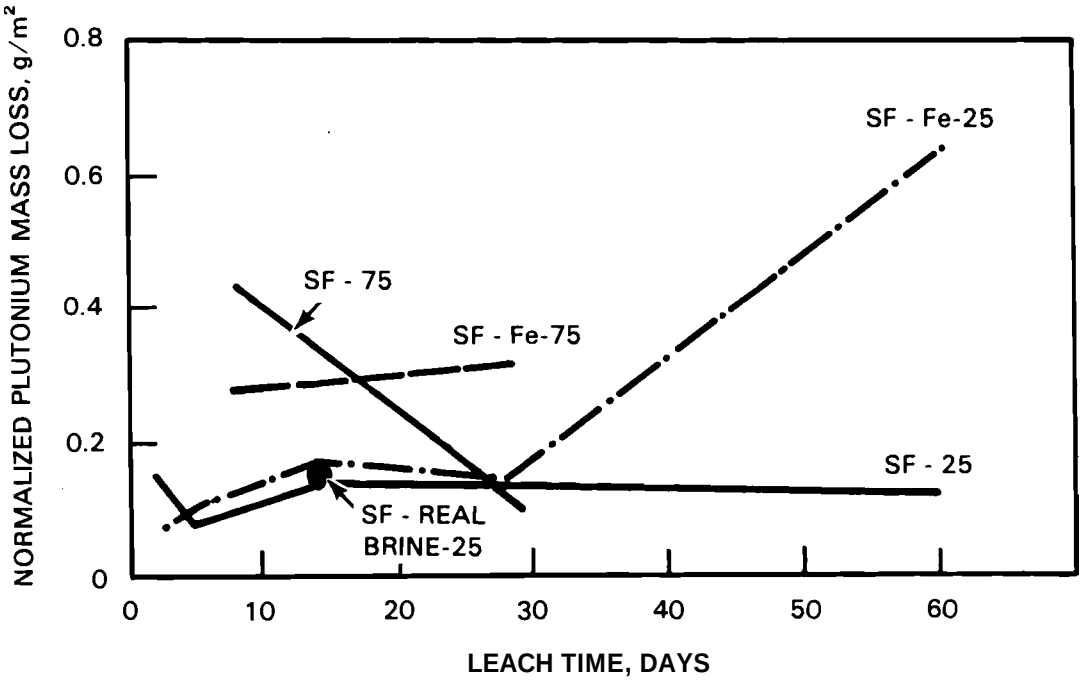

FIGURE 4.31. Plutonium in Leachate in Spent Fuel Systems at 25 and 75°C in Synthetic and Real Brine (obtained by preparing a saturated solution of salt cored from the Permian Basin)

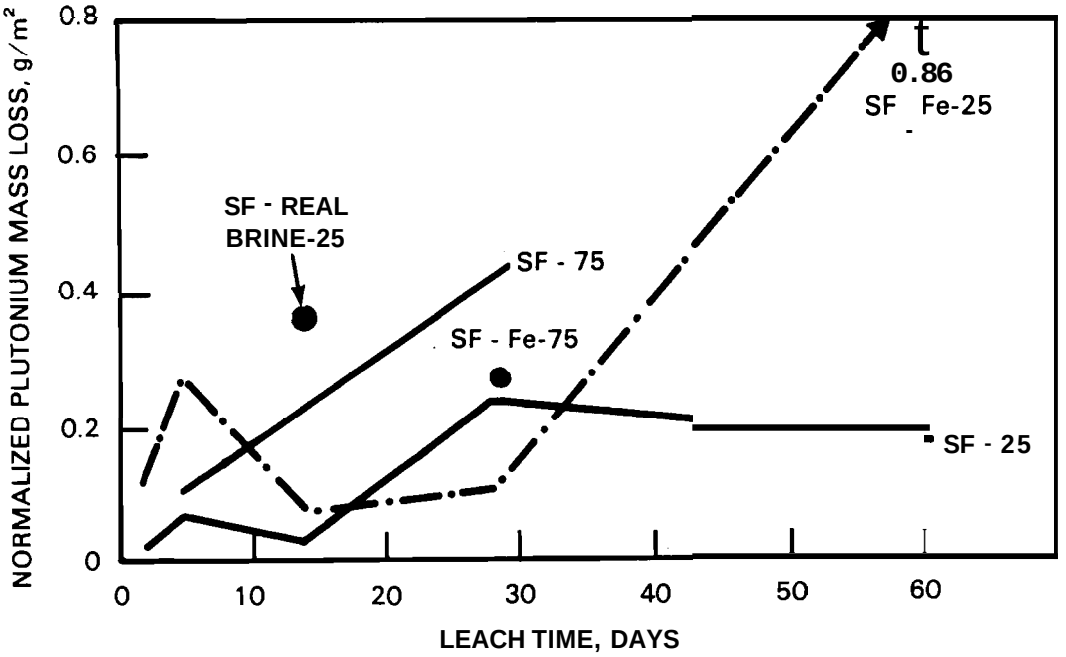

FIGURE 4.32. Plutonium Plateout in Spent Fuel Systems at 25 and 75°C in Synthetic and Real Brine (obtained by preparing a saturated solution of salt cored from the Permian Basin)

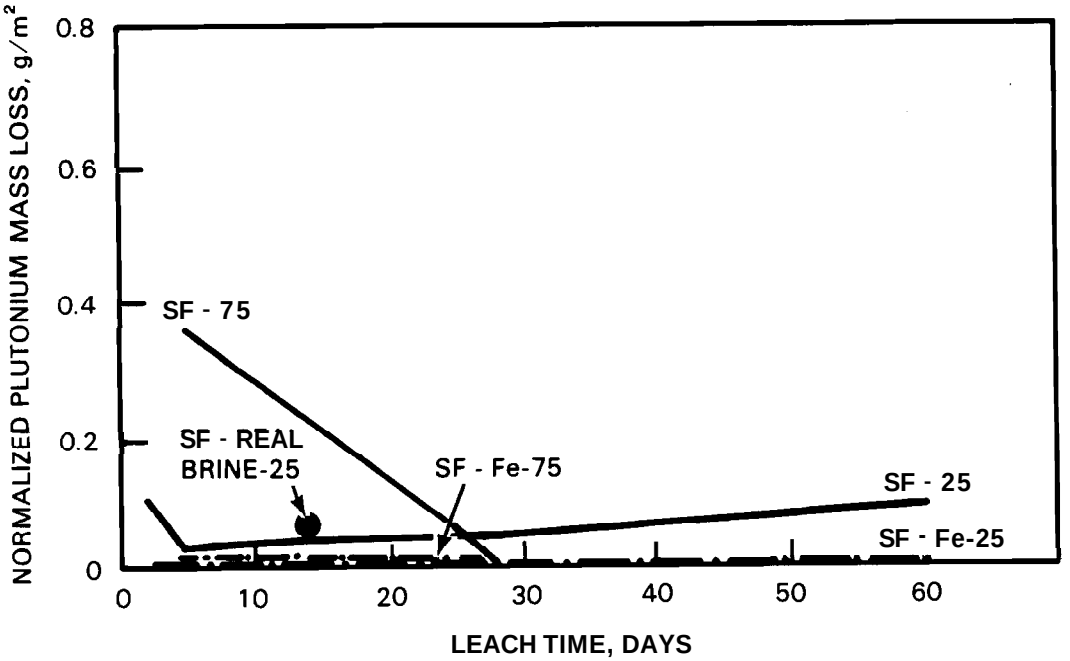

FIGURE 4.33. Plutonium in Filtrate in Spent Fuel Systems at 25 and 75°C in Synthetic and Real Brine (obtained by preparing a saturated solution of salt cored from the Permian Basin)

