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RESULTS FROM NNWSI SERIES 2
BARE FUEL DISSOLUTION TESTS

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SUMMARY

The dissolution and radionuclide release behavior of spent fuel in groundwater is being studied by the Nevada Nuclear Waste Storage Investigations (NNWSI) Project. Two bare spent fuel specimens plus the empty cladding hulls were tested in NNWSI J-13 well water in unsealed fused silica vessels under ambient hot cell air conditions (25°C) in the currently reported tests. One of the specimens was prepared from a rod irradiated in the H. B. Robinson Unit 2 reactor and the other from a rod irradiated in the Turkey Point Unit 3 reactor. Both fuels were low gas release and moderate burnup. The specimen particle size range (2 to 3 mm) was that which occurs in the fuel as a result of thermal cracking. A semi-static test method was used in which the specimens were tested for multiple cycles starting in fresh water with periodic water samples taken during each cycle. The specimens were tested for five cycles for a total time of 34 months.

Results indicate that most radionuclides of interest fall into three groups for release modeling. The first group principally includes the actinides (U, Np, Pu, Am, and Cm), all of which reached solubility-limited concentrations that were orders of magnitude below those necessary to meet the NRC 10 CFR 60.113 release limits for any realistic water flux predicted for the Yucca Mountain repository site. The second group is nuclides of soluble elements such as Cs, Tc, and I, for which release rates do not appear to be solubility-limited and may depend on the dissolution rate of fuel. In later test cycles, ^{137}Cs , ^{90}Sr , ^{99}Tc , and ^{129}I were continuously released at rates between about 5×10^{-5} and 1×10^{-4} of inventory per year. It appeared that these soluble nuclide release rates may not have greatly exceeded the dissolution rate of the fuel matrix phase in the later test cycles. The third group is radionuclides that may be transported in the vapor phase, of which ^{14}C is of primary concern. Detailed test results are presented and discussed.

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The NNWSI Series 2 Spent Fuel Dissolution Tests were conducted at the Hanford Engineering Development Laboratory, which was operated for the DOE by Westinghouse Hanford Company (WHC) under Contract DE-AC06-76FF02170. The NNWSI work along with most of the personnel involved were transferred from WHC to Pacific Northwest Laboratory (PNL) on June 29, 1987, as part of the DOE Hanford Site consolidation. PNL is operated for the DOE by Battelle Memorial Institute under Contract DE-AC06-76RLO 1830. Laboratory activities associated with the Series 2 tests were in general completed before transfer to PNL. Much of the data evaluation and preparation of this report occurred after transfer to PNL. Many people were involved in different aspects of the NNWSI Series 2 Spent Fuel Dissolution Tests. The following list identifies the principal contributors.

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ACRONYMS

BCL	Battelle Columbus Laboratories
cpm	counts per minute
DOE	U.S. Department of Energy
EDS	energy-dispersive spectrometry
HBR	H. B. Robinson (reactor)
IC	ion chromatography
ICP	inductively coupled plasma (emission spectrometry)
JCPDS	Joint Committee on Powder Diffraction Standards
LLNL	Lawrence Livermore National Laboratory
LWR	light water reactor
MCC	Materials Characterization Center
NNWSI	Nevada Nuclear Waste Storage Investigations
NRC	Nuclear Regulatory Commission
PNL	Pacific Northwest Laboratory
ppb	parts per billion (mass basis)
ppm	parts per million (mass basis)
PWR	pressurized water reactor
SEM	scanning electron microscopy
TP	Turkey Point (reactor)
YMP	Yucca Mountain Project
WHC	Westinghouse Hanford Company
XRD	X-ray diffraction

1.0 BACKGROUND

The Yucca Mountain Project^(a) (YMP) is investigating the suitability of the Topopah Spring Tuff at Yucca Mountain, Nevada, for potential use as a disposal site for spent nuclear fuel and other high-level waste forms. The repository horizon under study lies ~200 m to 400 m above the water table in the unsaturated zone. Contact of the spent fuel by liquid water will not occur until the repository has cooled below the 95°C boiling temperature at the repository elevation. At that time, which is predicted to be hundreds of years after disposal, a limited quantity of water infiltrating the rock could potentially enter a failed waste container and contact the spent fuel where cladding failures have also occurred. Migration of a limited quantity of such water from a failed waste container is considered to be the most probable mechanism for radionuclide release. In addition, there is the potential that ^{14}C (as CO_2) and possibly ^{129}I (as I_2) may migrate in the vapor phase.

Lawrence Livermore National Laboratory (LLNL) is the lead contractor for the Waste Package Task of NNWSI. Westinghouse Hanford Company (WHC) became a subcontractor to LLNL in 1984, assisting them in determining the requirements for successful disposal of spent fuel at the Yucca Mountain Site.^(b) The work at WHC focused primarily on hot cell testing with spent fuel materials. Areas of investigation included leaching/dissolution behavior, cladding corrosion, and spent fuel low-temperature oxidation behavior. In the Spent Fuel Leaching/Dissolution Task at WHC, three laboratory test series were conducted with pressurized water reactor (PWR) spent fuel specimens to characterize radionuclide release under NNWSI-relevant conditions.

In the Series 1 tests,⁽¹⁾ specimens prepared from Turkey Point Reactor Unit 3 fuel were tested in deionized distilled water in unsealed fused silica

(a) Formerly the Nevada Nuclear Waste Investigations (NNWSI) Project.

(b) This work was transferred from Westinghouse Hanford Company to Pacific Northwest Laboratory (PNL) on June 29, 1987, as part of the U.S. Department of Energy Hanford Site Consolidation.

vessels under ambient hot cell air and temperature^(a) conditions. Four specimen configurations were tested: 1) undefected fuel rod segments with water-tight end fittings, 2) fuel rod segments containing small (~200- μ m diameter) laser-drilled holes through the cladding and with water-tight end fittings, 3) fuel rod segments with a machined slit through the cladding and water-tight end fittings, and 4) bare fuel particles removed from the cladding plus the cladding hulls. A "semi-static" test procedure was developed in which periodic solution samples were taken with the sample volume replenished with fresh deionized distilled water. Cycle 1 of the Series 1 tests was started during July 1983 and was 240 days in duration. At the end of the first cycle the tests were sampled, the vessels stripped in 8 M HNO_3 , and the specimens restarted in fresh deionized distilled water for a second cycle. Cycle 2 of the Series 1 tests was terminated at 128 days in July 1984.

The Series 2 tests were similar to the Series 1 tests except that: 1) the Series 2 tests were run in NNWSI reference J-13 well water, 2) each of the four specimen configurations was duplicated using both the Turkey Point Reactor and H. B. Robinson Reactor PWR spent fuels, and 3) a vessel and specimen rinse procedure was added to the cycle termination procedures. Filtration of the collected rinse solution provided solids residues that were later examined for secondary-phase formation. Cycle 1 of the Series 2 tests was started in June 1984. All eight Series 2 specimens were run for a second cycle. Results from Cycles 1 and 2 of the Series 2 tests (all eight specimens) were reported in Reference 2. The two bare fuel specimens were continued for Cycles 3, 4, and 5. Cycle 5 of the Series 2 bare fuel tests was terminated in June 1987 for a total five-cycle testing time of ~34 months. Results from all five cycles of the Series 2 bare fuel specimens are reported in this report.

