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FY-1984 Annual Report: Spent Fuel and $U0_2$ Source Term Evaluation Results

W. J. Gray
G. L. McVay

February 1986

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

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FY-1984 ANNUAL REPORT:

SPENT FUEL AND UO_2 SOURCE TERM EVALUATION RESULTS

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ABSTRACT

Spent fuel is being considered as a possible waste form for disposal in a salt repository. Static leach tests of spent fuel in salt brine have been conducted to provide a source term that can be used for modeling the potential for release of radionuclides from the repository. The distribution of radionuclides after release from the spent fuel has also been investigated. In addition, leach tests on unirradiated UO_2 pellets have been conducted as an aid toward interpretation of the spent fuel results.

Results from the spent fuel tests showed that almost all of the release of the four elements tested (U, Pu, Tc, and Cs) occurred in the first 5 days and that very little additional release occurred over the next 175 days. Iron coupons in some of the tests had no effect on the total release, but it drastically reduced solution concentrations for all elements tested except Cs. Zircaloy coupons had little or no effect.

The purpose of the UO_2 tests conducted in FY-1984 was to determine whether a U_3O_7 surface layer on UO_2 pellets had any effect on leaching characteristics. It did not.

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INTRODUCTION

Spent fuel is being considered as a possible waste form for disposal in a salt repository. To adequately model the potential release of radionuclides, it is necessary to understand the leach behavior in salt brines and the chemical interactions of the released radionuclides with their environment. Toward this end, leach tests were conducted during fiscal years 1982 and 1983 on unclad spent fuel in salt brine.(1) Some of the tests contained coupons of ductile iron, which was used to simulate a typical container material. In addition, the distribution of radionuclides after release from the spent fuel was investigated; that is, the proportion of radionuclides in true solution, suspended in solution as small particulates, or plated out on the container walls or iron coupons was established.

Besides experiments with actual spent fuel, some experiments were conducted using unirradiated UO_2 pellets. The latter are much less difficult to work with than spent fuel, and a greater variety of analytical tools (primarily surface analytical techniques) can be used with the less radioactive UO_2. Since the matrix material of spent fuel is primarily UO_2, one would expect the uranium release from these two materials to be similar.

A number of noteworthy observations were listed with the FY 1982-83 test results(1) and are reproduced below:

- The leaching characteristics of uranium from both UO_2 and spent fuel in PBB1 brine have a very small temperature dependence over the range studied (25 to 150°C).
- The leaching characteristics of both UO_2 and spent fuel in PBB1 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 50 to 300% higher at 150°C than at 75°C and a factor of 3 to 5 higher at 75°C than at 25°C.
- Considerably more uranium leaches from UO_2 in deionized water than in PBB1 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 a factor of about 5 at 25°C but increases to a factor of about 20 at 75°C. This trend continues at 150°C to the point where the release in deionized water is greater than that in brine by a factor of about 50. When iron is present, there is little difference between deionized water and brine at 25 and 75°C, and the difference at 150°C is only a factor of 3 to 4.
- Uranium leached from spent fuel in PBB1 brine is more than 100 times greater than from UO_2, over the temperature range of 25-75°C.
- 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 it causes a substantial reduction in solution concentration of uranium--probably because it lowers the oxidation potential, thereby lowering uranium solubility or by selective sorption of 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 pH values, with few exceptions, are in the range 5.0 to 7.5, which is a change of no more than about one pH unit from the starting values of both PBB1 brine and deionized water.

The FY-1984 tests were designed to help answer some of the questions raised by the earlier tests as follows:

- The observation that the leaching characteristics of UO_2 and spent fuel was independent of time after the first few days was tested only out to 60 days. Also, there was an exception in that Pu release at 25°C after 60 days was much higher than at shorter time periods. To be sure that the Pu release at 60 days was not the beginning of a trend, as well as to generally extend the data to longer time periods, the FY-1984 test matrix was extended to 180 days. Some replication was also included to improve our knowledge of the reproducibility of these data.
- The time independence phenomenon suggests that the release of the different elements is solubility limited or that there exists a surface phase that is more readily soluble than the bulk material. It was thought that the surfaces of the spent fuel and UO_2 pellets might be more highly oxidized and therefore more soluble. If more of this "oxidized" surface layer was present on the spent fuel than on the UO_2, this could even help to explain why uranium release from spent fuel was so much higher than from UO_2. To test this hypothesis, leach tests in both PBB1 and deionized water were performed on UO_2 pellets, half of which had more highly oxidized surfaces.

EXPERIMENTAL

SPENT FUEL

Spent fuel specimens were leach tested in Permian Basin brine No. 1 (PBB1) at the times and temperatures given in Table 1. Fourteen of the tests included a ductile iron coupon and four tests included both a ductile iron coupon and a short piece of Zircaloy-4 cladding. The 30°C tests were conducted at ambient hot-cell temperature. The 90°C tests were carried out in a specially constructed oven whose temperature was controlled at 90 ±1°C.

TABLE 1. Leach Test Matrix (FY-84)

Time (days)	Number of Tests Run Under Each Condition					
	Spent Fuel		Spent Fuel-Fe		Spent Fuel-Fe-Zr	
	30°C	90°C	30°C	90°C	30°C	90°C
28	1	1	1	1		
60	1	1	1	1		
90	1	1	2	2	1	1
120	1	1	1	1		
180	2	2	2	2	1	1

The spent fuel used in the study was from fuel bundle BO-5 which was discharged from the H. B. Robinson II reactor on June 6, 1974, at an average burnup of 28 MWd/kgM. Fuel was removed from the fuel rod at the Battelle-Columbus Hot Laboratory. Unclad fuel fragments from more than one rod were combined and shipped to PNL for testing. About 5g of spent fuel fragments were dissolved and chemically analysed for uranium, plutonium, and two important fission products; the results are listed in Table 2.

TABLE 2. Selected Chemical Analysis for the Spent Fuel

Species	Value
U	8.4×10^5 µg/g
^{239}Pu and ^{240}Pu	2.47×10^7 Bq/g
^{99}Tc	4.55×10^5 Bq/g
^{137}Cs	2.37×10^9 Bq/g

Each leach specimen consisted of three fuel fragments whose ^{137}Cs activity had been individually measured (^{137}Cs activity is considered to be proportional to burnup). The individual fragments were combined into groups of three in such a manner that the total activity of all the three-fragment specimens was the same within ±5%. This assured that each specimen represented an overall average of the fuel being tested.

Photographs were taken of each specimen together with a millimeter scale and used to estimate surface areas. Total surface areas of the three-fragment specimens ranged from 2.6 to 4.1cm^2. The ratio of spent fuel surface area to leachant volume (SA/V) was 10 m^{-1} for all tests.

The iron and zircaloy coupons were prepared to have surface areas equal to the average spent fuel specimen, about 3.3 cm^2. The iron coupons were freshly machined parellelepipeds whereas the zircaloy coupons were short segments of PWR tubing. Table 3 lists the composition of the iron; Table 4 lists the nominal composition of Zircaloy-4 tubing. The iron was cleaned with 6 M HCl and the zircaloy was cleaned with 6 M HNO_3/1 M HF prior to the leach tests.

TABLE 3. Ductile Iron Composition for UO_2 and Spent Fuel Leach Tests

Element	Wt%
C	3.53
Mn	0.31
Si	2.51
P	0.05
S	0.004
Fe	Balance

The leach tests were conducted in fused silica containers that were cleaned in accordance with MCC-1 requirements.(2) Fused silica or Pyrex lids were sealed to the ground surfaces of the leach containers using silicone rubber gaskets. The spent fuel, iron, and zircaloy specimens

TABLE 4. Nominal Composition of Zircaloy-4 Tubing Used in the Spent Fuel Leach Tests

Element	Wt%
Sn	1.20 to 1.70
Fe	0.18 to 0.24
Cr	0.07 to 0.13
Fe + Cr	0.28 to 0.37
Zr	Balance

were placed in separate compartments of perforated fused silica baskets to minimize contact between the specimens and the container, and to maximize the surfaces of the specimens exposed to the leachant. Figure 1 is a schematic diagram of the leach container used for the ambient tests. The 90°C tests used the same type of container, but the fused silica lid was held in place by the lid of the specially constructed oven.

The Permian Basin brine No. 1 (PBB1) used in these studies represents an intrusion brine because it simulates the saturated solution obtained by dissolving Permian Basin Cycle 4 salt (a blended composite of 10 core samples from G. Friemel Core Hole No. 1, approximately 10-foot intervals in the 2440.2 to 2575.5 ft. section) in deionized water. It was prepared by dissolving the following salts in deionized water to produce 1.0 liter of solution.

Salt	Weight (g)
NaCl	309.7
$CaCl_2 \cdot 2H_2O$	5.73
Na_2SO_4	4.73
$MgCl_2 \cdot 6H_2O$	1.12
KCl	0.075
$SrCl_2$	0.064
$NaHCO_3$	0.032
NaBr	0.031
$ZnCl_2$	0.016
NaF	0.0021

Following the leach period, the containers were cooled to ambient hot-cell temperature (see the following discussion regarding this practice) and weighed to determine if any leachate had been lost. The leachate pH was measured, and two aliquots of leachate were taken for chemical analyses; one aliquot was passed through a 1.8 nm filter. Both aliquots were then acidified with 0.1 ml of concentrated nitric acid per 10 ml of solution to assure that the U and Pu remained in solution. After removing the spent fuel specimen and metal coupons, any remaining fragments of spent fuel were rinsed from the container and basket using fresh leachant. Then, a 5 M HNO_3, 0.05 M HF solution was placed in the leach container with its associated specimen basket in volume slightly greater than the original leachate volume and allowed to stand at least 2 hours at ambient temperature to dissolve any material plated out on the container walls or on the basket. Material plated out on the iron and zircaloy coupons was removed by placing each coupon in a series of three fresh solutions of 6 M HCl for ten minutes each. Then the solutions were combined and diluted to a known volume.

Each solution was analyzed for uranium, plutonium, cesium, and technetium. Cesium was determined by direct gamma counting each solution. 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. Plutonium was separated from other radioactive species by extraction into thenoyltrifluoroacetone in xylene, evaporating onto a plate and alpha-counting the residue. 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.

It was stated earlier that the leach containers in the 90°C tests were cooled to ambient temperature prior to preparation of aliquots for analyses. This practice was considered to be acceptable because the brine is saturated at 25°C, not at 90°C. Thus, no precipitation of the brine components, which could drag down some of the elements of interest, is expected. Furthermore, no procedure has yet been developed for elevated temperature filtration. Table 5 shows that the concentrations of U, Tc, and Cs are higher in the 90°C tests indicating that these elements, at least, are not reduced to the 30°C concentrations by this practice. Plutonium concentrations at the two temperatures were about the same. Whether or not this is an artifact of the cooldown prior to filtration is not known, but it is to be noted that other aspects of the Pu leach behavior (e.g., total release) are only mildly temperature dependent over this range. The bulk of the evidence, therefore, points toward the cooldown prior to filtration being an acceptable practice for these tests.