TABLE 4.9. Technetium Released from Spent Fuel (SF) in Synthetic and Real Brine

Sample Type	No. Days	Technetium Released, g/m^2				
		Leachate	Plateout on Quartz	Plateout on Iron	Total	Filtrate
25°C Tests						
Synthetic Brine						
SF	2	21.3	0.66	-	22.0	20.7
	5	8.5	0.30	-	8.8	8.06
	14	23.8	0.62	-	24.4	22.7
	28	10.2	0.21	-	10.4	9.9
	60	12.9	0.70	-	13.6	11.7
SF-Fe	2	0.38	<0.09	6.23	6.7	0.26
	5	0.91	0.18	14.65	15.7	0.48
	14	2.55	0.37	15.02	17.9	1.17
	28	3.67	0.40	21.24	25.3	2.67
	60	3.17	0.37	7.69	11.2	3.97
Real Brine[a]						
SF	14	14.1	0.73	-	14.8	13.0
75°C Tests						
Synthetic Brine						
SF	5	30.6	0.44	-	31.0	28.9
	28	18.1	0.59	-	18.7	16.8
SF-Fe	5	4.83	2.56	4.05	11.4	1.61
	28	4.52	0.77	5.05	10.3	2.67

(a) Obtained by preparing a saturated solution of salt cored from the Permian Basin.

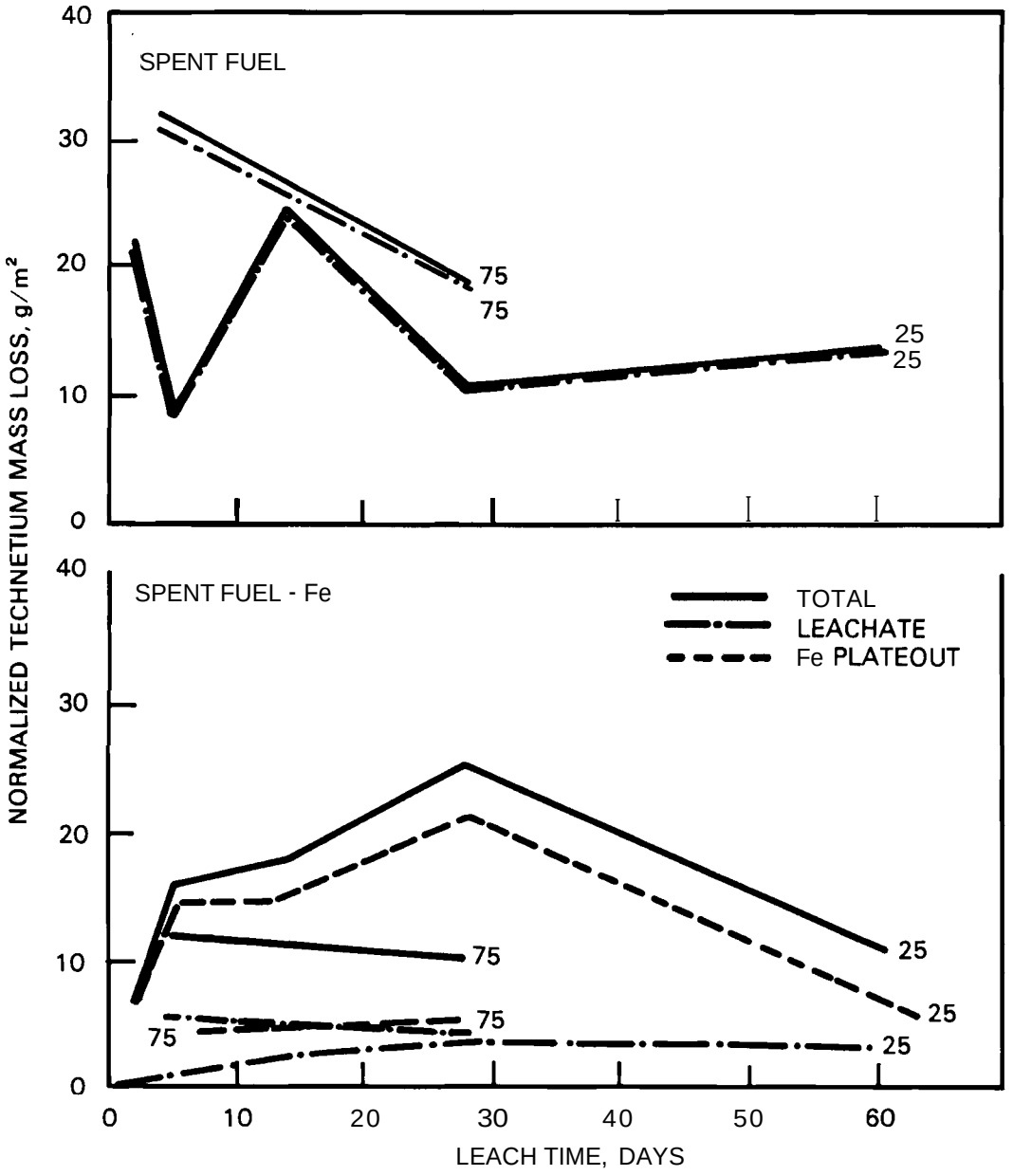

FIGURE 4.34. Comparison of Technetium Released in Systems Incorporating Spent Fuel and Brine at 25 and 75°C