The Series 3 tests were run for three cycles during the same approximate time period as Cycles 3, 4, and 5 of the Series 2 tests. The Series 3 tests were run in sealed stainless steel vessels and used the same four-specimen configurations used in Series 1 and Series 2 Cycles 1 and 2. Five specimens

(a) Hot cell temperature range is about 21°C to 28°C depending on time of year and time of day. An average value of 25°C was assumed for these ambient temperature tests.

(one each of the four configurations using H. B. Robinson fuel plus an additional bare fuel specimen using Turkey Point fuel) were tested at 85°C, and a sixth specimen (H. B. Robinson bare fuel) was run at 25°C. Detailed results from the Series 3 tests are reported in Reference 3. Two additional scoping tests using preoxidized bare fuel specimens in Series-2-type silica vessels were started in August 1986. Selected results from Cycle 1 and from initial Cycle 2 samples from the oxidized fuel scoping tests were reported in Reference 4. The Series 1 and 2 tests were originally entitled "Cladding Containment Credit Tests." All of the test series were later referred to as "Spent Fuel Dissolution Tests."

This work has been conducted under NNWSI work breakdown structure (WBS) element number 1.2.2.3.1.1.L and activity D-20-42 of the Scientific Investigation Plan for NNWSI Waste Form Testing.⁽⁵⁾ Except where noted, the Series 2 and Series 3 work has been conducted at Quality Level Assignment I.

2.0 TEST DESCRIPTION

A detailed description of the Series 2 tests is provided by the test plan⁽⁶⁾ and in the Cycles 1 and 2 report.⁽²⁾ The intent of this section is to provide the reader sufficient information about the test methods and fuel specimens to follow the discussion of results (Section 3.0). The Series 2 tests were conducted in unsealed silica vessels under ambient hot cell air and temperature conditions. The bare fuel specimen and vessel configuration are shown in Figure 2.1.

Cycle 2 was started the day after Cycle 1 termination, reusing the same vessels. The decision to continue testing the two Series 2 bare fuel specimens for additional cycles was made after completing Cycle 2. The time period between Cycle 2 termination and Cycle 3 start was 14 days for the H. B. Robinson (HBR) bare fuel test and 7 days for the Turkey Point (TP) bare fuel test, during which time the fuel was allowed to dry in air. New vessels were used for Cycles 1, 3, 4, and 5. Cycles 4 and 5 were started the same day the previous cycle was terminated.

2.1 TEST SPECIMENS

The two Series 2 bare fuel specimens were prepared from fuel rod segments irradiated in H. B. Robinson Unit 2 and Turkey Point Unit 3 pressurized water reactors. These two fuel specimens are henceforth referred to as HBR and TP in this report. Principal characteristics of the two fuels are given in Table 2.1. Both fuels are from similar Westinghouse 15 x 15 assemblies. Probably the most significant differences between the two fuels from the standpoint of dissolution testing are the difference in grain size and period of air exposure prior to testing for the TP fuel in comparison to the HBR fuel.

The HBR fuel was obtained through the PNL Materials Characterization Center (MCC) as an "approved testing material" (ATM) for waste form testing and was identified as ATM-101.⁽⁷⁾ The HBR bare fuel test specimen was prepared from a 5-in.-long rod segment (C5C-H) taken near the axial midpoint of the rod and away from burnup depression regions at the axial location of assembly spacer grids. The C5 HBR rod was originally sectioned in 1983 a few

FIGURE 2.1. Series 2 Bare Fuel Test Configuration

TABLE 2.1. Characteristics of H. B. Robinson Unit 2 and Turkey Point Unit 3 Fuels

<u>Characteristic</u>	<u>H. B. Robinson</u>	<u>Turkey Point</u>
Fuel type	PWR 15 x 15	PWR 15 x 15
Assembly identification	B0-5	B-17
Rod identification	C5	F6
Discharge date	May 6, 1974	November 25, 1975
Nominal burnup	30 MWd/kgU	27 MWd/kgU
Fission gas release	0.2%	0.3%
Initial enrichment	2.55 wt% ²³⁵ U	2.559 wt% ²³⁵ U
Initial pellet density	92% TD (UO ₂)	92% TD (UO ₂)
Initial fuel grain size	~6 μm	~25 μm
Initial rod diameter	10.7 mm OD	10.7 mm OD
Cladding material	Zircaloy-4	Zircaloy-4
Cladding thickness	0.62 mm	0.62 mm
PNL-MCC identification	ATM-101	--

months prior to preparation of the present NNWSI Series 2 test specimens. The rod sections were stored in sealed stainless steel tubes (air atmosphere) until preparation of the test specimens just prior to starting the Series 2 tests in June 1984. The TP fuel specimen was prepared from one of several 5-in. sections from rods previously sectioned at Battelle Columbus Laboratories (BCL) for stress-rupture testing that had been planned to occur at Hanford.⁽⁸⁾ The section used for the Series 2 TP bare fuel specimen was I-9-24 from the axial position 29 to 34 in. from the top of Rod I-9. The I-9 rod was originally sectioned at BCL in 1979 and the sections were stored in sealed metal tubes (air atmosphere) until preparation of the present specimens just prior to starting the Series 2 tests.

The fuel particle specimen weights for each test cycle are given in Table 2.2. Fuel was removed from both ends of the stress-rupture rod segments at BCL; and, therefore, the fuel weight for the TP specimen was less than that of the HBR specimen. The reductions in weight from cycle to cycle are due to removal of particles for ceramographic examination at the ends of the first three test cycles, to loss of loose grains rinsed from the specimens between

TABLE 2.2. Specimen Fuel Weights (g)

<u>Specimen</u>	<u>Cycle 1</u>	<u>Cycle 2</u>	<u>Cycle 3</u>	<u>Cycle 4</u>	<u>Cycle 5</u>
HBR	83.10	81.92	81.45	81.03	80.85
TP	27.21	26.66	26.13	26.02	25.96

cycles, and (to a smaller degree) to dissolution weight loss. Dry specimen weights were obtained at the start of Cycles 1 and 3 and after Cycle 5. Specimen weights for Cycles 2, 4, and 5 were estimated by considering rinse residue and ceramographic sample weights. The mass of the cladding hulls was about 16.4 g for all cycles with both specimens.

Inventories for the radionuclides analyzed were calculated from ORIGEN-2 data tabulated in Appendix E of Reference 7. The calculated inventory values along with radiochemically measured values on an H. B. Robinson fuel sample are given in Table 2.3. The average times from discharge assumed (12 years for H. B. Robinson and 10.5 years for Turkey Point) correspond to midway through Cycle 3. Linear interpolation was used to adjust the tabulated ORIGEN-2 data for burnup and time from discharge. Fractional release calculations for this report use a single ORIGEN-2 based inventory value for each radionuclide and are not corrected for changes in inventory during the 3-yr testing period. Of the principal radionuclides discussed, ^{244}Cm inventory would change the most during the testing period, varying from ~4% more to ~7% less than the Table 2.3 value from the beginning of Cycle 1 through the end of Cycle 5. The specimen inventories used in fractional release calculations were obtained by multiplying the per gram inventories in Table 2.3 times the specimen weights given in Table 2.2.