UO_2

Surface-oxidized and as-prepared UO_2 pellets were leached in this series of experiments to test the hypothesis that a more highly oxidized, and therefore more soluble, surface layer on UO_2 pellets dissolves quickly after which the leach rate decreases. The pellets were prepared by cold-pressing UO_2 powder depleted in ^{235}U, and then sintering at 1700°C in a 50% H_2-50% Ar atmosphere for 4h to 96% theoretical density. Then they were centerless ground, cleaned with ethanol and dried, and fired again at 1045°C in a 50% H_2-50% Ar atmosphere for 4h to assure that any surface material that might have been oxidized during centerless grinding was again reduced to UO_2. Half the pellets were then heated at 215°C in air for 10 days to generate a thin U_3O_7 film.(3) Weight gains for the 16 pellets ranged from 1.3 to 4.4 mg and averaged 2.0 mg. This weight gain is equivalent to 5.5×10^{-4} g/cm^2, and since the density of U_3O_7 is 11.05 g/cm^3 compared to 10.75 g/cm^3 for UO_2, this weight gain is equivalent to a U_3O_7 surface layer 18 μm thick. The pellets were 9.1 mm diameter by 8.2 mm long and, although the lengths varied slightly, they all had surface areas within 2% of 3.62 cm^2.

FY 84 Test Matrix for UO_2

The UO_2 pellets were leach tested according to the following matrix:

1 Temperature	-	90°C
2 Sample types	-	Oxide film; no oxide film
2 Leachants	-	PBB1; deionized water
4 Time periods	-	5,14, 28, 28 days (i.e., duplicate samples were run at 28 days.)
2 Environments	-	With iron; without iron

Total number of samples = 1 x 2 x 2 x 4 x 2 = 32

Ductile iron coupons with surface areas of 3.6 cm^2 (same as the UO_2 pellets) and the composition listed in Table 3 were used in half the UO_2 leach tests. The ratio of UO_2 surface area to leachant volume (SA/V) was 10 m^{-1} for all tests.

The leach tests conducted on UO_2 used Teflon leach containers and specimen baskets, which were cleaned in accordance with MCC-1 requirements.(2) The baskets prevented contact between the UO_2 pellets and iron coupons and minimized contact of both with the container surfaces.

Following the leach period, the containers were cooled to ambient laboratory temperature (see the discussion of this practice in the spent fuel section) and weighed. In all cases, the loss of water was found to be less than 2 percent. The leachate pH was measured and the different analytical solutions were prepared in the same manner used in the spent fuel leach tests as described earlier. An exception was the dissolution of material off the iron coupons; only one 6 M HCl solution was used for 15 minutes because it was felt that this was adequate to remove at least 90% of any deposited uranium. Uranium concentrations in each solution were then analyzed using the same method as in the spent fuel tests.

RESULTS AND DISCUSSION

GENERAL

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:

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

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 = 0.84 for spent fuel and 0.88 for UO_2;
SA = surface area of the leach specimen, m^2.

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

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

where $(NL)_i$ = normalized elemental mass loss, g/m^2;
a_i = activity of the element in the solution, Bq;
a_0 = activity of the element in the spent fuel fragment, Bq,
W_0 = mass of the specimen, g, and
SA = surface area of the leach specimen, m^2.

The results are presented in terms of location of the released radioactive species. The sum of species in the leachate, plateout on the container, and plateouts on the iron and zircaloy coupons, 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 and can be considered to be in solution. The difference between the leachate value and the filtrate value represents that which was suspended, possibly in colloidal form, in the leachate.

The data are presented in both graphic and tabular form in Figures 2-5 and Tables 7-10. In addition, raw data are given in Appendix A. Data points are not shown on the graphs because they would become too busy. Rather, straight lines were drawn between data points; averages were used where data points were replicated. Error bars shown on the graphs represent the spread in the total release data for the replicated points (i.e., the bars extend from the maximum to minimum measurement rather than a standard deviation).

SPENT FUEL

The FY-1984 spent fuel program extended the leaching data from 60 to 180 days and provided some needed replication. A number of observations regarding the data, which are presented in Figures 2-5 and Tables 7-10, are given below. It is also important to note at the beginning that: (a) these observations also apply almost without exception to earlier spent fuel leaching data;(1) and (b) the values obtained for the normalized mass losses in the two sets of experiments were in good agreement. The good agreement was particularly reassuring since the two sets of experiments were done in different laboratories by different people using different equipment. However, the source of spent fuel specimens was the same; the same leach procedures were used; and the chemical analyses were done in the same laboratory for the two sets of experiments. Note that ambient hot cell temperature was 25 and 30°C for the earlier and later tests, respectively. The difference in terms of its effect on leach results is insignificant.

- All four elements (U, Pu, Tc, and Cs) exhibit very little time dependence in the total release data. That is, nearly as much was released in 28 to 60 days as in 180 days. This trend was also observed in the earlier tests where leaching time periods of 5 days produced nearly as much total release as times up to 60 days. A possible exception with the present data is uranium at 90 days without iron. However, despite the high release by replicate samples at 180 days, the apparent increase in total release at long times must be viewed with suspicion since it is the exception to a general trend.
- Of the four elements tested, cesium release exhibits the largest temperature dependence with the 90°C release being about 3 times the 30°C release. Pu release is essentially independent of temperature in the range 30 to 90°C. The temperature dependence of U and Tc release falls between that of Cs and Pu. Only Cs was shown to have a small temperature dependence between 75 and 25°C in the earlier data.
- The presence of iron has no effect on total release (Pu at 90°C appears to be an exception but, again, this must be viewed with suspicion since it is contrary to the trend), but it does reduce solution concentrations (filtrate) nearly to zero for all elements tested except Cs.
- Zircaloy has no noticeable effect on leaching although some Tc collected on it in the 90-day tests (none at 180 days).

The following statements can be made regarding the dispositions of the different elements in the various tests:

Uranium (See Figure 2)

- Most of the released U was found in solution when iron was absent, but almost none when iron was present.
- When iron was present, about half the released U was deposited on the iron; the other half was in filterable form.
- Very little released U was plated out on the container walls at 30°C, but there was some at 90°C.

Plutonium (See Figure 3)

- About half the released Pu was found in solution when iron was absent, but almost none when iron was present.
- Almost none of the released Pu deposited on the iron.
- Roughly half the released Pu plated out on the container walls in all cases; the other half was in solution when iron was absent and it was filterable when iron was present.

Technetium (See Figure 4)

- Essentially all the released Tc was found in solution when iron was absent, but only very little when iron was present.
- A large portion (more than half) of the released Tc deposited on the iron.
- Almost none of the released Tc plated out on the container walls in any case.
- Some of the released Tc was plated out on the zircaloy after 90 days but none after 180 days. Because only a few tests contained zircaloy, no generalizations can be made.

Cesium (See Figure 5)

- Essentially all the released Cs was found in solution with or without iron present.
- Essentially none of the released Cs plated out on either the walls or the iron in any case.

Tables 7-10 and Figures 2-5 show that the amounts of elements in the filtrates (i.e., solution concentrations) do not vary much with time over periods from 28 to 180 days. Therefore, solution concentrations were averaged over time for each element and each condition and are listed, along with detection limits, in Table 5. These concentrations were assumed to represent elements truly in solution since they have passed through a 1.8 nm filter. In the presence of iron, only small fractions of the total release of U, Pu, and Tc were found in solution. Even without iron, only about half the released Pu was found in solution. Solution concentrations in these cases can, therefore, be taken to represent solubility limits since solution and solid phases coexisted over considerable periods of time.

TABLE 5. Solution Concentrations in Spent Fuel Tests in Brine (mol/liter)*

	Without Iron		With Iron		Detection Limit
	$30^{\circ}C$	$90^{\circ}C$	$30^{\circ}C$	$90^{\circ}C$	
Uranium	8×10^{-5}	3×10^{-4}	2×10^{-6}	8×10^{-6}	8×10^{-9}
Plutonium	2×10^{-8}	2×10^{-8}	2×10^{-10}	1×10^{-10}	2×10^{-11}
Technetium	1×10^{-6}	2×10^{-6}	5×10^{-8}	1×10^{-7}	5×10^{-9}
Cesium	3×10^{-6}	1×10^{-5}	3×10^{-6}	1×10^{-5}	1×10^{-8}

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*These approximate concentrations were measured after the solutions were cooled to ambient temperature (30°C) and passed through a 1.8 nm filter.

It has been noted that there is a general lack of time dependence in the total release data for either the present or earlier test matrix. Furthermore, as has also been noted, the presence of iron has little effect on the total release of U, Pu, and Tc despite its marked effect on the solution concentrations of these elements. It appears that the release simply stops after a short time, independent of an apparent difference in redox conditions caused by the presence or absence of an iron coupon. This phenomenon suggests that one of three conditions exists: (1) a relatively reactive phase has been depleted leaving a less reactive phase and an apparent halt in the release; (2) some type of protective layer has been formed; or (3) some reactant in the brine has become depleted.

pH values were measured at the end of the leach tests using a combination glass electrode, but the electrode was subsequently found to be faulty. Evidence for this derived from measured values that were consistently lower than measured in previous tests(1) despite the fact that the instrument was frequently calibrated using buffers at pH 4 and 7. In addition, the measured pH values of two blank samples (PBB1 brine allowed to stand in the hot cell at 30°C in leach containers for 43 days with no spent fuel or metal coupons present) were similarly low. For reasons that are not well understood, a pH electrode can sometimes fail to read high ionic strength brine correctly despite the fact that it properly reads the low ionic strength buffers. This problem is usually not observed except with electrodes that have been used in brines for periods of several weeks to several months. Unfortunately, in the present case the faulty electrode was discarded and a new one installed before a direct comparison between the two, using the same test solution, could be made. However, since the pH of the blank samples should have been nearly the same as fresh PBB1 brine, a means of correcting the erroneous pH measurements is provided. The new electrode registered a pH of 5.65 for fresh PBB1 brine, whereas the faulty electrode registered 4.69 and 4.65 for the two blank samples. Thus, the correction is 1.0 pH units. In addition, there is a correction of 0.9 pH units for the liquid junction potential of the combination electrode.(4) Therefore, a total correction of 1.9 was applied to the data in Tables 7-10. Relative uncertainties of ±0.1 pH unit are probably applicable when comparing values for the different samples. Because of the above correction, however, absolute uncertainties are somewhat larger, probably ±0.5 pH units.