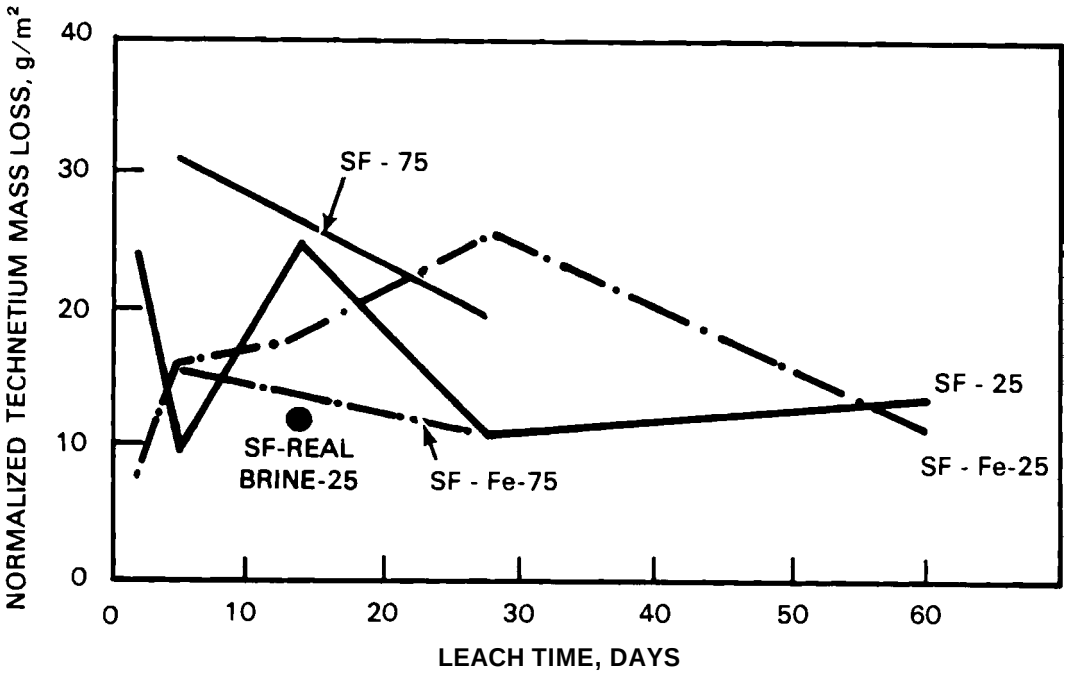

FIGURE 4.35. Total Technetium Released in Spent Fuel Systems at 25 and 75°C in Synthetic and Real Brine (obtained by preparing a saturated solution of salt cored from the Permian Basin)

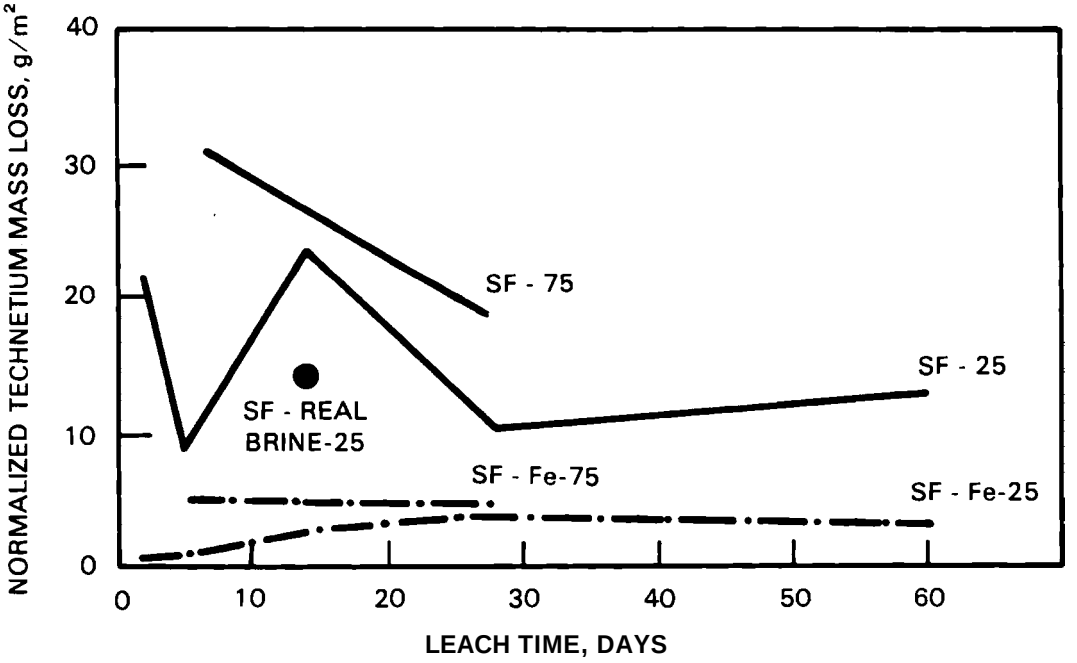

FIGURE 4.36. Technetium in Leachate in Spent Fuel Systems at 25 and 75°C in Synthetic and Real Brine (obtained by preparing a saturated solution of salt cored from the Permian Basin)

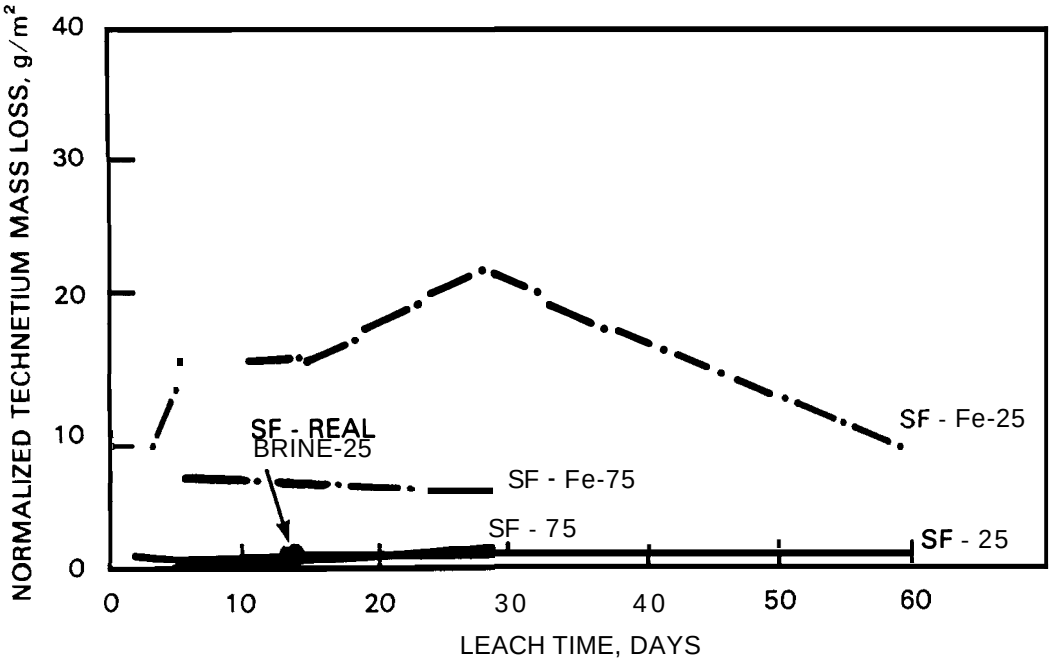

FIGURE 4.37. Technetium Plateout in Spent Fuel Systems at 25 and 75°C in Synthetic and Real Brine (obtained by preparing a saturated solution of salt cored from the Permian Basin)