2.2 TEST SAMPLES AND ANALYSES

The sampling schedule and specified analyses for Cycle 1 were given in the test plan.⁽⁶⁾ Sampling and analysis schedules were specified for Cycles 2 through 5 by memoranda. A summary of types of samples, sampling procedures, and analyses performed follows. Identifications and summaries of analytical procedures used are contained in Appendix D of PNL-7170.⁽³⁾

TABLE 2.3. Series 2 Specimen Radionuclide Inventories ($\mu\text{Ci/g}$ of fuel except as indicated)

Nuclide	H. B. Robinson ORIGEN-2 ^(a)	H. B. Robinson Measured ^(b)	Turkey Point ORIGEN-2 ^(a)
Burnup (MWd/kgM)	30.2 ^(b)	30.2	27.5 ^(c)
Uranium ($\mu\text{g/g}$ of fuel)	8.45×10^5	--	8.48×10^5
²⁴⁴ Cm	1.28×10^3	1.43×10^3 ^(d)	9.90×10^2
²⁴¹ Am	1.77×10^3	1.63×10^3	1.51×10^3
²³⁹⁺²⁴⁰ Pu	7.44×10^2	7.16×10^2	7.04×10^2
²³⁷ Np	2.43×10^{-1}	2.35×10^{-1}	2.18×10^{-1}
¹³⁷ Cs	6.37×10^4	6.57×10^4	6.04×10^4
¹²⁹ I	2.65×10^{-2}	--	2.42×10^{-2}
¹²⁶ Sn	6.74×10^{-1}	--	6.11×10^{-1}
⁹⁹ Tc	1.05×10^1	8.34×10^0	9.74×10^0
⁹⁰ Sr	4.17×10^4	--	4.03×10^4
¹⁴ C	--	(e)	--

(a) Calculated from ORIGEN-2 data in PNL-5109,⁽⁷⁾ assuming 12 years from discharge for H. B. Robinson and 10.5 years from discharge for Turkey Point.

(b) Radiochemically determined (September 1985) from Sample C5C-D.

(c) Reported burnup for Sample G7-15.⁽⁸⁾

(d) Actually ²⁴³⁺²⁴⁴Cm since both isotopes have similar alpha energies; ORIGEN-2 data indicate that ²⁴³Cm is ~1% of ²⁴³⁺²⁴⁴Cm.

(e) ¹⁴C average of values measured on Samples C5C-J and C5B-C:

Fuel = $0.49 \mu\text{Ci/g}$

Cladding = $0.53 \mu\text{Ci/g}$.

2.2.1 Starting J-13 Water

At the beginning of each test cycle the J-13 water used to start the cycle was analyzed. The following analyses were performed: pH, inductively coupled plasma (ICP) emission spectrometry for cations, ion chromatography (IC) for anions, and inorganic carbon for bicarbonate ion concentration calculation. Results of J-13 water analyses are contained in Appendix B.

2.2.2 Periodic Solution Samples

Periodic solution samples were taken using preleached glass pipettes attached to syringes. The sampling depth was slightly higher than the upper

lip of the internal bare-fuel sample basket, which is shown to scale in Figure 2.1. Before drawing the sample, ~50 mL of air were bubbled through the vessel from the syringe at the sampling depth. The original purpose of the bubbling in the Series 1 tests was to provide for a small amount of convection or mixing effect just prior to sampling. Although this procedure is probably of questionable value for mixing, it was retained so that the sampling procedure would be consistent in all test series. Periodic solution sample volumes ranged from 10 mL to 30 mL, depending on the specified analyses. After the sample was removed, the sample volume was replenished with fresh J-13 well water.

Solution samples were placed in preleached glass vials and capped in the hot cell. The samples were removed from the hot cell to a glovebox; and aliquots were prepared for analysis, usually within an hour of sampling. The first step when the sample vial was opened was to measure pH on an aliquot from the vial. Aliquots were also taken and placed in sealed vials for ^{14}C and ^{129}I analyses, if specified. The remaining sample was then usually separated into aliquots for the unfiltered, 0.4- μm filtered, and 18-Å filtered^(a) fractions. Analysis of 18-Å filtered solution samples was deleted after the 148-day Cycle 4 sample. Uranium, alpha (for $^{239+240}\text{Pu}$), gamma (for ^{137}Cs), ^{241}Am , ^{237}Np , ^{126}Sn , and ^{99}Tc analyses were usually performed on all three filtered fractions; and ^{129}I , ^{90}Sr , and ^{14}C analyses were usually performed on the unfiltered fractions. Solution chemistry (ICP, IC, and inorganic carbon) analyses were performed on the 0.4- μm filtered fraction, when specified. Not all analyses were performed on every solution sample. For sampling schedules, volumes and analyses performed refer to the radiochemistry and solution chemistry data tables in Appendixes A and B.

(a) What is referred to as an 18-Å filtered fraction in this report is a sample filtered through a centrifuge membrane cone filter (Amicon Corp., Lexington, MA, Model CF-25) that, according to the manufacturer, filters molecular weights above ~25,000. This filter has traditionally been referred to as "18 Å" in the WHC chemistry laboratory, apparently because an 18-Å-diameter fuel particle would have a molecular weight on the order of 25,000.

2.2.3 Rod Samples

Several 3-mm-diameter fused silica rods were included in Cycles 1, 2, and 3. Individual rods were periodically removed and were stripped using 10 mL of 8 M HNO_3 ; and the acid strip solution was analyzed. The purpose of the rod samples was to monitor the amount of radionuclide "plate-out" during the test cycles. The rod data indicated that essentially all nuclide plate-out occurred early in the test cycles and the rods were deleted in Cycles 4 and 5. Additional precipitation of nuclides as phases that did not adhere to the rods may have occurred. (Samples of these phases were presumed to be removed in the cycle termination rinse procedure.)

2.2.4 Final Solution Samples

A final solution sample was taken immediately before termination of a test cycle on the termination day. The procedure for the final solution sampling is identical to that for the periodic solution samples except that the sample volume was not replenished with fresh J-13 well water. For the purposes of data evaluation, the volume of the final solution sample is assumed to be the entire 250 mL of solution in the test.

2.2.5 Rinse Samples

After the final solution sample was taken, the bare-fuel particles were removed to a 250-mL beaker, and the remaining final solution was decanted off. The fuel particles were rinsed in the 250-mL beaker with J-13 water (~50 mL), rocking the beaker from side to side ten times and allowing the particles to tumble in the bottom of the beaker. The bare-fuel rinse water was then decanted into a 1000-mL beaker, and the bare-fuel rinse was repeated four more times. The bare-fuel rinse solution routinely became dark and turbid in appearance during the first few rinse cycles as surface grains (loosened by grain boundary dissolution) and sediment from the test became temporarily suspended. Specimen baskets and interior vessel surfaces were thoroughly rinsed with J-13 water from a squirt bottle, and the rinse water drained into the 1000-mL beaker. The rinse water volumes in the 1000-mL beakers were made up to 600 mL after specimen and vessel rinsing by addition of fresh J-13 well water and then were left covered overnight to settle. A sample was removed from the top of the settled rinse solutions the next morning. Routine

requested analyses for the rinse solution samples included uranium, alpha spectrometry, gamma spectrometry, ^{241}Am , ^{237}Np , ^{99}Tc , and ^{14}C . Rinse samples were 0.4- μm filtered prior to the analyses.