The pH data listed in Tables 7-10 show that the pH of the 180-day samples is about 0.6 units less than at all shorter time periods. This decrease appears to be real since measurements on the 180-day samples were made 2 to 3 weeks prior to those on the 120-day and 60-day samples. pH measurements on the 28-day and 90-day samples were made 5 months and 3 months prior to those on the 180-day samples, respectively. Thus, a gradual deterioration of the electrode does not seem to be the reason for the lower pH values of the 180-day samples.

Table 11 lists pH values for leachates from UO_2 leach tests that are somewhat lower than those listed in Tables 7-10 for the spent fuel leach tests. Because of the uncertainties in the absolute values of the latter, this difference is not considered to be significant.

$\underline{UO_2}$

Data for the UO_2 tests are presented in Figures 5 and 6 as well as in Tables 11 and 12. The purpose of these tests was to determine whether a U_3O_7 surface layer had any effect on leaching characteristics of UO_2. Apparently, it did not since the only case where oxidized pellets did release more uranium was at 28 days, without iron, in deionized water. Since this was the only exception, it can probably be discounted. Thus, if a more readily soluble surface layer, different from the bulk, is part of the reason for the difference between UO_2 and spent fuel, it must be something other than U_3O_7.

When comparing the earlier data(1) with the present results, it must be remembered that the former data were from 25 and 75°C tests, whereas the latter were from 90°C tests. Although only a few of the earlier tests were conducted with deionized water, some temperature dependence of uranium release in deionized water was observed--at least in the absence of iron. In brine, there was little or no temperature dependence. The present data, in the absence of iron, are quite consistent with the earlier results. However, data from the present tests conducted in the presence of iron are not consistent with the earlier results. In deionized water, iron had little effect on the total uranium release in the present results. In contrast, the earlier results show a marked decrease in total uranium release when iron was present at both 25 and 75°C, although the effect was greater at 75°C. The reverse happened in brine. That is, the present data show an increase in total uranium release in the presence of iron, whereas the earlier data show no effect of iron on total uranium release at either 25 or 75°C.

Rationalization of these seemingly inconsistent results will require further studies to discover the mechanisms involved in UO_2 leaching. The most important point to be made here, however, is that the U_3O_7 surface layer had no effect, which was the question these present tests were designed to answer.

Table 6 lists solution concentrations, averaged over time, of uranium in the filtrates. These data can be compared with those for the spent fuel in Table 5. Solution concentrations in the spent fuel tests averaged 400 and 3000 times greater than in the UO_2 tests for the tests with and without iron, respectively. Perhaps the alpha and/or gamma radiation from the spent fuel makes the solutions more oxidizing and makes the uranium, therefore, more soluble. Tests designed to answer this question are planned.

TABLE 6. Uranium Concentrations (mol/liter) in 90°C UO_2 Tests

	Without Iron		With Iron	
	Unoxidized	Oxidized	Unoxidized	Oxidized
Deionized Water	8×10^{-7}	8×10^{-6}	4×10^{-8}	4×10^{-8}
PBB1 Brine	1×10^{-7}	3×10^{-7}	2×10^{-8}	2×10^{-8}

DISCUSSION OF ERRORS

Errors in leaching results can arise through improper preparation and/or handling of the different analytical solutions that are represented in the figures and tables that follow. Errors can also occur in the analytical measurements themselves. Estimates of both types of errors could best be made through some type of systematic study using standard solutions, but this has not been done.

There can also be real differences between samples such as different surface conditions or differences in crack structure, particularly with regard to fission product distributions. Such differences would be expected to be more pronounced with spent fuel than with UO_2, and there does seem to be more scatter in the spent fuel data.

Tables 11 and 12 are tabulations of the data shown graphically by Figures 5 and 6. These tables also show that the amount of time that the leachate solutions are allowed to stand before analysis can affect the results. Only solutions from samples leached for 28 days were analyzed at different times. Because of the sample-to-sample scatter in the data, it is difficult to compare the "A" samples, which were analyzed immediately, with the "B" samples, which were allowed to stand for one week before analyzing. Not enough re-analyses were made on the Teflon and iron plate-out solutions to establish trends. Trends for the leachate and filtrate solutions appear to be in opposite directions. A possible explanation for the increase with time of uranium in the leachate solutions is that some of the uranium initally remains in particulate form and is, therefore, not "seen" by the analyzer. This, despite the fact that the solution is acidified (0.1 ml concentrated HNO_3 per 10 ml of solution) for the purpose of dissolving and keeping all of the uranium in true solution. With time, more of the uranium may dissolve and be "seen" by the analyzer. If this explanation is true, one would not expect to observe an increase in uranium with time if all the uranium were truly in solution to begin with (i.e., before acidification). Such a situation would be indicated by having leachate and filtrate concentrations the same. That situation existed only for the unoxidized UO_2 without iron in brine (samples BU28A and BU28B). Although the data for sample BU28A, in particular, do not support the explanation offered, the explanation otherwise seems reasonable and should not be discounted unless and until further contradictory evidence is obtained. In the meantime, a change in the way the analytical solutions are prepared and/or handled may be warranted.

A logical explanation for the apparent decrease with time of uranium in the filtrate solutions is more open to question. Evidently, some of the uranium precipitates or plates out on the sample container walls with time, but that is an unsatisfactory explanation because the solutions were acidified for the express purpose of preventing this from happening. Additional studies would be required to demonstrate whether that is the right explanation and, if so, how to prevent it.

The spent fuel data, in particular, exhibit considerable scatter. It was pointed out earlier that more scatter might be expected in the spent fuel data than in the UO_2 data. Since the leach results should be represented by some type of smooth curve, some feel for the uncertainty can be obtained just by looking at this scatter. In an effort to obtain more information on uncertainties, some of the tests were replicated in both the spent fuel and UO_2 tests. Complete tabulations of these data are given in the tables. Replications are shown only for the total release data in the figures where the spread between the maximum and minimum results are shown as verticle bars. On average, the replicate tests show a scatter that is similar to all the other data in general. Based on the observed scatter, it is estimated that the average leach data are accurate only to within a factor of about 2 for both the spent fuel and UO_2.

CONCLUSIONS

Conclusions based on the spent fuel studies are as follows:

- Release of the four elements analyzed (U, Pu, Tc, and Cs) was almost independent of time over the period 5 to 180 days. That is, almost all of the release occurred in the first 5 days.
- The presence of iron had no effect on the total release, but it did reduce solution concentrations nearly to zero for all elements tested except Cs.
- Zircaloy had no effect on the leaching although some Tc collected on it during some of the tests.

Conclusions based on the UO_2 studies are as follows:

- The purpose of the UO_2 tests was to determine whether a U_3O_7 surface layer had any effect on the leaching characteristics of UO_2. With one exception, which can probably be discounted, it did not.

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2. DOE/TIC-11400, Nuclear Waste Materials Handbook, Test Methods. MCC-1. Static Leach Test Method, p. 1-4.
3. White, G. D., C. A. Knox, E. R. Gilbert and A. B. Johnson, Jr. 1983. "Oxidation of UO_2 at 150 to 350°C." In Workshop on Spent Fuel/Cladding Reaction During Dry Storage, NUREG/CP-0049, Appendix F, ed. D. W. Reisenweaver.
4. Pederson, L. R. 1985. Radiation and Heat Damage Effects in Natural Rock Salts. PNL-5212, Pacific Northwest Laboratory, Richland, WA 99352.

TABLES/FIGURES

TABLE 7. Data for 30°C Tests of Unclad Spent Fuel in PBB1 Brine

Element	Sample	Normalized Mass Loss (g/m^2) at Different Times (Days)						
		28	60	90	120	180		
						#1	#2	Ave.
Uranium	Leachate	2.01	0.666	1.69	1.56	1.14	2.34	1.74
	Filtrate	1.91	0.666	1.69	1.45	0.457	2.23	1.34
	Plate-Out	0.046	0.381	0.14	0.102	0.226	0.080	0.153
	Total	2.06	1.05	1.83	1.66	1.37	2.42	1.90
Plutonium	Leachate	0.269	0.023	0.0775	0.0277	0.0716	0.237	0.154
	Filtrate	0.180	0.016	0.0561	0.0126	0.0653	0.225	0.145
	Plate-Out	0.436	0.112	0.0724	0.0828	0.154	0.121	0.138
	Total	0.705	0.135	0.150	0.111	0.226	0.358	0.292
Technetium	Leachate	15.8	19.0	13.2	16.4	12.2	18.0	15.1
	Filtrate	15.2	17.5	13.2	16.7	12.2	18.2	15.2
	Plate-Out	<0.43	0.127	1.11	<0.081	<0.08	<0.08	<0.08
	Total	16.2	19.1	14.3	16.4	12.2	18.0	15.1
Cesium	Leachate	26.2	24.3	30.8	33.2	19.4	33.9	26.7
	Filtrate	26.0	24.0	31.2	32.0	18.8	32.9	25.9
	Plate-Out	0.202	0.077	0.119	0.021	0.045	0.04	0.023
	Total	26.4	24.4	30.9	33.2	19.4	33.9	26.7
Measured pH	Leachate	5.2	5.0	4.9	5.1	4.4	4.3	
Corrected pH*	Leachate	7.1	6.9	6.8	7.0	6.3	6.2	

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*A correction of 0.9 for the liquid junction potential plus 1.0 for the electrode error described in the text was applied to these data.