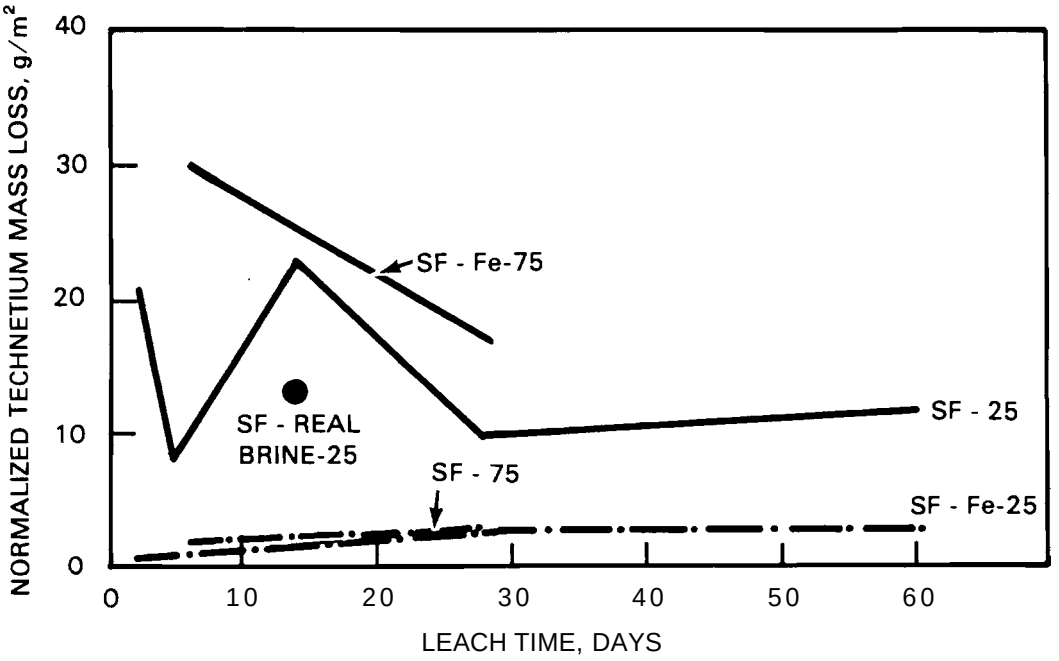

FIGURE 4.38. Technetium in Filtrate in Spent Fuel Systerns at 25 and 75°C in Synthetic and Real Brine (obtained by preparing a saturated solution of salt cored from the Permian Basin)

TABLE 4.10. Cesium Released from Spent Fuel (SF) in Synthetic and Real Rrine

Sample Type	No. Days	Cesium Released, g/m^2				
		Leachate	Plateout on Quartz	Plateout on Iron	Total	Filtrate
25°C Tests						
Synthetic Brine						
SF	2	21.3	0.70	-	22.0	17.6
	5	7.3	0.30	-	7.6	7.0
	14	26.3	0.77	-	27.1	25.4
	28	23.4	0.77	-	24.2	22.5
	60	21.6	1.62	-	23.2	20.4
SF-Fe	2	26.6	1.97	0.27	28.8	25.4
	5	24.3	1.06	0.04	25.4	23.2
	14	27.4	1.55	0.06	29.0	26.1
	28	22.7	0.64	0.04	23.4	21.1
	60	41.2	1.69	0.07	43.0	38.7
Real Brine[(a)]						
SF	14	32.5	1.76	-	34.3	1.5
75°C Tests						
Synthetic Brine						
SF	5	58.4	1.34	-	59.7	54.2
	28	50.1	1.62	-	51.7	46.5
SF-Fe	5	49.9	1.62	0.06	51.6	47.9
	28	58.4	1.48	0.16	60.0	55.6

(a) Obtained by preparing a saturated solution of salt cored from the Permian Basin.

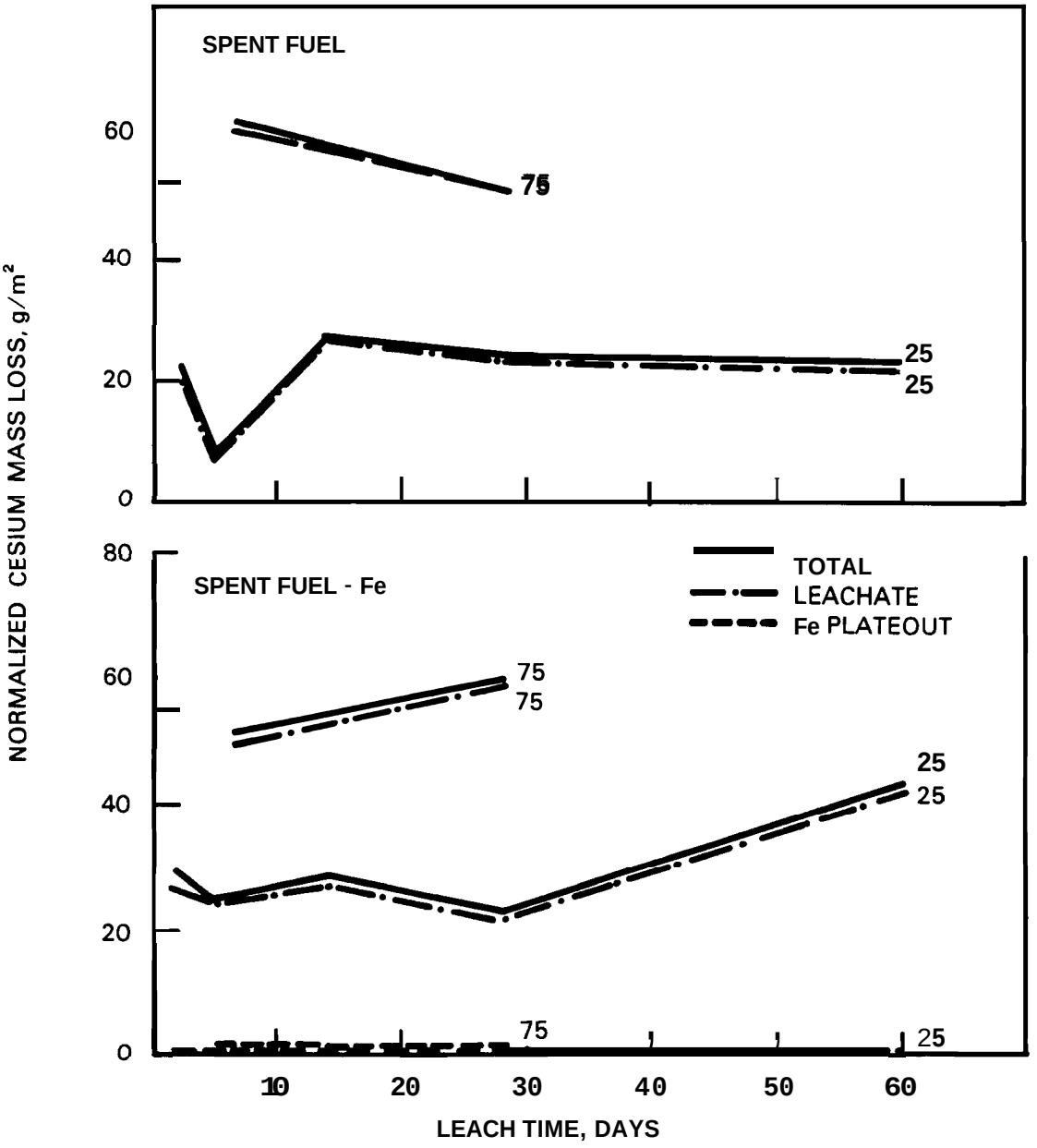

FIGURE 4.39. Comparison of Cesium Released in Systems Incorporating Spent Fuel and Brine at 25 and 75°C