2.2.6 Acid Strip Samples

After rinsing, the specimen baskets and any remaining fused silica rods were placed back in their respective vessels; and 300 mL of 8 M HNO_3 were added. The next day the acid was poured into a bottle, back into the vessel, and then back into the bottle again. The acid strip solution in the bottles was then sampled for analysis. Requested analyses routinely included uranium alpha spectrometry, gamma spectrometry, ^{241}Am , ^{237}Np , and ^{99}Tc .

2.2.7 Ceramographic Samples

Random fuel particles were removed from the 250-mL fuel rinse beakers for ceramographic examination. The particles were mounted in resin, ground to expose an internal section, polished, and examined in the as-polished condition. The intersection of the section plane with the particle surface was of primary interest and was examined for evidence of grain boundary dissolution or any other type of observable preferential dissolution. The Cycle 3 TP ceramographic sample was deleted so as not to further lower the fuel inventory in the TP test. No ceramographic samples were taken from Cycles 4 and 5 because very little of interest was observed on previous samples. Three particles were taken for the HBR Cycle 1 sample, and two particles were taken for all other ceramographic samples.

2.2.8 Rinse Filters

After the rinse solution samples were taken, the remaining solutions in the 1000-mL beakers were stirred to get the finer sediments back into suspension. The rinse solutions were then filtered through 0.4- μm filters, and the filtrate solutions were discarded. The filters were weighed to determine the net amount of residue filtered from each rinse solution. The filters were later examined by scanning electron microscopy (SEM) and x-ray diffraction (XRD) to identify and characterize secondary solid phases. SEM examination included energy-dispersive spectrometry (EDS) analysis of selected particles or phases.

2.2.9 Coarse Rinse Sediments

Coarser particles that would not remain in suspension long enough to be decanted off during filtration of the rinse solution were allowed to settle back to the bottom of the 1000-mL rinse collection beakers. These sediments were allowed to dry in the 1000-mL beaker and were removed and weighed after Cycles 1, 3 and 4. Weights for the coarse rinse sediments, along with weights of material collected on the rinse filters and ceramographic sample weights, are given in Table 2.4. Samples of these coarse sediments were then examined in the SEM. The coarse rinse sediments consisted primarily of small particles of fuel.

2.3 CHEMISTRY

Chemical analyses of solution samples were of two types: analyses of uranium and radionuclides originating from the fuel specimens ("radio-chemistry") and analyses of species contained in the starting J-13 well water ("solution chemistry").

2.3.1 Radiochemistry

A summary of radiochemistry methods is given in Table 2.5. Selenium-79 analyses were discontinued after Cycle 2 since attempts to measure it by liquid scintillation following separation in Cycles 1 and 2 failed.⁽²⁾ Selenium-79 analyses were replaced by ^{126}Sn analyses in Cycles 3, 4, and 5. The approximate detection limits for each radionuclide are compared in Table 2.5 to the activity that would result if 10^{-5} of the specimen inventory were dissolved in the 250 mL of test solution. The capability to measure better than 10^{-5} of inventory in solution is significant based on the Nuclear Regulatory Commission (NRC) stated⁽⁹⁾ 10^{-5} of 1000-yr inventory annual release limit.

2.3.2 Solution Chemistry

Solution chemistry measurements included pH, ICP for cations, IC for anions, and inorganic carbon. Bicarbonate (HCO_3^-) concentration in $\mu\text{g/mL}$ was calculated by multiplying the inorganic carbon results (also in $\mu\text{g/mL}$) by 5.0833 to correct for molecular weight. For certain analyses, organic carbon and total carbon were also reported. The detection limits of the solution

TABLE 2.4. Rinse Residue and Ceramography Specimen Weights (mg)

	Cycle 1		Cycle 2		Cycle 3 ^(a)		Cycle 4 ^(b)		Cycle 5 ^(b,c)	
	HBR	TP	HBR	TP	HBR	TP	HBR	TP	HBR	TP
Course sediment	158.8	12.3	Not weighed	Not weighed	123.2	106.9	179.7	60.4	Not weighed	Not weighed
Rinse filter	Not weighed	Not weighed	Not weighed	Not weighed	5.2	1.8	2.3	2.3	3.1	1.7
Ceramography	1003	524	465	519	284	--	--	--	--	--

(a) Fuel specimens were allowed to dry between Cycles 2 and 3 and weighed 81.4469 g (HBR) and 26.1327 g (TP).

(b) No ceramography samples were taken.

(c) Final dry fuel specimens weighed 80.5671 g (HBR) and 25.9272 g (TP).

TABLE 2.5. Summary of Radiochemistry Methods

Radionuclide	Method	Detection Limits		10 ⁻⁵ Inventory (pCi/mL) ^(b)
		(pCi/mL)	(ng/mL) ^(a)	
²⁴⁴ Cm	α-spectrometry	0.2	3 x 10 ⁻⁶	4100
²⁴¹ Am	α-spectrometry following separation	0.1	3 x 10 ⁻⁵	5700
²³⁹⁺²⁴⁰ Pu	α-spectrometry	0.2	0.003	2380
²³⁷ Np	α-spectrometry following separation	0.1	0.14	0.8
¹³⁷ Cs	γ-spectrometry	200	0.002	2.0 x 10 ⁵
¹²⁹ I	Neutron activation analysis	10 ⁻⁵	0.0001	0.08
¹²⁶ Sn	GeLi well γ-spectrom- etry following separation	0.2	0.02	2.2
⁹⁹ Tc	β-proportional counting following separation	10	0.6	34
⁹⁰ Sr	β-proportional counting following separation	20	0.0001	1.3 x 10 ⁵
⁷⁹ Se	Liquid scintillation counting following separation	20	0.3	1.2
⁶⁰ Co	γ-spectrometry	200	0.0002	(c)
¹⁴ C	Liquid scintillation counting following separation	20	0.004	2
U	Fluorescence	--	1	(3 ppm)

(a) Equivalent mass concentration for indicated isotope.

(b) Activities for 10⁻⁵ of H. B. Robinson test specimen inventories dissolved in 250 mL.

(b) ⁶⁰Co inventory is variable.

chemistry analyses were generally on the order of 0.1 $\mu\text{g/mL}$, which was adequate for following the concentrations of ionic species in J-13 well water in order to determine if these species were being precipitated during the tests. Another purpose for the solution chemistry data was to indicate any test contamination or sample contamination with nonradioactive species.