TABLE 8. Data for 30°C Tests of Unclad Spent Fuel/Iron in PBB1 Brine

Element	Sample	Normalized Mass Loss (g/m^2) at Different Times (Days)										
		28	60	90				120	180			
				#1	#2	#3	Ave.		#1	#2	#3	Ave.
Uranium	Leachate	1.26	2.18	1.74	1.35	1.45	1.51	3.71	4.23	1.80	3.41	3.15
	Filtrate	0.126	0.041	0.031	0.020	0.031	0.027	0.0456	0.68	0.017	0.026	0.24
	Plate-Out	0.159	0.14	0.70	0.19	0.165	0.35	0.258	0.88	0.23	0.29	0.47
	Iron	2.07	1.21	1.99	3.06	2.50	2.52	1.72	2.27	2.79	2.03	2.36
	Zirconium			0.011			0.011		0.002			0.002
	Total	3.49	3.53	4.44	4.60	4.12	4.39	5.69	7.38	4.82	5.73	5.98
Plutonium	Leachate	0.131	0.344	0.601	0.242	0.197	0.347	0.354	0.564	0.237	0.462	0.421
	Filtrate	0.00028	0.00056	0.00023	0.00075	0.00034	0.00044	0.0015	0.501	0.001	0.003	0.17
	Plate-Out	0.0932	0.072	0.0001	0.055	0.060	0.038	0.157	0.001	0.070	0.092	0.054
	Iron	0.0049	0.012	0.006	0.0058	0.0043	0.0054	0.0233	0.037	0.126	0.0226	0.062
	Zirconium			0.001			0.001		0.001			0.001
	Total	0.229	0.428	0.618	0.303	0.261	0.394	0.534	0.603	0.320	0.577	0.500
Technetium	Leachate	4.40	1.97	2.71	1.56	1.87	2.05	2.23	1.74	3.34	2.42	2.50
	Filtrate	2.03	0.668	0.44	0.39	0.719	0.52	0.821	0.854	1.64	3.30	1.93
	Plate-Out	0.640	0.221	0.863	<0.27	0.62	0.58	0.125	0.622	0.259	0.081	0.321
	Iron	2.41	8.81	11.0	12.8	12.8	12.2	10.6	11.65	8.87	11.3	10.6
	Zirconium			2.41			2.41		0.168			0.168
	Total	7.45	11.0	17.0	14.6	15.3	15.6	13.0	14.2	12.5	13.8	13.5
Cesium	Leachate	17.7	25.3	31.2	16.8	14.8	20.9	30.4	45.0	29.2	32.0	35.4
	Filtrate	17.6	25.8	27.4	16.8	15.0	19.7	29.5	43.7	29.0	30.7	34.5
	Plate-Out	0.033	0.103	0.064	0.037	0.041	0.047	0.079	0.036	0.021	0.02	0.026
	Iron	0.0051	0.0083	0.003	0.0014	0.0087	0.0044	0.006	0.128	0.026	0.028	0.061
	Zirconium			0.0008			0.0008		0.004			0.004
	Total	17.7	25.4	31.3	16.8	14.9	21.0	30.4	45.1	29.2	32.0	35.4
Measured pH	Leachate	4.9	4.8	4.9	5.0	4.9		5.0	4.3	4.4	4.2	
Corrected pH*	Leachate	6.8	6.7	6.8	6.9	6.8		6.9	6.2	6.3	6.1	

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*A correction of 0.9 for the liquid junction potential plus 1.0 for the electrode error described in the text was applied to these data.

TABLE 9. Data for 90°C Tests of Unclad Spent Fuel in PBB1 Brine

Element	Sample	Normalized Mass Loss (g/m^2) at Different Times (Days)						
		28	60	90	120	180		
						#1	#2	Ave.
Uranium	Leachate	0.647	2.33	5.39	1.22	16.2	17.1	16.7
	Filtrate	0.534	1.56	5.15	0.79	13.0	15.3	14.2
	Plate-Out	0.647	0.54	3.37	1.48	1.29	1.39	1.34
	Total	1.29	2.87	8.76	2.70	17.5	18.5	18.0
Plutonium	Leachate	0.058	0.124	0.0602	0.0946	0.206	0.131	0.169
	Filtrate	0.036	0.085	0.0880	0.0574	0.070	0.069	0.070
	Plate-Out	0.051	0.123	0.0704	0.0483	0.0335	0.199	0.116
	Total	0.109	0.247	0.131	0.143	0.240	0.330	0.285
Technetium	Leachate	0.523	23.3	29.1	28.0	24.1	30.6	27.4
	Filtrate	0.468	21.5	30.4	27.6	26.6	31.3	29.0
	Plate-Out	<0.0077	<0.089	0.348	<0.087	0.15	<0.15	0.15
	Total	0.531	23.3	29.4	28.0	24.3	30.6	27.5
Cesium	Leachate	81.1	79.0	109.0	66.5	74.5	81.6	78.1
	Filtrate	80.1	77.0	111.0	62.2	73.8	80.1	79.0
	Plate-Out	0.022	0.45	0.0197	0.0098	0.071	0.20	0.14
	Total	81.1	79.5	109.0	66.5	74.6	81.8	78.2
Measured pH	Leachate	5.0	5.0	4.7	4.9	4.4	4.3	
Corrected pH*	Leachate	6.9	6.9	6.6	6.8	6.3	6.2	

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*A correction of 0.9 for the liquid junction potential plus 1.0 for the electrode error described in the text was applied to these data.

TABLE 10. Data for 90°C Tests of Unclad Spent Fuel/Iron in PBB1 Brine

Element	Sample	Normalized Mass Loss (g/m^2) at Different Times (Days)										
		28	60	90				120	180			
				#1	#2	#3	Ave.		#1	#2	#3	Ave.
Uranium	Leachate	6.97	2.12	1.45	1.19	11.9	4.85	1.86	0.125	0.105	0.65	0.293
	Filtrate	1.00	0.648	0.107	0.139	0.24	0.162	0.339	0.314	0.141	0.051	0.169
	Plate-Out	1.45	9.11	8.46	4.07	1.59	4.71	1.55	3.30	1.17	0.060	1.51
	Iron	1.89	4.35	4.32	9.30	5.80	6.47	10.5	5.91	9.45	8.14	7.83
	Zirconium			0.002			0.002		0.014			0.014
	Total	10.3	15.6	14.2	14.6	19.3	16.0	13.9	9.35	10.7	9.39	9.81
Plutonium	Leachate	0.442	0.344	0.981	0.982	1.39	1.18	0.418	0.210	0.0306	0.146	0.129
	Filtrate	0.00096	0.00049	0.00046	0.00013	0.00026	0.00028	0.0004	0.001	0.0007	0.0004	0.0007
	Plate-Out	0.467	0.106	0.566	0.157	0.150	0.291	0.140	2.45	0.391	1.61	1.48
	Iron	0.005	0.0305	0.8084	0.0075	0.0086	0.0082	0.0078	0.019	0.0127	0.014	0.015
	Zirconium			0.0023			0.0023		0.01			0.01
	Total	0.914	0.481	1.56	1.15	1.55	1.42	0.566	2.68	0.434	1.77	1.63
Technetium	Leachate	7.97	1.62	1.46	1.19	11.2	4.62	8.46	4.62	1.79	4.54	3.65
	Filtrate	1.92	1.12	1.12	0.595	1.30	1.01	2.19	2.14	2.84	1.63	2.20
	Plate-Out	0.511	1.56	4.15	1.36	0.936	2.15	1.00	0.871	0.772	0.481	0.708
	Iron	3.89	7.09	15.3	20.8	16.0	17.4	19.0	11.1	13.8	12.0	12.3
	Zirconium			3.34			3.34		<0.18			<0.18
	Total	12.4	10.3	24.3	23.4	28.1	25.3	28.5	16.6	16.4	17.0	16.7
Cesium	Leachate	92.3	104	118	87.7	73.0	92.9	103	111	71.9	105	96.0
	Filtrate	91.4	102	121	90.8	72.4	94.7	102	105	69.3	104	92.8
	Plate-Out	0.601	0.075	0.039	0.019	0.024	0.027	0.0074	0.18	0.34	0.052	0.19
	Iron	0.027	0.029	0.055	0.071	0.029	0.052	0.057	0.12	0.079	0.105	0.10
	Zirconium			0.00072			0.00072		<0.004			<0.004
	Total	92.9	104	118	87.8	73.0	92.9	103	111	72.3	105	96.0
Measured pH	Leachate	4.9	5.0	4.7	4.9	4.8		4.9	4.5	4.5	4.4	
Corrected pH*	Leachate	6.8	6.9	6.6	6.8	6.7		6.8	6.4	6.4	6.3	

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*A correction of 0.9 for the liquid junction potential plus 1.0 for the electrode error described in the text was applied to these data.

TABLE 11. Data for 90°C Tests of UO_2 in PBB1 Brine

Sample Number	No. Days	Normalized Uranium Mass Loss (mg/m²)					Final* pH
		Leachate(1)	Plate-Out on Teflon(2)	Plate-Out on Iron(3)	Filtrate(1)	Total	
UO_2							
BU5 (4)	5	5.0	7.6		4.9	12.6	6.65
BU14 (4)	14	3.4	17.2		2.8	20.6	6.30
BU28A (4)	28	3.4	11.5		3.4	14.9	6.21
(5)		4.1					
BU28B (6)	28	3.1	28.4		3.0	31.5	6.24
(7)		3.2					
(8)					2.6		
UO_2-Fe							
BUF5 (4)	5	3.9	15.4	1.5	1.0	20.8	6.58
BUF14 (4)	14	9.5	35.9	2.2	0.3	47.6	5.92
BUF28A (4)	28	9.1	229	3.5	0.0	242	6.28
(5)		70.4					
BUF28B (6)	28	23.6	81.3	2.9	0.2	108	6.09
(7)		21.6	97.0	3.2	0.0		
(8)					0.0		
Oxidized UO_2							
OBU5 (4)	5	10.7	5.4		9.4	16.1	6.45
OBU14 (4)	14	6.5	4.7		6.8	11.2	6.44
OBU28A (4)	28	4.4	7.4		4.4	11.8	6.44
OBU28B (6)	28	7.8	16.2		4.7	24.0	6.46
(7)		7.2					
(8)					4.3		
Oxidized UO_2-Fe							
OBUF5 (4)	5	7.2	18.9	3.9	0.7	30.0	6.46
OBUF14 (4)	14	10.9	36.3	5.4	0.0	52.6	5.69
OBUF28A (4)	28	10.0	86.1	6.1	0.0	102	6.07
(5)		38.6					
OBUF288 (6)	28	45.4	39.4	10.9	0.2	95.7	5.87
(7)				13.4			
(8)					0.0		

(1) 36 ml sample
(2) 50 ml sample
(3) 25 ml sample
(4) analyzed immediately
(5) reanalyzed two weeks later
(6) set aside for one week before analyzing
(7) reanalyzed one week later
(8) reanalyzed three weeks later

*These values have been corrected for liquid junction potential; i.e., a value of 0.90 was added to the measured pH values.