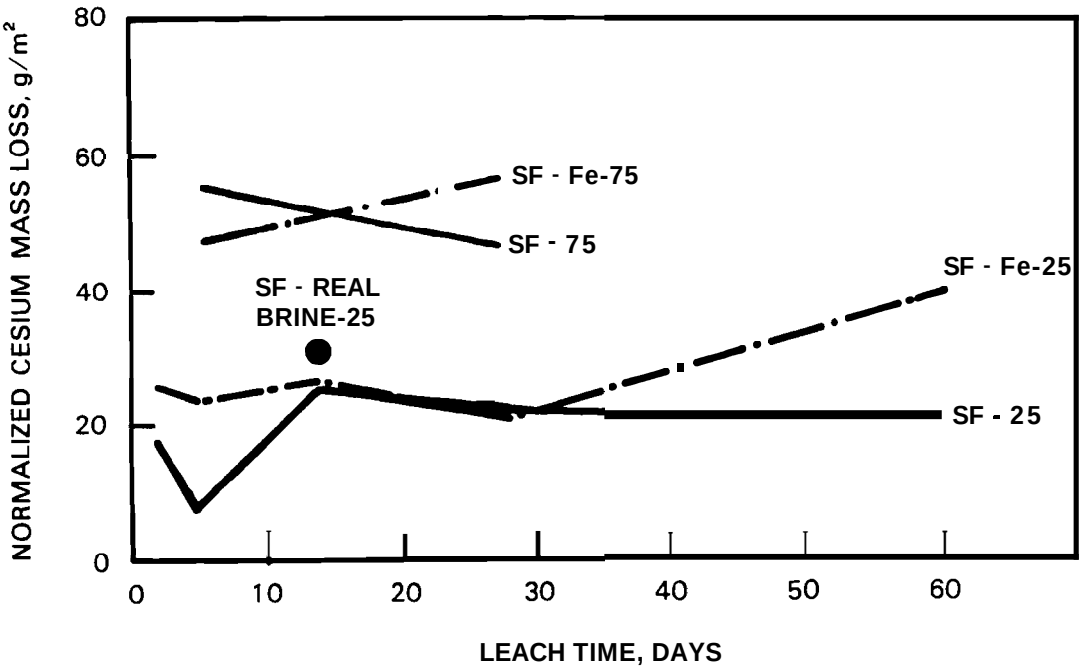

FIGURE 4.40. Total Cesium Released in Spent Fuel Systems at 25 and 75°C in Synthetic and Real Brine (obtained by preparing a saturated solution of salt cored from the Permian Basin)

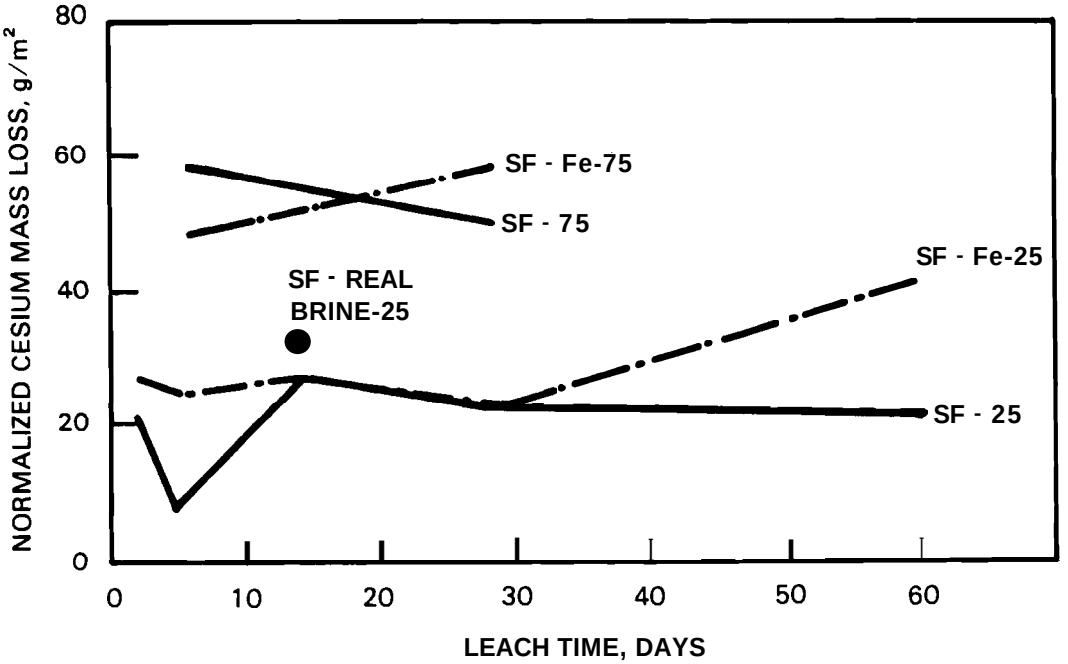

FIGURE 4.41. Cesium in Leachate in Spent Fuel Systems at 25 and 75°C in Synthetic and Real Brine (obtained by preparing a saturated solution of salt cored from the Permian Basin)

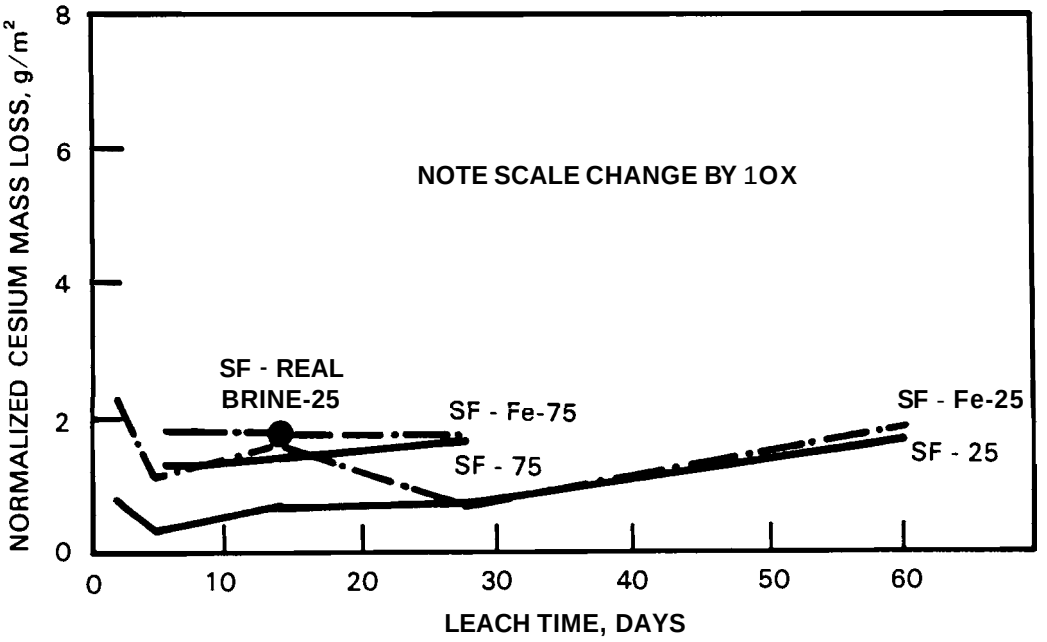

FIGURE 4.42. Total Cesium Plateout in Spent Fuel Systems at 25 and 75°C in Synthetic and Real Brine (obtained by preparing a saturated solution of salt cored from the Permian Basin)