3.0 RESULTS AND DISCUSSION

3.1 GENERAL COMMENTS ON DATA PRESENTATION

A complete tabulation of radiochemical results reported in pCi activity units (μg units for uranium) is contained in Appendix A along with activity/concentration conversion formulas and radiochemistry error estimates. A complete tabulation of non-nuclide solution chemistry data is contained in Appendix B. Discussion of the radiochemical results is organized by nuclide, starting with the actinides (uranium, $^{239+240}\text{Pu}$, ^{241}Am , ^{244}Cm , and ^{237}Np), followed by fission product (^{137}Cs , ^{90}Sr , ^{99}Tc , and ^{129}I) and activation product (^{14}C and ^{60}Co) nuclides. A discussion of solution chemistry and secondary-phase examination results follows the radiochemistry discussions.

3.1.1 Plotted Data

Radiochemical data from periodic and final solution samples are plotted as a function of time in composite plots showing data for the five sequential test cycles in adjacent boxes along the x-axis. Data from the HBR test are plotted as open symbols, and data from the TP test are plotted as closed symbols. Round symbols are used for unfiltered data, square symbols for 0.4- μm filtered data, and triangular symbols are used for 18- $\text{\AA}$ filtered data. A downward-pointing arrow attached to plotted data points indicates data reported as "less than" values. The $^{239+240}\text{Pu}$, ^{241}Am , and ^{244}Cm data are plotted as semi-log plots because these data varied over several orders of magnitude as a function of time and/or filtered fraction. Linear plotting is used for uranium, ^{237}Np , and fission product (^{137}Cs , ^{90}Sr , ^{99}Tc , and ^{129}I) data because these data did not vary over several orders of magnitude. The actinide data are plotted as activity (concentration for uranium) versus time, since these nuclides tended to reach steady-state concentrations in solution and it was not possible to estimate the inventory fractions dissolved based on the quantities measured in solution. The activity levels that would result if 10^{-5} of the HBR or TP specimen actinide inventories were in solution are indicated on each plot for comparison purposes. The 10^{-5} of inventory in solution was chosen as a convenient reference level for comparison of the actinide data and is not intended to be interpreted relative to the NRC annual release limit of 10^{-5} of the 1000-yr inventory for specific nuclides.

The fission product data are plotted as a specimen inventory fraction measured in solution versus time to give these plots a stronger basis for comparison since these nuclides were relatively soluble and appeared to remain mostly in solution. Each plotted fission product data point represents the inventory fraction determined to be in the 250 mL of test solution at the sampling date plus the sum of inventory fractions removed in previous samples during the test cycle. Inventory fractions removed in previous samples were estimated in cases where the fission product activity was not measured in all samples.

3.1.2 "Quantities Measured" Tables

Tables of the quantities of nuclides measured in the various types of liquid samples were compiled for each nuclide. The "periodic samples" values given in these tables are the sum of the unfiltered sample activities (concentrations for U) times sample volumes for each periodic solution sample (excluding the final solution sample) in which detectable activities were measured. Since not all nuclides were measured in every sample, the periodic sample values may be in some cases less than the actual total quantity removed in periodic samples. In certain cases, such as ^{129}I in Cycles 1 and 2, where only a single periodic sample was analyzed, that value was assumed as an estimate of the ^{129}I in all periodic samples and is indicated by a footnote. The "final solution" value is the quantity of the nuclide determined to be in the 250 mL of test solution at the end of a test cycle. The "final solution" values are based on 0.4- μm filtered data. For ^{99}Tc , ^{90}Sr , ^{129}I , and ^{14}C the "periodic solution" and "final solution" values are based on unfiltered data. Concentration^(a) of the element in the final solution sample is given in parentheses below the "final solution" value in the indicated units. The "rod samples" value is the total quantity of the nuclide measured on rod samples periodically removed during Cycles 1, 2, and 3 of the test.

The "rinse" value is the quantity of the nuclide determined to be dissolved in the 600-mL rinse solutions when sampled the day after cycle

(a) Elemental concentrations were calculated from nuclide activities using Equation (A.1) and isotope/element mass ratios given in Table A.1 of Appendix A.

termination. Rinse solution samples were 0.4- μ m filtered prior to analysis. The "acid strip" value is the quantity of the nuclide determined to be dissolved in the 300 mL of 8 M HNO_3 used to strip the internal vessel surfaces, specimen basket, and remaining fused silica rods at cycle termination. The "cycle total" values are the sum of the periodic samples, final solution, rinse and acid strip values. The " $\div 10^{-5}$ Inv." value is the cycle total value divided by 10^{-5} of the inventory of that nuclide calculated to be present in the initial spent fuel test specimen. The "% in solution" value is the sum of the periodic samples plus the final solution values divided by the cycle total value times 100. "Less than" symbols indicate either: 1) the value was reported as a "less than" value for the particular sample, or 2) the value is a sum in which greater than 5% is based on "less than" values.

It should be noted that the tabulated "Cycle Total" and " $\div 10^{-5}$ Inv." values for the actinides cannot be directly equated with the actual quantity of fuel matrix dissolution that occurred because the quantities of actinides presumed to have precipitated as secondary phases were not quantitatively measured. Much of the secondary phase inventory formed during each test cycle was likely removed by the rinsing procedure. Partial dissolution from secondary phases may have contributed along with dissolution of fuel fines to the nuclide quantities measured in the rinse solutions. Nuclide inventories contained in secondary phases were not determined. Some previously undissolved fuel fines may also have been dissolved in the acid strip solution, adding further uncertainty as to the meaning of the calculated "Cycle Total" values. For the soluble fission product nuclides (^{137}Cs , ^{90}Sr , ^{99}Tc , and ^{129}I), the Cycle Total and $\div 10^{-5}$ Inv. values tabulated are probably fair estimates for the release quantities in each cycle and provide upper limits for the amounts of fuel matrix dissolution in later test cycles.

3.2 ACTINIDES

Actinides account for the majority of the radioactivity in spent fuel during the postcontainment period. Actinide concentrations measured in the periodic solution samples tended to reach maximum steady-state levels early during test cycles, suggesting that actinide release will be solubility-controlled. The steady-state concentrations (filtered and unfiltered) reached

by the actinides were orders of magnitude below those necessary to meet the NRC 10 CFR 60.113⁽⁹⁾ release limits (less than 10^{-5} per year of 1000-yr inventories) for any reasonable water flux predicted for the Yucca Mountain repository site. Results for the actinides are discussed in the following subsections.

3.2.1 Uranium

Uranium concentration measured in 0.4- μm filtered periodic and final solution samples are plotted in Figure 3.1. Concentrations measured in unfiltered and 18- $\text{\AA}$ filtered aliquots were quite similar to those shown for the 0.4- μm filtered aliquots. This may be an artifact of the laser fluorimetry technique used, that measures fluorescence of a uranium species formed in solution after the addition of a complexing reagent and is not sensitive to uncomplexed uranium contained in suspended particles. However, after filtration, sample aliquots were acidified to approximately 1% HNO_3 prior to analysis to prevent uranium plate-out. Peak concentrations were observed early in Cycle 1 and then decreased during the cycle. The apparent initial supersaturation in Cycle 1 is attributed to dissolution from an oxidized UO_{2+x} surface film on the fuel and to slow kinetics for the nucleation and growth of more stable secondary uranium phases. The more rapid decrease in uranium concentration during Cycle 1 observed with the HBR fuel is attributed to depletion of a less extensive oxidized surface phase in this test relative to the TP Cycle 1 test. The period between initial rod sectioning and testing was about five years for the TP fuel versus a few months for the HBR fuel. In the later test cycles, uranium concentration tended toward a 1 to 2 $\mu\text{g/mL}$ (ppm) range in both tests. Slightly higher concentrations in the HBR test compared to the TP test in later test cycles may be due to effects of the greater fuel-to-water ratio in the HBR test on concentrations achieved in the steady-state process of dissolution and secondary-phase formation.