TABLE 12. Data for 90°C Tests of UO_2 in Deionized Water

Sample Number		No. Days	Normalized Uranium Mass Loss (mg/m²)					Final pH
			Leachate(1)	Plate-Out on Teflon(2)	Plate-Out on Iron(3)	Filtrate(1)	Total	
UO_2								
WU5	(4)	5	51.6	14.2		22.4	65.8	5.38
WU14	(4)	14	31.2	155		10.0	186	5.47
WU28A	(4)	28	103	140		28.4	243	5.76
	(5)		106					
WU28B	(6)	28	77.2	169		19.3	246	5.64
	(7)		85.1	287		21.6		
	(8)					22.0		
UO_2-Fe								
WUF5	(4)	5	11.9	181	2.2	0.5	195	6.07
WUF14	(4)	14	35.4	227	6.1	0.8	269	5.91
WUF28A	(4)	28	90.8	136	51.2	1.1	278	6.42
WUF288	(6)	28	88.5	328	14.2	1.6	431	6.17
	(7)		107.8					
	(8)					0.8		
Oxidized UO_2								
OWU5	(4)	5	151	12.3		115	163	5.77
OWU14	(4)	14	140	118		77.2	258	5.11
OWU28A	(4)	28	312	539		244	851	5.50
	(5)		397					
OWU28B	(6)	28	848	336		697	1184	5.31
	(7)		1090					
	(8)					602		
Oxidized UO_2-Fe								
OWUF5	(4)	5	21.0	88.9	1.7	0.6	112	6.02
OWUF14	(4)	14	23.8	55.2	46.6	1.1	126	5.90
OWUF28A	(4)	28	72.6	218	74.9	2.5	366	6.39
	(5)		131					
OWUF288	(6)	28	184	232	52.5	1.6	469	5.99
	(7)		207					
	(8)					1.2		

(1) 36 ml sample
(2) 50 ml sample
(3) 25 ml sample
(4) analyzed immediately
(5) reanalyzed two weeks later
(6) set aside for one week before analyzing
(7) reanalyzed one week later
(8) reanalyzed three weeks later

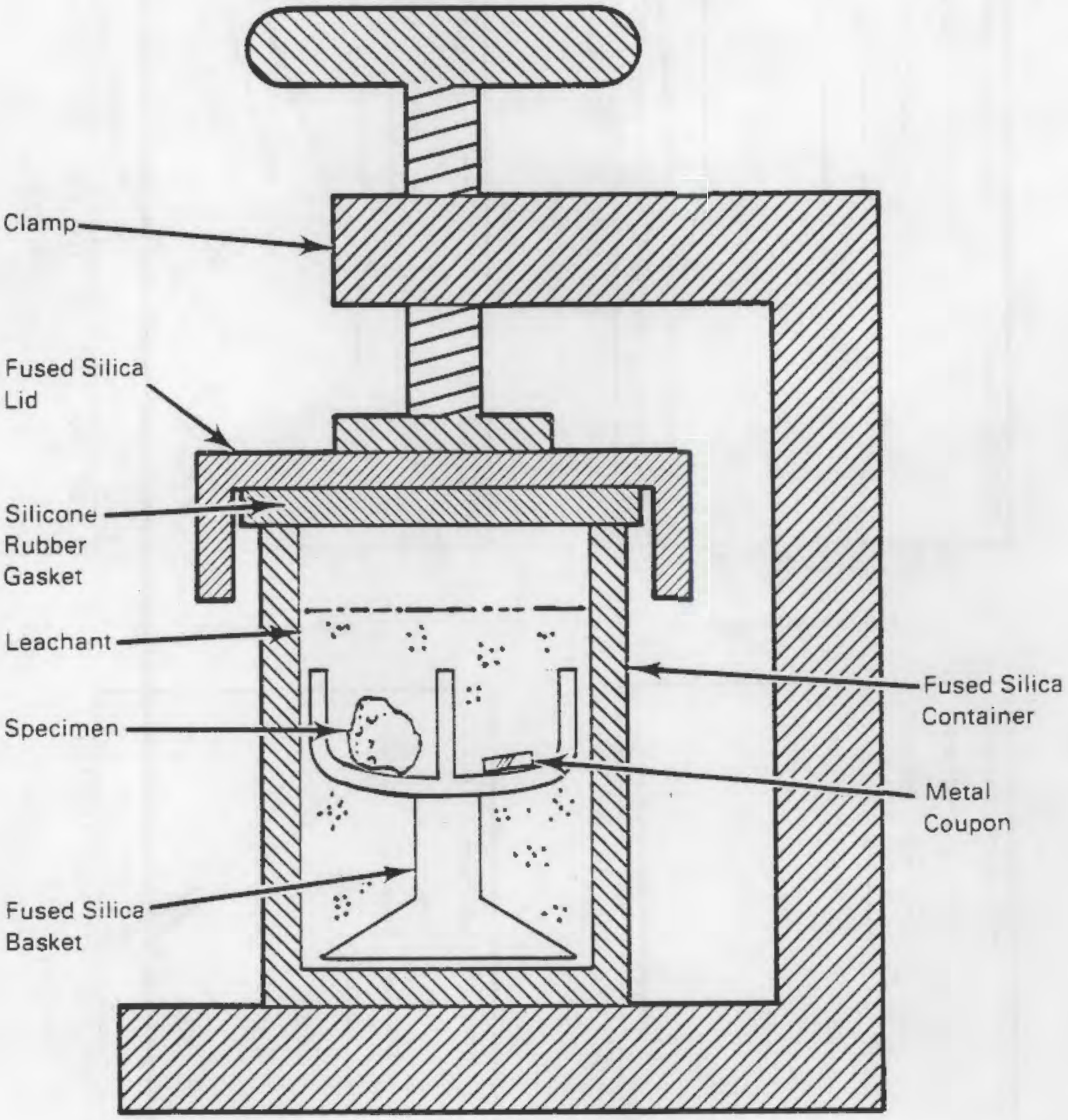

FIGURE 1. Schematic Diagram of Container Used for Ambient Temperature Spent Fuel Leach Tests