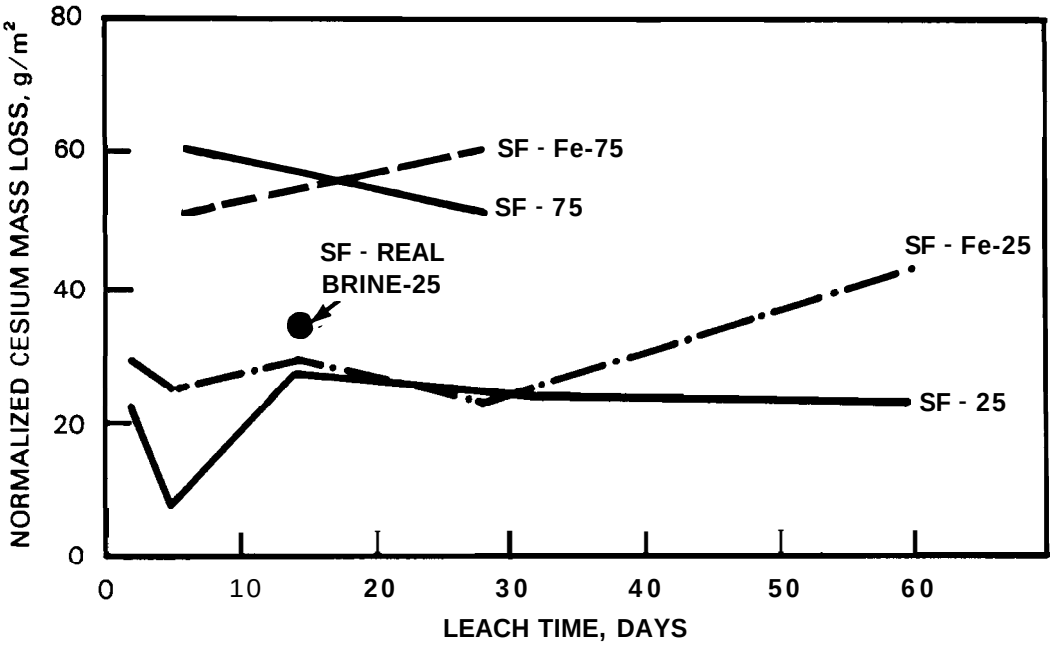

FIGURE 4.43. Cesium in Filtrate in Spent Fuel Systems at 25 and 75°C in Synthetic and Real Brine (obtained by preparing a saturated solution of salt cored from the Permian Basin)

5.0 REFERENCES

DOE/TIC-11400. Nuclear Waste Materials Handbook Waste Form Test Methods, September 1981. Prepared by Materials Characterization Center, Pacific Northwest Laboratory, Richland, Washington.

APPENDIX A

SUMMARY OF RAW DATA

TABLE A.1. Uranium Concentration Data for 25°C Tests of UO_2 in Brine

Sample No.	No. Days	Uranium Concentration, μg/ml					Final Measured pH
			Plateout on				
		Leachate[a]	Teflon[b]	Iron[c]	Zircaloy-4[c]	Filtrate[a]	
UO_2							
U022	2	0.11	<0.001	-	-	0.092	6.66
U025	5	0.092	0.012	-	-	0.088	6.09
U014	14	0.14	0.007	-	-	0.12	6.36
U0228	28	0.17	0.007	-	-	0.17	6.41
U0260	60	0.22	0.006	-	-	0.22	6.41
U0260B	60	0.19	0.006	-	-	0.18	6.75
U0260C	60	0.20	0.009	-	-	0.20	6.41
UO_2-Fe							
U0F22	2	0.045	0.002	0.051	-	0.045	6.04
U0F25	5	0.033	0.009	0.25	-	0.033	6.41
U0F214	14	0.024	0.007	0.054	-	0.013	4.85
U0F228	28	0.021	0.006	0.54	-	0.017	5.87
U0F260	60	0.004	0.014	0.59	-	0.002	6.01
U0F260B	60	0.012	0.005	0.74	-	0.003	6.07
U0F260C	60	0.007	0.010	0.52	-	0.002	5.99
UO_2-Zr							
U0Z22	2	0.13	0.003	-	<0.001	0.10	6.80
U0Z25	5	0.15	0.001	-	<0.001	0.15	6.65
U0Z214	14	0.15	0.001	-	ND[d]	0.15	6.36
U0Z228	28	0.17	0.002	-	ND	0.17	6.44
U0Z260	60	0.18	0.002	-	<0.001	0.18	6.89
U0Z260B	60	0.16	0.003	-	<0.001	0.16	7.04
U0Z260C	60	0.17	<0.003	-	<0.001	0.17	6.46
UO_2-Fe-Zr							
U0ZF22	2	0.047	<0.001	0.12	<0.001	0.044	6.52
U0ZF25	5	0.044	0.002	0.28	<0.001	0.039	6.57
U0ZF214	14	0.016	0.005	0.31	ND	0.014	6.04
U0ZF228	28	0.014	0.005	0.46	ND	0.007	6.34
U0ZF260	60	0.011	0.010	1.1	<0.001	0.006	6.27
U0ZF260B	60	0.010	0.007	0.85	<0.001	0.006	6.27
U0ZF260C	60	0.003	0.004	0.42	<0.001	<0.003	6.28
Releach of U0ZF260							
RU0ZF260	14	0.011	<0.001	0.123	0.015	0.011	6.35