Quantities of uranium measured in the various sample types are given in Table 3.1. With the exception of Cycle 1 of the HBR test, most of the uranium was measured in solution. The greater quantities of uranium in the rinse and acid strip samples in Cycle 1 of the HBR test correlate with the larger amount of uranium precipitation indicated in Figure 3.1 for this test cycle. The total quantity of uranium measured over the five cycles was 10.06×10^{-5} of

FIGURE 3.1. Uranium Concentration Measured in 0.4- μ m Filtered Samples

TABLE 3.1. Quantities of Uranium Measured (μg)

	Cycle 1		Cycle 2		Cycle 3		Cycle 4		Cycle 5	
	HBR	TP	HBR	TP	HBR	TP	HBR	TP	HBR	TP
Periodic samples	253	351	142	135	183	73	89	58	31	14
Final solution	300	1000	500	600	350	300	300	275	425	218
[U (ppm)]	(1.2)	(4.0)	(2.0)	(2.4)	(1.4)	(1.2)	(1.2)	(1.1)	(1.7)	(0.9)
Rod samples	36	15	18	3	5	3	--	--	--	--
Rinse	660	366	102	39	66	25	43	13	114	22
Acid strip	2700	960	300	156	93	159	75	29	183	42
Cycle total	3949	2692	1062	933	697	560	507	375	753	296
$\div 10^{-5}$ Inv.	5.66	11.67	1.54	4.13	1.02	2.52	0.74	1.70	1.10	1.35
% in Solution	14.00	50.19	60.45	78.78	76.47	66.61	76.23	88.80	60.60	78.38

Summary of Cycles

Σ Cycle totals	3949	2692	5011	3625	5708	4185	6215	4560	6968	4856
$\div 10^{-5}$ Inv.	5.66	11.67	7.20	15.80	8.22	18.32	8.96	20.02	10.06	21.37

inventory (6968 μg) for the HBR test and 21.37×10^{-5} of inventory (4856 μg) for the TP test. The greater uranium fractional release for the TP test results from the lower fuel-to-water ratio in this test.

3.2.2 Plutonium

The $^{239+240}\text{Pu}$ activities measured in unfiltered, 0.4- μm filtered, and 18- $\text{\AA}$ filtered samples are plotted in Figure 3.2. The peak activities measured in the first unfiltered Cycle 1 samples may be due to initially dispersed fuel fines or formation of floccules that later settle from the solution. The dispersed fuel fines hypothesis is supported by the nearly congruent actinide quantities (U, Pu, Am, and Cm) measured in the initial unfiltered Cycle 1 samples. Silica floccules were observed⁽²⁾ during SEM examination of filters used to filter solution samples, and actinide adsorption by such floccules may have occurred at levels below the detection threshold for EDS analysis in the SEM.

The high $^{239+240}\text{Pu}$ data measured in the 62-day Cycle 2 sample from the HBR test is probably in error. The ^{244}Cm counted on the same source disks showed similarly high activities, while separated ^{241}Am counted on separate source disks did not show high activity. The $^{239+240}\text{Pu}$, ^{241}Am and ^{244}Cm activities measured in the 148-day Cycle 4 unfiltered sample aliquot from the HBR test were also high "flyer" data points. The remaining data points clearly indicate lower $^{239+240}\text{Pu}$ activities in the HBR test than in the TP test. The activity measured (unfiltered and 0.4- μm filtered) at the end of Cycle 5 of the HBR test was about 18 pCi/mL versus about 110 pCi/mL in the TP test. An explanation for this difference in Pu activities has not been found. The relatively small effects of filtration (generally less than a factor of 4) suggest that the formation of stable Pu-colloids was limited and that a substantial portion of the Pu measured in solution samples was in true solution.

The quantities of $^{239+240}\text{Pu}$ measured in the various liquid sample types are given in Table 3.2. The solubilities measured in final solution samples were on the order of 1 ppb ($4 \times 10^{-9} \text{ M}$) for Pu (all isotopes combined) versus on the order of 1 ppm ($4 \times 10^{-6} \text{ M}$) for uranium. The greatest portions of Pu were measured in the acid strip samples. The five-cycle total inventory

FIGURE 3.2. Activities of $^{239} + ^{240}\text{Pu}$ Measured in Solution Samples

TABLE 3.2. $^{239+240}\text{Pu}$ Quantities Measured (nCi)

	Cycle 1		Cycle 2		Cycle 3		Cycle 4		Cycle 5	
	HBR	TP	HBR	TP	HBR	TP	HBR	TP	HBR	TP
Periodic samples	41	62.7	23.8	13.4	13.6	15.2	10.3	15.6	3.5	4.0
Final solution	28	114	8.3	45.0	23.5	49.3	4.8	38.3	4.7	27.5
[Pu (ppb)]	(1.2)	(4.9)	(0.36)	(1.9)	(1.0)	(2.1)	(0.20)	(1.6)	(0.20)	(1.2)
Rod samples	59	21.6	173	5.4	7.4	4.8	--	--	--	--
Rinse	254	62.7	253	26.5	58.6	20.3	40.5	15.4	73.2	15.7
Acid strip	4054	1140	339	204	165	193	173	42.7	301	50.1
Cycle total	4436	1401	779	294	268	283	229	112	382	97.3
$\div 10^{-5}$ Inv.	7.18	7.31	1.28	1.57	0.44	1.54	0.38	0.61	0.64	0.54
% in Solution	1.56	12.61	4.12	19.84	13.84	22.82	6.61	48.12	2.14	32.37

Summary of Cycles

Σ Cycle totals	4436	1401	5215	1695	5483	1978	5712	2090	6094	2187
$\div 10^{-5}$ Inv.	7.18	7.31	8.46	8.88	8.90	10.42	9.28	11.03	9.92	11.57

fraction of $^{239+240}\text{Pu}$ measured in the HBR test (9.92×10^{-5}) is about equal to that measured for uranium. The five-cycle total $^{239+240}\text{Pu}$ inventory fraction measured in the TP test (11.57×10^{-5}) is comparable to that measured in the HBR test, but less than that measured for uranium in the TP test. The total measured inventory fraction results for actinides other than uranium suggest congruent release of these actinides. However, this result is heavily weighted by the Cycle 1 results where relatively large quantities of nuclides measured in the acid strip samples may have partially originated from previously undissolved fuel fines. Another limitation in using the cycle total inventory fractions measured for different actinides as evidence for incongruent or congruent dissolution is that the quantities of actinides removed as secondary phases in the rinse solutions were not accurately accounted for.