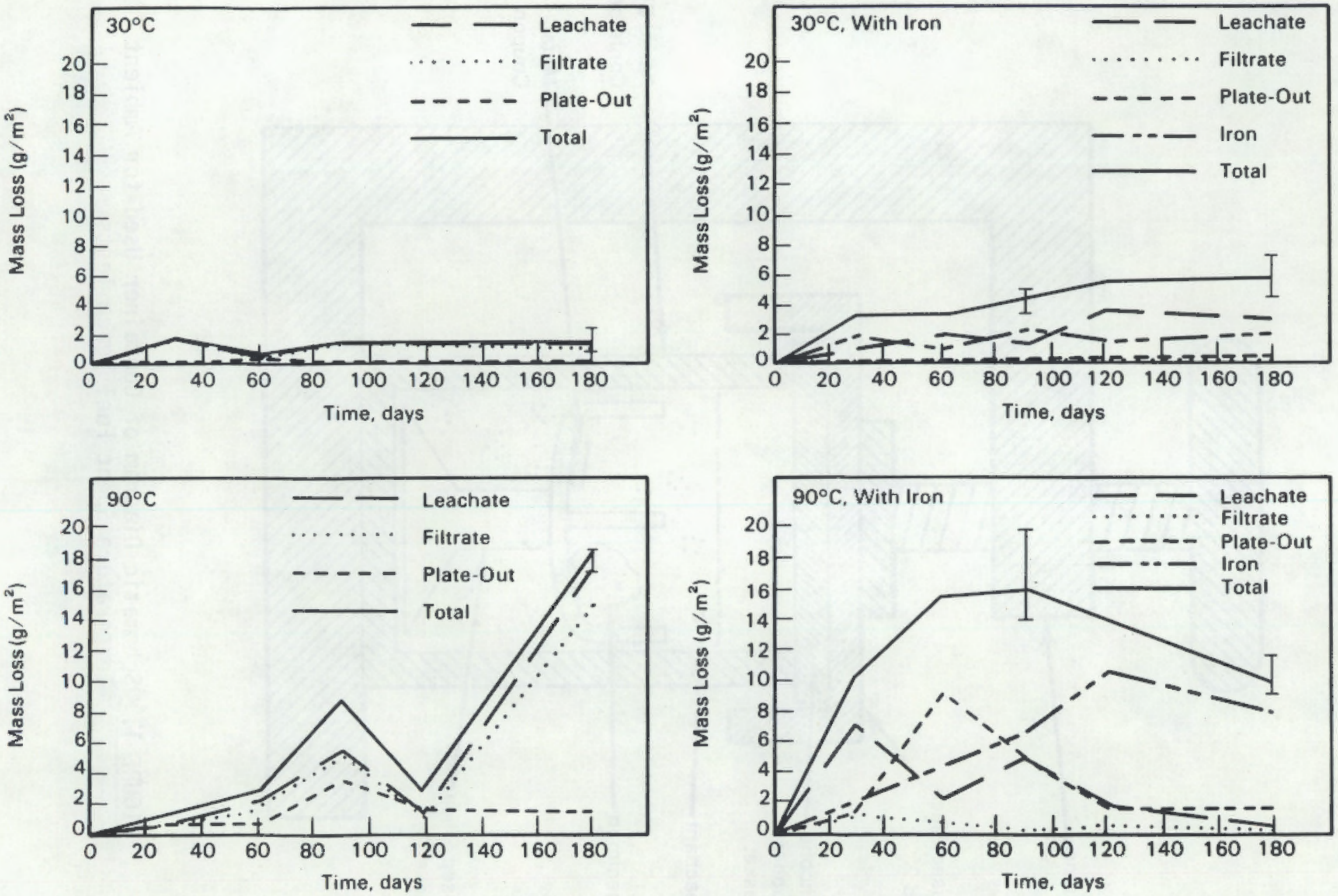

FIGURE 2. Normalized Uranium Mass Loss from Spent Fuel in PBB1 Brine

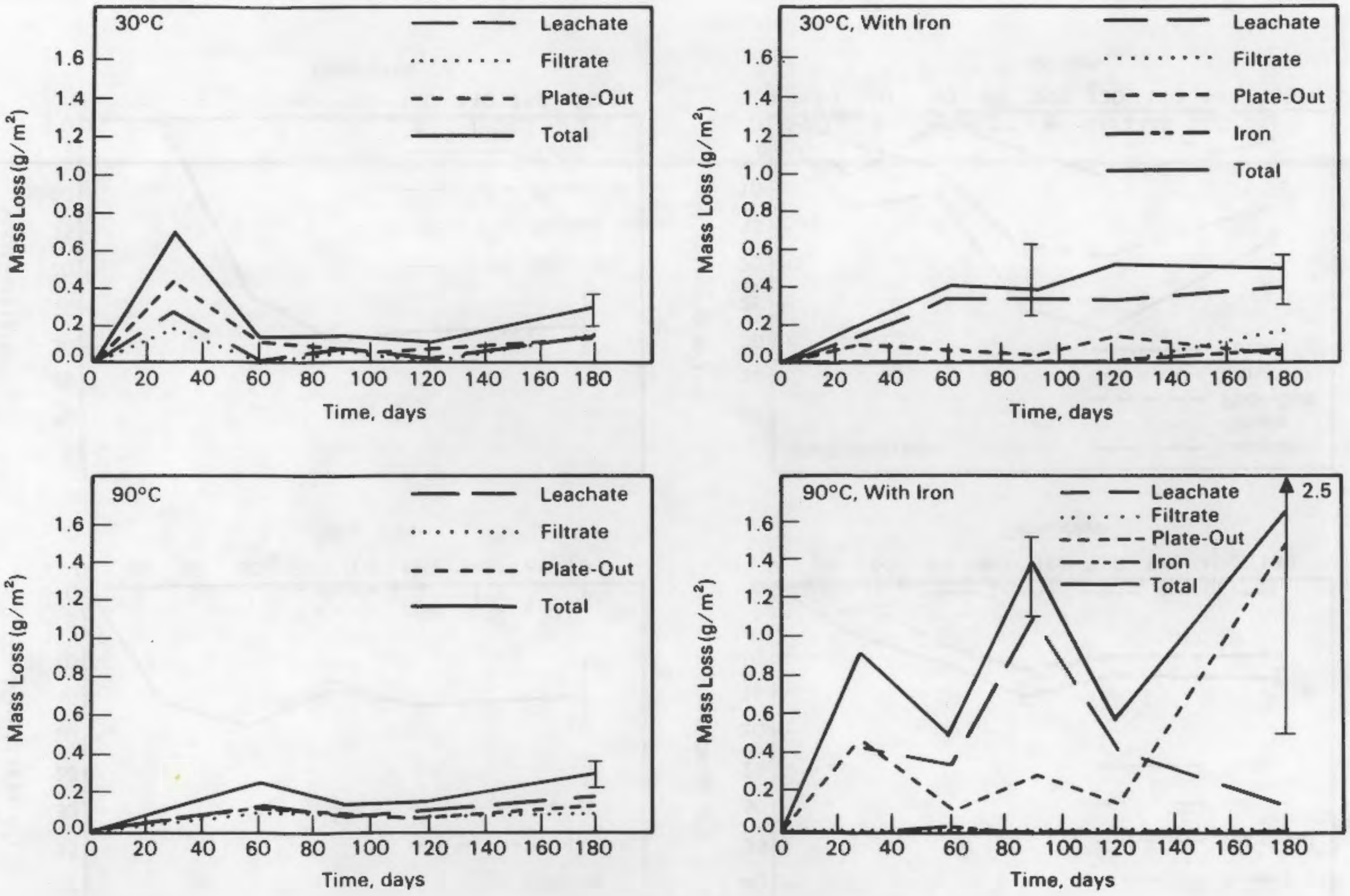

FIGURE 3. Normalized Plutonium Mass Loss from Spent Fuel in PBB1 Brine

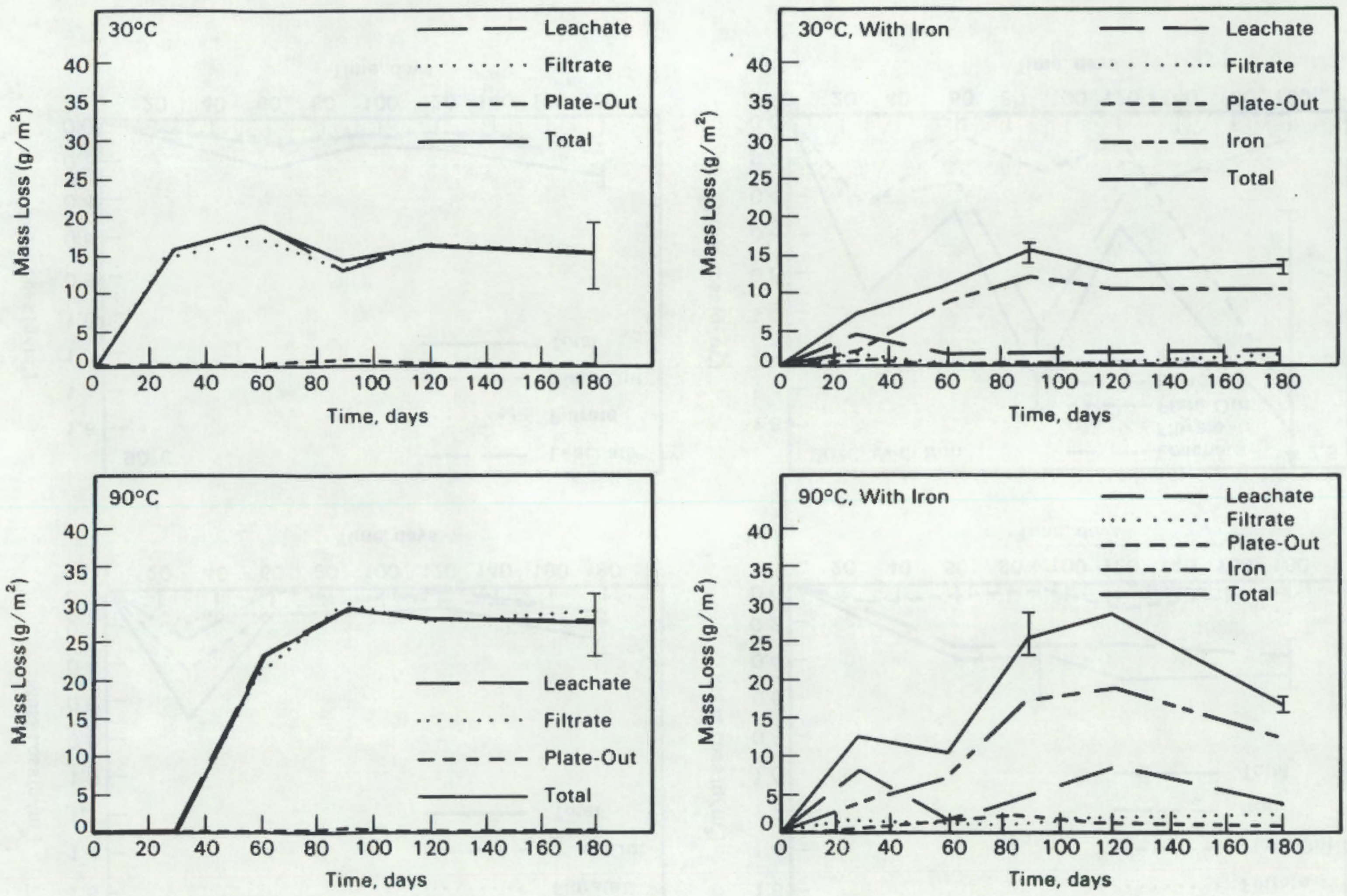

FIGURE 4. Normalized Technetium Mass Loss from Spent Fuel in PBB1 Brine

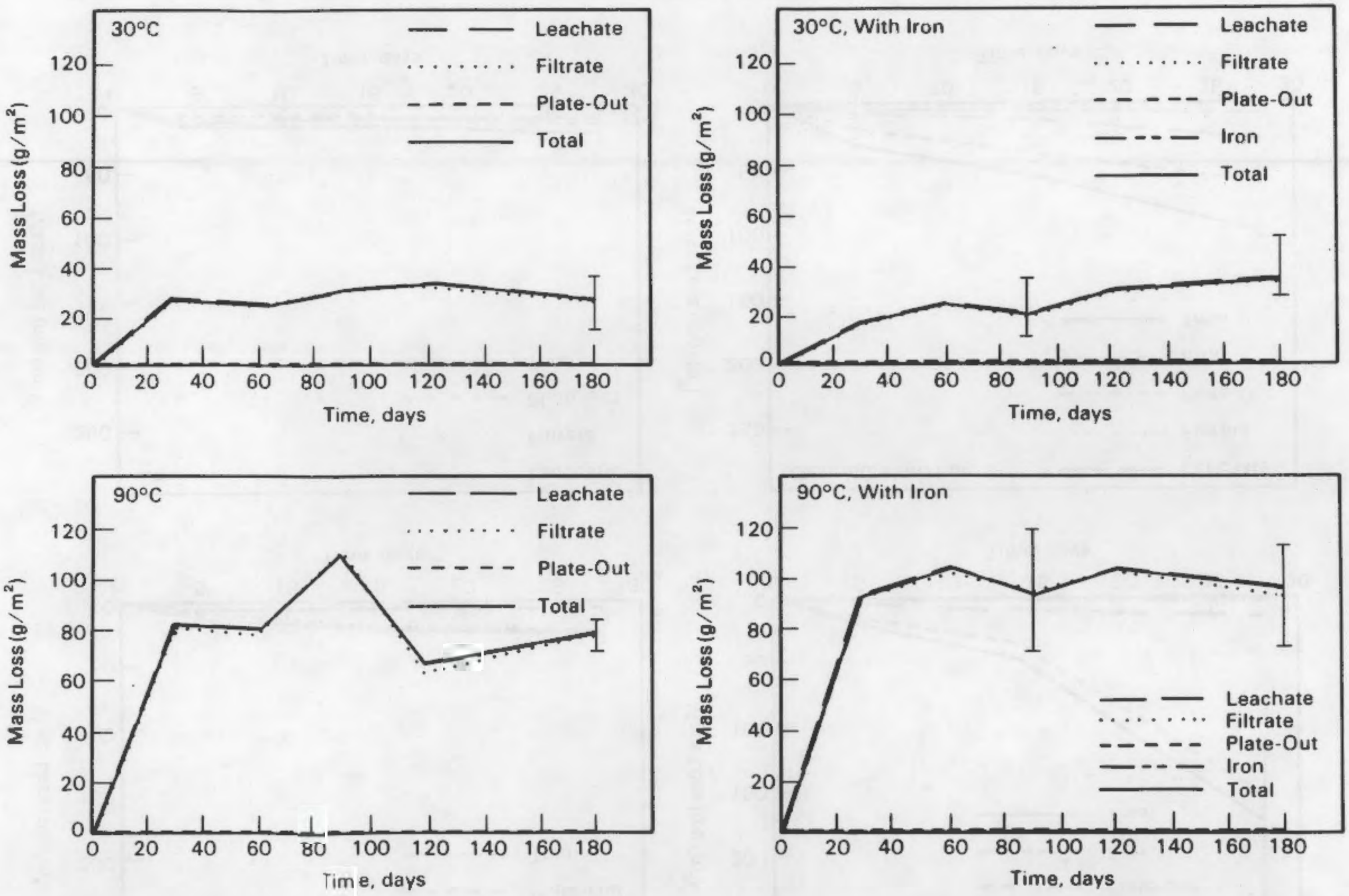

FIGURE 5. Normalized Cesium Mass Loss from Spent Fuel in PBB1 Brine

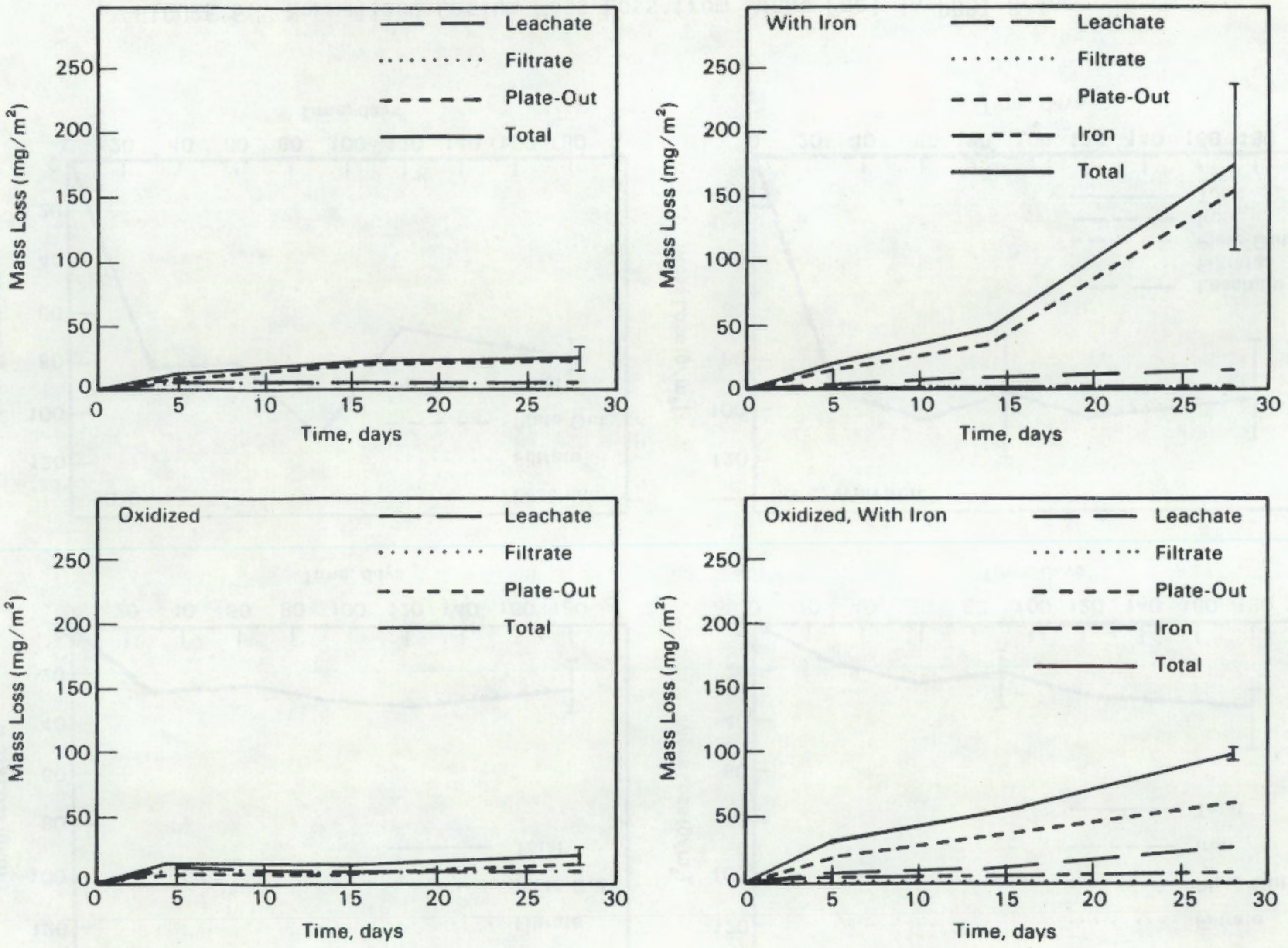

FIGURE 6. Normalized Uranium Mass Loss from UO_2 in 90°C PBB1 Brine

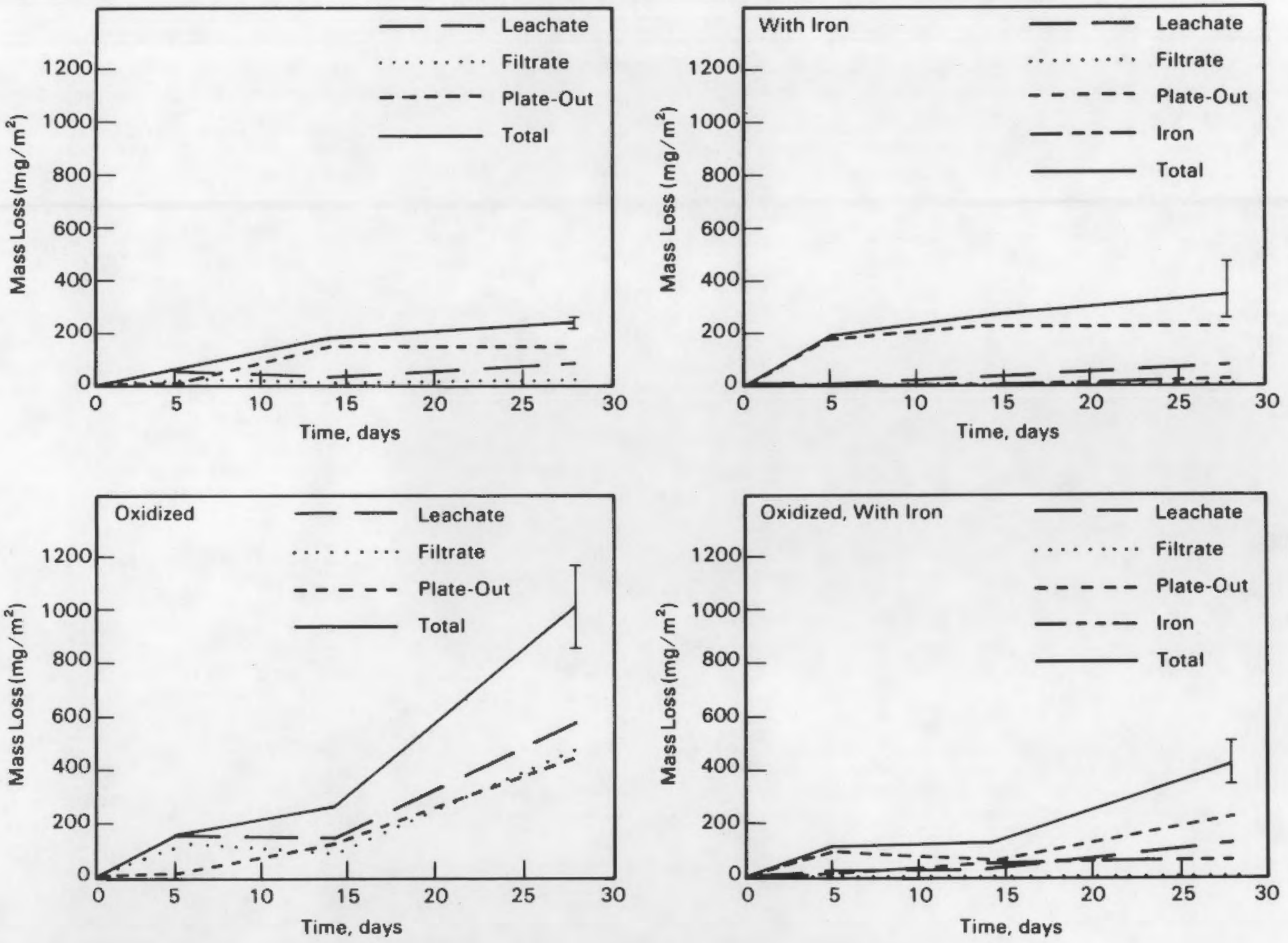

FIGURE 7. Normalized Uranium Mass Loss from UO_2 in 90°C Deionized Water

APPENDIX A

TABLE A.1. RAW URANIUM DATA FOR 30°C TESTS OF UNCLAD SPENT FUEL IN PBB1 BRINE

Sample Number	No. Days	Leachate Vol. (ml)(1)	Plate-Out Vol. (ml)(2)	Uranium Concentration (μg/ml)				
					Plate-Out on			
				Leachate	Quartz	Iron(3)	Zircaloy(3)	Filtrate
26	28	35	47	20	0.35			19
4	60	40	45	5.9	3.0			5.9
32	90	27	45	22	1.1			22
7	120	37	45	15	0.81			14
14	180	41	45	10	1.8			4.0
16	180	40	45	21	0.64			20
10	28	37	46	12	1.2	720		1.2
34	60	40	45	19	1.1	420		0.36
27	90	24	45	23	5.0	640	3.4	0.41
28	90	37	45	12	1.4	1000		0.18
11	90	37	45	13	1.2	820		0.28
3	120	37	45	35	2.0	601		0.43
1	180	35	45	40	6.4	743	0.60	6.4
8	180	37	45	16	1.7	924		0.15
17	180	39	45	30	2.2	688		0.23

(1) This volume applies to both leachate and filtrate.
(2) This volume applies to plate-out on the quartz only.
(3) Total μg per coupon.

TABLE A.2. RAW PLUTONIUM DATA FOR 30°C TESTS OF UNCLAD SPENT FUEL IN PBB1 BRINE

Sample Number	No. Days	Leachate Vol. (ml)(2)	Plate-Out Vol. (ml)(3)	Plutonium Activity (Bq/ml)(1)				