(a) 36 ml sample
(b) 50 ml sample
(c) 10 ml sample
(d) not detected

TABLE A.2. Uranium Concentration Data for 75°C Tests of UO_2 in Brine

Sample No.	No. Days	Uranium Concentration, µg/ml					Final Measured pH
			Plateout on				
		Leachate[a]	Teflon[b]	Iron[c]	Zircaloy-4[c]	Filtrate[a]	
UO_2							
UO72	2	0.066	0.007	-	-	0.063	7.05
UO75	5	0.092	0.059	-	-	0.083	5.59
UO714	14	0.064	0.10	-	-	0.055	5.89
UO728	28	0.058	0.098	-	-	0.056	5.91
UO760	60	0.064	0.081	-	-	0.054	6.78
UO760B	60	0.068	0.059	-	-	0.066	6.62
UO760C	60	0.076	0.038	-	-	0.076	6.46
UO_2-Fe							
UOF72	2	0.041	0.031	0.067	-	0.025	6.68
UOF75	5	0.016	0.13	0.14	-	0.003	5.55
UOF714	14	0.17	0.24	0.34	-	0.041	4.41
UOF728	28	0.038	0.067	0.083	-	<0.003	5.83
UOF760	60	0.033	0.068	0.20	-	<0.003	5.79
UOF760B	60	0.14	0.084	0.40	-	0.010	5.70
UOF760C	60	0.022	0.058	0.12	-	<0.003	5.81
UO_2-Zr							
UOZ72	2	0.13	0.003	-	0.001	0.13	7.11
UOZ75	5	0.10	0.006	-	0.003	0.094	6.40
UOZ714	14	0.083	0.011	-	ND	0.085	6.49
UOZ728	28	0.067	0.014	-	0.013	0.065	6.66
UOZ760	60	0.073	0.015	-	0.006	0.073	6.93
UOZ760B	60	0.068	0.019	-	0.008	0.075	7.22
UOZ760C	60	0.091	0.009	-	0.008	0.082	7.21
UO_2-Fe-Zr							
UOZF72	2	0.031	0.017	0.067	<0.001	0.017	6.58
UOZF75	5	0.052	0.021	0.084	0.002	0.008	5.84
UOZF714	14	0.044	0.049	0.16	ND[d]	ND	5.72
UOZF728	28	0.075	0.055	0.11	ND	ND	6.23
UOZF760	60	0.023	0.020	0.28	<0.001	<0.003	5.97
UOZF760B	60	0.019	0.018	0.22	<0.001	<0.003	6.16
UOZF760C	60	0.036	0.019	0.19	<0.001	<0.003	6.03
Releach of UOZF760							
RUOZF760	14	0.025	0.025	0.018	<0.001	0.003	6.20
UO_2-Oxidized Iron							
OUOF714	14	0.035	0.038	0.007	-	ND	5.90
OUOF714B	14	0.041	0.058	0.008	-	ND	5.95

(a) 36 ml sample
(b) 50 ml sample
(c) 10 ml sample
(d) ND - not detected

TABLE A.3. Uranium Concentration Data for 25 and 75°C Tests of UO_2 in Deionized Water

Sample No.	No. Days	Uranium Concentration, μg/ml					Final Measured pH
		Leachate[a]	Plateout on Teflon[b]	Plateout on Iron[c]	Plateout on Zircaloy-4[c]	Filtrate[a]	
25°C Tests							
DUO214	14	0.59	0.037	-	-	0.34	4.28
DUOF214	14	0.036	0.032	0.32	-	0.002	5.81
DUOZ214	7	0.85	0.026	-	0.021	0.47	5.30
DUOZ274	14	0.83	0.018	-	0.043	0.56	6.18
DUOZF214	7	0.077	0.040	0.004	0.011	0.007	6.93
DUOZF274	14	0.16	0.017	0.045	0.009	0.009	6.28
75°C Tests							
DUO714	14	3.2	0.31	-	-	2.0	3.90
DUOF714	14	0.083	0.30	0.30	-	0.003	6.07
DUOZ714	7	2.5	0.092	-	0.073	1.9	7.41
DUOZ774	14	4.4	0.061	-	0.075	4.9	5.38
DUOZF714	7	0.076	0.24	0.062	0.014	0.013	6.95
DUOZF774	14	(d)	0.86	0.42	0.041	0.001	5.19

(a) 36 ml sample
(b) 50 ml sample
(c) 10 ml sample
(d) Not analyzed. Precipitated solution.

TABLE A.4. Uranium Concentration Data for 150°C Tests of UO_2 in Brine

Sample No.	No. Days	Uranium Concentration, µg/ml					Final Measured pH
			Plateout on				
		Leachate[a]	Teflon[a]	Iron[b]	Zircaloy-4[b]	Filtrate[a]	
UO_2							
B-01	7	0.04	0.15	-	-	0.04	6.67
B-02	15	0.04	0.38	-	-	0.03	6.39
B-03	28	0.03	0.94	-	-	0.02	7.12
B-04	28	0.09	0.03	-	-	0.04	6.18
B-05	60	0.02	0.008	-	-	ND	6.38
B-06	60	1.7	0.01	-	-	1.0	5.09
UO_2-Fe							
B-13	7	0.08	0.003	9.4	-	0.03	6.02
B-14	14	0.04	0.0004	7.6	-	0.02	6.14
B-15	28	0.005	-	5.7	-	<0.001	6.33
B-16	28	0.01	0.57	1.1	-	<0.001	6.40
B-17	62	0.02	0.06	7.2	-	<0.003	6.37
UO_2-Zr							
B-23	7	0.03	0.09	-	2.2	0.03	6.69
B-24	14	0.06	0.02	-	0.25	0.04	6.14
B-25	28	0.008	0.003	-	-	0.007	7.57
B-26	28	0.13	0.004	-	-	0.08	5.75
B-27	60	0.01	-	-	-	0.01	6.65

(a) 72 ml sample. Two pellets were used rather than one as in the 25 and 75°C tests.
(b) µg per metal coupon.

TABLE A.5. Uranium Concentration Data for 150°C Tests of UO_2 in Deionized Water

Sample No.	No. Days	Uranium Concentration, µg/ml					Final Measured pH
			Plateout on				
		Leachate[a]	Teflon[a]	Iron[b]	Zircaloy-4[b]	Filtrate[a]	
UO_2							
Q-07	7	5.1	5.9	-	-	3.0	4.13
Q-08	15	2.0	12.0	-	-	0.9	4.61
Q-09	28	1.2	4.6	-	-	0.8	5.67
Q-10	28	2.8	0.9	-	-	1.4	4.60
Q-11	60	1.9	0.9	-	-	1.0	5.66
Q-12	60	2.9	0.8	-	-	2.0	5.24
UO_2-Fe							
Q-18	7	0.043	0.018	14.8	-	0.003	6.65
Q-14	14	0.35	0.05	30.0	-	0.10	6.05
Q-20	28	0.02	0.08	60.0	-	0.002	7.21
Q-21	28	0.9	0.08	18.0	-	0.001	7.17
Q-22	60	0.10	0.18	17.0	-	0.001	8.07
UO_2-Zr							
Q-28	7	33.8	0.07	-	2.4	31.3	5.19
Q-29	14	20.0	0.03	-	1.1	20.5	4.48
Q-30	28	24.0	0.006	-	-	21.0	5.25
Q-31	28	12.4	0.03	-	-	10.0	4.77
Q-32	60	0.1	0.18	-	-	0.001	4.64

(a) 72 ml sample. Two pellets were used rather than one as in the 25 and 75°C tests.
(b) µg per metal coupon.