The $^{239+240}\text{Pu}$ isotopes account for about 45% of the activity present in 1000-year old spent fuel.⁽¹⁰⁾ The half-lives of ^{239}Pu and ^{240}Pu are 24,130 and 6570 years, respectively. Assuming a water flux through the repository horizon of 20 L per year per waste package containing 3140 kg of spent fuel^(11,12) becomes saturated with 100 pCi/mL of $^{239+240}\text{Pu}$, about 1×10^{-9} of the $^{239+240}\text{Pu}$ inventory in the waste packages at 1000 years would be transported per year. This value is very much lower than the NRC release requirement of less than 1×10^{-5} of the 1000-year inventory per year for individual nuclides.

3.2.3 Americium

The ^{241}Am activities measured^(a) in unfiltered, 0.4- μm filtered, and 18-Å filtered samples are plotted in Figure 3.3. A prominent feature of this data is its range, which is greater than three orders of magnitude. The high activities for the initial Cycle 1 unfiltered samples are similar to those observed in the $^{239+240}\text{Pu}$ data, and probably result from initially dispersed

(a) Cycle 1 ^{241}Am activities up to (but not including) the final solution samples were calculated from $^{241}\text{Am} + ^{238}\text{Pu}$ and $^{239+240}\text{Pu}$ data using $^{238}\text{Pu}/^{239+240}\text{Pu}$ ratios radiochemically measured on single samples from each test. Other ^{241}Am activities were counted on sources prepared following Am separation.

FIGURE 3.3. Activities of ²⁴¹Am Measured in Solution Samples

fuel fines or actinide containing floccules. Steady-state activities measured on sample aliquots 0.4- μm filtered activities ranged from about 1000 pCi/mL in Cycle 1 of the TP test down to less than 10 pCi/mL in Cycle 4 of both tests and in Cycle 5 of the HBR test. Excluding the initial Cycle 1 samples, the unfiltered data cover a similar range. A curious feature of the data is the order of magnitude activity increases that occurred between Cycles 2 and 3 of the HBR test, possibly related to the 14-day period of air exposure between Cycles 2 and 3.

The apparent effects of filtration, especially notable in the activity reductions following 18-Å filtration, suggest that most of the ^{241}Am activity in solution is associated with suspended particles or colloids. However, the actual concentrations involved (100 pCi/mL of ^{241}Am corresponds to $1.5 \times 10^{-10} \text{ M}$) are very low, so the possibility exists that the activity reductions associated with 18-Å filtration could be artifacts of adsorption of small quantities of Am by the filters or other surfaces. Activity associated with particles or floccules that are retained by the 0.4- μm filters would likely settle or be filtered by the rock, while activity that passes the 0.4- μm filters may stay in suspension and move with the water. Based on the foregoing considerations, use of the 0.4- μm filtered data would seem most appropriate for transport and release estimation. Although the current data do not provide a well-defined, stable 0.4- μm filtered ^{241}Am activity value, the range of the log scale data is centered around a value of about 100 pCi/mL. A relatively stable value for 0.4- μm filtered ^{241}Am activity of 100 pCi/mL was observed at 25°C in Cycles 2 and 3 of the Series 3 tests.⁽³⁾

The quantities of ^{241}Am measured in the various sample types are given in Table 3.3. The total ^{241}Am inventory fractions measured over the five cycles of both tests are similar but are heavily weighted by the ^{241}Am quantities measured in the Cycle 1 acid strip solutions. These inventory fractions are also similar to those measured for $^{239+240}\text{Pu}$ and ^{244}Cm .

TABLE 3.3. ^{241}Am Quantities Measured (nCi)

	Cycle 1		Cycle 2		Cycle 3		Cycle 4		Cycle 5	
	HBR	TP	HBR	TP	HBR	TP	HBR	TP	HBR	TP
Periodic samples	91.9	138	3.6	5.7	44.6	6.9	29.4	5.5	4.9	2.1
Final solution	71.4	243	6.2	21.1	63.7	22.2	2.4	16.2	2.0	18.2
[Am (pg/mL)]	(105)	(348)	(9.1)	(30)	(94)	(32)	(3.5)	(23)	(3.0)	(26)
Rod samples	132	42.1	59.4	38.4	22.9	10.7	--	--	--	--
Rinse	532	111	139	50.5	118	42.2	106	39.5	175	39.5
Acid strip	9595	2180	773	400	396	392	446	103	774	134
Cycle total	10422	2714	981	515	645	474	584	164	956	194
$\div 10^{-5}$ Inv.	8.04	6.88	0.77	1.33	0.51	1.25	0.46	0.44	0.76	0.52
% in Solution	1.57	14.04	1.00	5.15	16.79	6.14	5.45	13.22	0.72	10.47

Summary of Cycles

Σ Cycle totals	10422	2714	11403	3229	12048	3703	12632	3867	13588	4061
$\div 10^{-5}$ Inv.	8.04	6.88	8.81	8.21	9.32	9.46	9.78	9.90	10.54	10.42

Americium-241 accounts for about 51% of the total activity in spent fuel at 1000 years, and Pu and Am isotopes combined^(a) account for about 98% of the 1000-yr activity. With a 432-yr half-life, ²⁴¹Am decays to a much lower portion of the total activity after a few thousand years. Assuming that a ²⁴¹Am activity of 100 pCi/mL is transported in water with a flow rate of 20 L per year per waste package containing 3140 kg of fuel, the annual release would correspond to about 8×10^{-10} of the 1000-yr ²⁴¹Am inventory. As with ²³⁹⁺²⁴⁰Pu, this value is much less than the NRC annual release limit of 1×10^{-5} of the 1000-yr inventory. The assumptions used in the preceding release estimates for Pu and Am isotopes are conservative. Activities for ²³⁹⁺²⁴⁰Pu and ²⁴¹Am are likely to be less than 100 pCi/mL, considering that these activities measured in 0.4- μ m filtered samples at 85°C in the Series 3 tests^(3,12,13) were on the order of 1 pCi/mL or less. The 20 L per year per waste package is probably a conservative estimate for the water flow rate. Release estimates should be further lowered by consideration of a realistic time distribution for waste package failures, probabilities for saturation of failed waste packages with water, and retardation of actinides as a result of sorption by the rock.

3.2.4 Curium

The activities of ²⁴⁴Cm measured in unfiltered, 0.4- μ m filtered, and 18-Å filtered samples are plotted in Figure 3.4. The ²⁴⁴Cm data are very similar to the ²⁴¹Am data. The data cover a range of nearly four orders of magnitude and show significant filtration effects. As with ²⁴¹Am, a substantial proportional activity reduction with the 18-Å filtration suggests that the majority of the ²⁴⁴Cm activity measured in unfiltered and 0.4- μ m filtered samples is associated with colloidal phases. However, the ²⁴⁴Cm concentrations involved (100 pCi/mL corresponds to a Cm concentration of about 5×10^{-12} M) are very low, and as with ²⁴¹Am, the activity reductions associated with 18-Å filtration could also be artifacts resulting from adsorption

(a) Includes the activity of ²³⁹Np, which is a short-lived daughter product of ²⁴³Am decay. The ²³⁹Np accounts for about 0.9% of the 1000-yr activity. All 1000-yr radionuclide inventory data cited in this report are from ORNL/TM-7431 (Ref. 10).