					Plate-Out on			
				Leachate	Quartz	Iron(4)	Zircaloy(4)	Filtrate
26	28	35	47	5.5	6.7			3.7
4	60	40	45	5.7	24			3.8
32	90	27	45	28	16			20
7	120	37	45	7.3	18			3.3
14	180	41	45	17	34			16
16	180	40	45	5B	27			56
10	28	37	46	34	19	47		0.073
34	60	40	45	82	15	110		0.13
27	90	24	45	218	19	51	13	0.083
28	90	37	45	59	11	52		0.18
11	90	37	45	48	12	38		0.083
3	120	37	45	92	33	223		0.38
1	180	35	45	146	0.23	328	11	132
8	180	37	45	58	14	115		0.27
17	180	39	45	112	19	210		0.67

(1) Total ^{239}Pu plus ^{240}Pu activity. This activity represents about 90% of the total Pu which has a specific activity of 4.0 GBq/g.

(2) This volume applies to both leachate and filtrate.

(3) This volume applies to plate-out on the quartz only.

(4) Total μg per coupon.

TABLE A.3. RAW TECHNETIUM DATA FOR 30°C TESTS OF UNCLAD SPENT FUEL IN PBB1 BRINE

Sample Number	No. Days	Leachate(2) Vol. (ml)	Plate-Out(3) Vol. (ml)	Technetium Activity (Bq/ml)(1) Leachate	Plate-Out on Quartz	Plate-Out on Iron(4)	Zircaloy(4)	Filtrate
26	28	35	47	82	<1.7			78
4	60	40	45	87	0.52			80
32	90	27	45	88	4.5			88
7	120	37	45	82	<0.33			83
14	180	41	45	55	<0.33			55
16	180	40	45	84	<0.33			85
10	28	37	46	22	2.5	430		10
34	60	40	45	8.8	0.88	1580		3.0
27	90	24	45	18	3.2	1830	400	3.0
28	90	37	45	7.2	<1	2170		1.8
11	90	37	45	8.7	2.3	2170		3.3
3	120	37	45	11	0.50	1920		4.0
1	180	35	45	8.5	2.3	1970	28	4.2
8	180	37	45	15	1.0	1520		7.5
17	180	39	45	11	0.32	1980		15

(1) Total ^{99}Tc activity. This activity represents 100% of the Tc which has a specific activity of 0.63 GBq/g.
(2) This volume applies to both leachate and filtrate.
(3) This volume applies to plate-out on the quartz only.
(4) Total μg per coupon.