TABLE A.6. Uranium Concentration in Filtrate for 150°C Rocking Autoclave Tests of UO_2 in Brine and Oeionized Water

Sample No.	No. Days	Uranium Concentration, µg/ml[a]	Final Measured pH[b]
UO_2-Deionized Water			
1	4	0.13	6.7
2	11	0.45	6.0
3	18	0.23	5.1
4	25	0.16	5.2
5	32	ND[c]	5.1
6	39	0.09	5.1
7	58	0.04	5.05
UO_2-Brine			
1	4	0.01	6.04
2	11	(3.0)[d]	5.6
3	18	0.18	5.9
4	25	0.05	6.1
5	32	0.03	7.1
6	48	0.007	7.2

(a) Uranium concentration in filtrate after passing through an 18Å filter. Initial leachate volume was 230 ml; approximately 5 ml was removed at each time for analysis.
(b) Measured at room temperature.
(c) ND - not detected
(d) Value in parentheses apparently in error and deleted from data plots.

TABLE A.7. Uranium Concentration Data and pH for 25 and 75°C Spent Fuel Tests

Sample No.	No. Days	Uranium Concentration, μg/ml				Final Measured pH	Leachate[c] Volume (ml)
		Filter[a]	Filtrate	Plateout on Quartz	Plateout on Iron[b]		
Spent Fuel, 25°C							
SR-1	2	0.8	13.2	1.9	-	5.94	25
SB-2	5	1.8	14.3	1.1	-	6.48	30
SB-3	14	1.3	15.5	2.5	-	6.38	20
SB-4	28	1.9	25.9	2.2	-	6.33	27
SB-5	60	2.1	23.4	4.1	-	6.17	18
Spent Fuel - Fe, 25°C							
SBF-1	2	9.0	0.8	33.3	137.5	6.0	18
SBF-2	5	3.5	8.9	5.0	179.3	6.4	23
SBF-3	14	9.3	2.5	1.4	322.5	6.38	23
SBF-4	28	7.8	1.8	1.9	582.5	5.96	28
SBF-5	60	10.0	0.9	5.2	993.8	5.67	21
Real Brine[d], 25°C							
RB-3	14	2.6	16.5	5.9	-	6.33	17
Spent Fuel, 75°C							
SB2-2	5	3.7	21.4	11.1	-	4.65	17
SB2-4	28	2.5	0.3	7.1	-	6.24	23
Spent Fuel - Fe, 75°C							
SBF-2	5	9.0	0.7	32.9	50.0	6.3	20
SBF-4	28	11.3	0.97	18.2	65.3	5.09	20

(a) 25 ml sample.
(b) μg per iron coupon.
(c) These volumes apply to the "filtrate" and "quartz plateout" solution.
(d) Obtained by preparing a saturated solution of salt cored from the Permian Basin.

TABLE A.8. Plutonium Concentration Data for 25 and 75°C Spent Fuel Tests

Sample No.	No. Day	Plutonium Concentration, Bq/ml				Leachate[c] Volume (ml)
				Plateout on		
		Filter[a]	Filtrate	Quartz	Iron[b]	
Spent Fuel, 25°C						
SB-1	2	11	27	5	-	25
SB-2	5	15	6.9	16	-	30
SB-3	14	20	10	6.9	-	20
SB-4	28	43	11	58	-	27
SB-5	60	6	22	42	-	18
Spent Fuel - Fe, 25°C						
SBF-1	2	12	0.37	23	73	18
SBF-2	5	25	0.50	65	58	23
SBF-3	14	37	0.47	15	67	23
SBF-4	28	40	0.37	20	160	28
SBF-5	60	130	0.50	110	2200	21
Real Brine[d], 25°C						
RB-3	14	16	16	89	-	17
Spent Fuel - 75°C						
SB2-2	5	12	90	38	-	17
SB2-4	28	20	2.7	100	-	23
Spent Fuel - Fe, 75°C						
SBF2-2	5	53	0.84	850	100	20
SBF2-4	28	62	1.0	58	170	20

(a) 25 ml sample.
(b) Bq per iron coupon.
(c) These volumes apply to the "filtrate" and "quartz plateout" solution.
(d) Obtained by preparing a saturated solution of salt cored from the Permian Basin.

TABLE A.9. Technetium Concentration Data for 25 and 75°C Spent Fuel Tests

Sample No.	No. Days	Plutonium Concentration, Bq/ml				Leachate[c] Volume (ml)
				Plateout on		
		Filter[a]	Filtrate	Quartz	Iron[b]	
Spent Fuel, 25°C						
SB-1	2	2.8	94	3.0	-	25
SB-2	5	2.5	37	1.4	-	30
SB-3	14	4.0	100	2.8	-	20
SB-4	28	1.6	45	0.98	-	27
SB-5	60	4.0	53	3.2	-	18
Spent Fuel - Fe, 25°C						
SBF-1	2	0.42	1.2	0.42	280	18
SBF-2	5	1.8	2.2	0.80	670	23
SBF-3	14	5.7	5.3	1.7	680	23
SBF-4	28	5.0	12	1.8	970	28
SBF-5	60	0.75	14	1.7	350	21
Real Brine[d], 25°C						
RB-3	14	3.5	59	3.3		17
Spent Fuel, 75°C						
SB2-2	5	5.3	130	2.0	-	17
SB2-4	28	5.3	77	2.7	-	23
Spent Fuel - Fe, 75°C						
SBF-2	5	12	7.3	12	370	20
SBF-4	28	6.8	12	3.5	470	20

(a) 25 ml sample.
(b) Bq per iron coupon.
(c) These volumes apply to the "filtrate" and "quartz plateout" solution.
(d) Obtained by preparing a saturated solution of salt cored from the Permian Basin.

TABLE A.10. Cesium Concentration Data for 25 and 75°C Spent Fuel Tests

Sample No.	No. Days	Plutonium Concentration, Bq/ml				Leachate[c] Volume (ml)
				Plateout on		
		Filter[a]	Filtrate	Quartz	Iron[b]	
Spent Fuel, 25°C						
SB-1	2	89	420	17		25
SB-2	5	9.8	170	7.2	-	30
SB-3	14	17	600	18	-	20
SB-4	28	22	530	18	-	27
SB-5	60	22	480	38	-	18
Spent Fuel - Fe, 25°C						
SBF-1	2	22	600	48	120	18
SBF-2	5	28	550	38	20	23
SBF-3	14	28	620	37	31	23
SBF-4	28	42	500	15	23	28
SBF-5	60	48	920	40	34	21
Real Brine[d], 25°C						
RB-3	14	25	730	42	-	17
Spent Fuel, 75°C						
SB2-2	5	68	1300	32	-	17
SB2-4	28	77	1100	38	-	23
Spent Fuel - Fe, 75°C						
SBF-2	5	38	1100	38	29	20
SBF-4	28	53	1300	35	77	20

(a) 25 ml sample.
(b) kBq per iron coupon.
(c) These volumes apply to the "filtrate" and "quartz plateout" solution.
(d) Obtained hy preparing a saturated solution of salt cored from the Permian Rasin.

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