TABLE A.4. RAW CESIUM DATA FOR 30°C TESTS OF UNCLAD SPENT FUEL IN PBB1 BRINE

				Cesium Activity (kBq/ml)(1)				
					Plate-Out on			
Sample Number	No. Days	Leachate(2) Vol. (ml)	Plate-Out(3) Vol. (ml)	Leachate	Quartz	Iron(4)	Zircaloy(4)	Filtrate
26	28	35	47	597	3.5			592
4	60	40	45	492	1.4			487
32	90	27	45	918	2.2			925
7	120	37	45	733	0.38			706
14	180	41	45	388	0.83			377
16	180	40	45	697	0.73			677
10	28	37	46	387	0.58	4.1		383
34	60	40	45	503	1.8	6.6		513
27	90	24	45	943	1.0	2.4	<0.58	828
28	90	37	45	342	0.61	1.0		342
11	90	37	45	305	0.69	6.6		308
3	120	37	45	655	1.4	4.4		637
1	180	35	45	973	0.60	96	<3.0	945
8	180	37	45	593	0.36	19		590
17	180	39	45	645	0.34	22		618

(1) Total ^{137}Cs activity. This activity represents about 42% of the total Cs which has a specific activity of 1350 GBq/g.
(2) This volume applies to both leachate and filtrate.
(3) This volume applies to plate-out on the quartz only.
(4) Total µg per coupon.

TABLE A.5. RAW URANIUM DATA FOR 90°C TESTS OF UNCLAD SPENT FUEL IN PBB1 BRINE

Sample Number	No. Days	Leachate(1) Vol. (ml)	Plate-Out(2) Vol. (ml)	Uranium Concentration (μg/ml)				
					Plate-Out on			
				Leachate	Quartz	Iron(3)	Zircaloy(3)	Filtrate
21	28	32	36	5.8	5.1			4.8
35	60	33	41	21	3.9			14
36	90	24	40	67	25			64
37	120	33	40	11	11			7.1
2	180	30	35	160	11			128
19	180	31	35	169	12			152
22	28	30	35	62	11	500		8.9
9	60	30	36	19	68	1170		5.8
38	90	25	35	15	56	1000	0.46	1.0
40	90	26	35	110	11	1400		2.2
5	90	28	35	11	30	2400		1.3
20	120	31	40	17	11	2970		3.1
18	180	24	35	1.2	22	1380	3.2	3.0
29	180	26	35	6.4	4.4	2100		0.50
12	180	28	35	1.0	8.8	2490		1.4

(1) This volume applies to both leachate and filtrate.
(2) This volume applies to plate-out on the quartz only.
(3) Total μg per coupon.

TABLE A.6. RAW PLUTONIUM DATA FOR 90°C TESTS OF UNCLAD SPENT FUEL IN PBB1 BRINE

Sample Number	No. Days	Leachate(2) Vol. (ml)	Plate-Out(3) Vol. (ml)	Plutonium Activity (Bq/ml)(1)				
					Plate-Out on			
				Leachate	Quartz	Iron(4)	Zircaloy(4)	Filtrate
21	28	32	36	14	11			8.8
35	60	33	41	31	25			21
36	90	24	40	21	14			30
37	120	33	40	23	9.8			14
2	180	30	35	56	7.8			19
19	180	31	35	36	47			19
22	28	30	35	108	97	37		0.23
9	60	30	36	85	22	225		0.1
38	90	25	35	248	103	53	15	0.1
40	90	26	35	350	28	57		0.07
5	90	28	35	248	32	53		0.03
20	120	31	40	105	27	60		0.1
18	180	24	35	55	448	121	62	0.30
29	180	26	35	39	325	102		0.1
12	180	28	35	8.0	81	92		0.18

(1) Total ^{239}Pu plus ^{240}Pu activity. This activity represents about 90% of the total Pu which has a specific activity of 4.0 GBq/g.
(2) This volume applies to both leachate and filtrate.
(3) This volume applies to plate-out on the quartz only.
(4) Total µg per coupon.

TABLE A.7. RAW TECHNETIUM DATA FOR 90°C TESTS OF UNCLAD SPENT FUEL IN PBB1 BRINE

Sample Number	No. Days	Leachate(2) Vol. (ml)	Plate-Out(3) Vol. (ml)	Technetium Activity (Bq/ml)(1)				
					Plate-Out on			
				Leachate	Quartz	Iron(4)	Zircaloy(4)	Filtrate
21	28	32	36	123	<1.7			115
35	60	33	41	108	<0.3			100
36	90	24	40	187	1.3			195
37	120	33	40	130	<0.3			128
2	180	30	35	123	<0.67			136
19	180	31	35	157	<0.67			160
22	28	30	35	37	2.0	530		8.8
9	60	30	36	7.5	6.0	983		5.2
38	90	25	35	7.0	14	1830	400	5.3
40	90	26	35	53	3.3	2000		6.2
5	90	28	35	5.7	5.2	2770		2.8
20	120	31	40	40	3.7	2780		10
18	180	24	35	23	3.0	1340	<22	11
29	180	26	35	23	1.8	1600		8.3
12	180	28	35	8.8	3.0	1880		14

(1) Total ^{99}Tc activity. This activity represents 100% of the Tc which has a specific activity of 0.63 GBq/g.
(2) This volume applies to both leachate and filtrate.
(3) This volume applies to plate-out on the quartz only.
(4) Total µg per coupon.

TABLE A.8. RAW CESIUM DATA FOR 90°C TESTS OF UNCLAD SPENT FUEL IN P8B1 BRINE

Sample Number	No. Days	Leachate Vol. (ml)(2)	Plate-Out Vol. (ml)(3)	Cesium Activity (kBq/ml)(1) Leachate	Plate-Out on Quartz	Plate-Out on Iron(4)	Plate-Out on Zircaloy(4)	Filtrate
21	28	32	36	1670	0.40			1650
35	60	33	41	1610	7.5			1590
36	90	24	40	3100	0.34			3170
37	120	33	40	1370	0.17			1280
2	180	30	35	1680	1.4			1670
19	180	31	35	1850	4.0			1820
22	28	30	35	1880	10	17		1870
9	60	30	36	2130	1.3	18		2100
38	90	25	35	2500	0.59	29	<0.38	2550
40	90	26	35	1540	0.38	16		1530
5	90	28	35	1850	0.33	42		1920
20	120	31	40	2170	0.12	37		2130
18	180	24	35	2430	2.7	63	<2.4	2300
29	180	26	35	2370	0.87	62		2330
12	180	28	35	1570	5.8	47		1510

(1) Total ^{137}Cs activity. This activity represents about 42% of the total Cs which has a specific activity of 1350 GBq/g.
(2) This volume applies to both leachate and filtrate.
(3) This volume applies to plate-out on the quartz only.
(4) Total μg per coupon.

TABLE A.9. Raw Data for 90°C Tests of UO_2 in PBB1 Brine

Sample Number		No. Days	Uranium Concentration, μg/ml				Final pH
			Leachate(1)	Plate-Out on		Filtrate(1)	
				Teflon(2)	Iron(3)		
UO_2							
BU5	(4)	5	0.044	0.048		0.043	5.75
BU14	(4)	14	0.030	0.109		0.025	5.40
BU28A	(4)	28	0.030	0.073		0.030	5.31
	(5)		0.036				
BU28B	(6)	28	0.027	0.180		0.026	5.34
	(7)		0.028				
	(8)					0.023	
UO_2-Fe							
BUF5	(4)	5	0.034	0.098	0.019	0.009	5.68
BUF14	(4)	14	0.084	0.228	0.028	0.003	5.02
BUF28A	(4)	28	0.080	1.450	0.044	0.000	5.38
	(5)		0.620				
BUF28B	(6)	28	0.208	0.516	0.037	0.002	5.19
	(7)		0.190	0.615	0.040	0.000	
	(8)					0.000	
Oxidized UO_2							
OBU5	(4)	5	0.094	0.034		0.083	5.55
OBU14	(4)	14	0.057	0.030		0.060	5.54
OBU28A	(4)	28	0.039	0.047		0.039	5.54
OBU288	(6)	28	0.069	0.103		0.041	5.56
	(7)		0.063				
	(8)					0.038	
Oxidized UO_2-Fe							
OBUF5	(4)	5	0.063	0.120	0.049	0.006	5.56
OBUF14	(4)	14	0.096	0.230	0.068	0.000	4.79
OBUF28A	(4)	28	0.088	0.546	0.078	0.000	5.17
	(5)		0.340				
OBUF28B	(6)	28	0.400	0.250	0.138	0.002	4.97
	(7)				0.170		
	(8)					0.000	

(1) 36 ml sample
(2) 50 ml sample
(3) 25 ml sample
(4) analyzed immediately
(5) reanalyzed two weeks later
(6) set aside for one week before analyzing
(7) reanalyzed one week later
(8) reanalyzed three weeks later

TABLE A.10. Raw Data for 90°C Tests of UO_2 in Deionized Water

Sample Number	No. Days	Uranium Concentration (µg/ml)				Final pH
			Plate-Out on			
		Leachate(1)	Teflon(2)	Iron(3)	Filtrate(1)	
UO_2						
WU5 (4)	5	0.455	0.090		0.197	5.38
WU14 (4)	14	0.275	0.985		0.088	5.47
WU28A (4)	28	0.910	0.888		0.250	5.76
(5)		0.936				
WU28B (6)	28	0.680	1.070		0.170	5.64
(7)		0.750	1.820		0.190	
(8)					0.194	
UO_2-Fe						
WUF5 (4)	5	0.105	1.150	0.028	0.004	6.07
WUF14 (4)	14	0.312	1.440	0.078	0.007	5.91
WUF28A (4)	28	0.800	0.860	0.650	0.010	6.42
WUF28B (6)	28	0.780	2.080	0.180	0.014	6.17
(7)		0.950				
(8)					0.007	
Oxidized UO_2						
OWU5 (4)	5	1.330	0.078		1.010	5.77
OWU14 (4)	14	1.230	0.750		0.680	5.11
OWU28A (4)	28	2.750	3.420		2.150	5.50
(5)		3.500				
OWU28B (6)	28	7.470	2.130		6.140	5.31
(7)		9.600				
(8)					5.300	
Oxidized UO_2-Fe						
OWUF5 (4)	5	0.185	0.564	0.021	0.005	6.02
OWUF14 (4)	14	0.210	0.350	0.591	0.010	5.90
OWUF28A (4)	28	0.640	1.380	0.950	0.022	6.39
(5)		1.150				
OWUF28B (6)	28	1.620	1.470	0.666	0.014	5.99
(7)		1.820				
(8)					0.011	

(1) 36 ml sample
(2) 50 ml sample
(3) 25 ml sample
(4) analyzed immediately
(5) reanalyzed two weeks later
(6) set aside for one week before analyzing
(7) reanalyzed one week later
(8) reanalyzed three weeks later

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