FIGURE 3.4. Activities of ^{244}Cm Measured in Solution Samples

by the 18 Å membrane filters or other surfaces. Most of the unfiltered and 0.4-μm filtered activities were in the 10 pCi/mL to 100 pCi/mL range during Cycles 2, 4, and 5. The quantities of ^{244}Cm activity measured in the different sample types are given in Table 3.4.

Curium-244 has a relatively short half-life of 18.1 yr and will decay out during the minimum 300-yr containment period. Other curium isotopes (primarily 242, 245, and 246) account for about 0.013% of the 1000-yr activity of spent fuel. If the most abundant of these isotopes, ^{245}Cm , saturated at 0.2 pCi/mL (corresponds to the same Cm concentration as 100 pCi/mL ^{244}Cm in the current tests) in a water flow of 20 L per year per waste package (3140 kg of fuel), about 1×10^{-8} of 1000-yr ^{245}Cm inventory would be transported per year.

3.2.5 Neptunium

Activities of ^{237}Np measured in unfiltered, 0.4-μm filtered, and 18-Å filtered samples are plotted in Figure 3.5. The measured activities were generally less than 1 pCi/mL and showed a relatively large degree of scatter because these activities were approaching the detection limits. With the exception of the initial samples from Cycle 1 of the TP test, most of the ^{237}Np activities measured fell in a narrow range between 0.1 and 0.8 pCi/mL. The data suggest that ^{237}Np activities approached a steady-state level of about 0.4 pCi/mL, corresponding to a Np concentration of about 2.4×10^{-9} M. No significant filtration effects on ^{237}Np activities were observed.

Quantities of ^{237}Np measured in the various sample types are given in Table 3.5. Considering that much of the data included in Cycle Totals were reported as "less than", the inventory fractions given for ^{237}Np compare reasonably well with those measured for the other actinides, suggesting that ^{237}Np may be released congruently with other actinides as the fuel dissolves.

Assuming a 20 L per year per waste package water flow becomes saturated with ^{237}Np at an activity of 0.4 pCi/mL, $\sim 3 \times 10^{-9}$ of the 1000-yr ^{237}Np inventory would be transported per year. The data thus indicate that the NRC release limit should also be met for ^{237}Np with a large factor for conservatism.

TABLE 3.4. ^{244}Cm Quantities Measured (nCi)

	Cycle 1		Cycle 2		Cycle 3		Cycle 4		Cycle 5	
	HBR	TP	HBR	TP	HBR	TP	HBR	TP	HBR	TP
Periodic samples	125	138	11.8	5.4	52.8	6.6	19.8	4.3	3.9	1.6
Final solution	102	250	7.0	20.3	84.5	18.5	2.9	15.7	2.6	8.7
[Cm (pg/mL)]	(5.3)	(12.9)	(0.24)	(1.05)	(4.4)	(0.95)	(0.15)	(0.81)	(0.13)	(0.45)
Rod samples	145	38.3	388	34.4	20	9.2	--	--	--	--
Rinse	565	104	678	36.8	106	26.5	84.6	24.3	144	24.3
Acid strip	8973	1610	732	285	349	297	374	71.4	595	89.2
Cycle total	9910	2140	1817	382	612	358	481	116	745	124
$\div 10^{-5}$ Inv.	8.64	7.79	1.61	1.42	0.55	1.36	0.43	0.44	0.67	0.47
% in Solution	2.29	18.13	1.04	6.73	22.42	7.02	4.72	17.29	0.87	8.31

Summary of Cycles

Σ Cycle totals	9910	2140	1727	2522	12339	2880	12820	2996	13565	3120
$\div 10^{-5}$ Inv.	8.64	7.79	10.25	9.21	10.80	10.57	11.23	11.01	11.90	11.48

FIGURE 3.5. Activities of ²³⁷Np Measured in Solution Samples

TABLE 3.5. ^{237}Np Quantities Measured (pCi)

	Cycle 1		Cycle 2		Cycle 3		Cycle 4		Cycle 5	
	HBR	TP	HBR	TP	HBR	TP	HBR	TP	HBR	TP
Periodic samples	<25	55	20	14	40.5	22.5	29.7	17.6	20.7	10.8
Final solution	<112	112	90	90	90	113	101	135	124	67.6
[Np (ppb)]	(<0.64)	(0.64)	(0.5)	(0.5)	(0.5)	(0.64)	(0.58)	(0.77)	(0.7)	(0.38)
Rod samples	<10	NM	2	2	<2.4	<1.5	--	--	--	--
Rinse	<270	<135	<135	<135	<135	<135	<81	<135	342	54.1
Acid strip	946	135	68	<68	<68	81.1	<40	405	108	27
Cycle total	<1363	<437	<315	<309	<336	<353	252	<693	595	160
$\div 10^{-5}$ Inv.	<6.8	<7.2	<1.6	<5.3	<1.7	<6.2	<1.3	<12.2	3.1	2.8
% in Solution	--	--	--	--	--	--	--	--	24.3	49.1
<u>Summary of Cycles</u>										
Σ Cycle totals	<1363	<437	<1678	<746	<2014	<1099	<2266	<1792	<2861	<1952
$\div 10^{-5}$ Inv.	<6.8	<7.2	<8.4	<12.5	<10.1	<18.7	<11.4	<30.9	<14.5	<33.7

 NM = Not measured.

3.2.6 Comparison with EQ3/6 Predictions

An important purpose of the NNWSI spent fuel dissolution tests is to provide validation data for computer codes being developed to simulate dissolution of spent fuel under Yucca Mountain site-specific conditions. Approximate actinide concentrations measured in the Series 2 bare fuel tests are compared in Table 3.6 to values calculated at 25°C by LLNL using Version 3270 of the EQ3/6 geochemical code and Version 3245R54 of the supporting thermodynamic database.⁽¹²⁾ The EQ3/6 values were calculated assuming atmospheric CO₂ gas fugacity and two different O₂ gas fugacities of 10^{-0.7} (atmospheric) and 10^{-12.0} bars with solubility control by the indicated phases.

TABLE 3.6. Comparison of Measured Actinide Concentrations to Those Calculated Using EQ3/6 (log M)

Actinide	Measured ^(a)		EQ3/6 ^(b)		Phase ^(c)
	0.4 μm	18 $\text{\AA}$	-0.7	-12.0	
U	-5.2	--	-7.2/-7.0 -7.0/-6.9 -6.9/-4.3 -4.3 -4.2	-7.1/-6.9 -6.9/-6.8 -6.8/-4.2 -4.2 -4.1	H H+S S S+Sch Sch
Np	-8.6	--	-6.2	-9.0	NpO ₂
Pu(HBR)	-8.4	--	-12.4	-13.8	PuO ₂
(TP)	-9.1	--	-5.7	-4.2	Pu(OH) ₄
Am	-9.8	-11.3	-8.4 -8.3	-8.4 -8.3	Am(OH) ₃ Am(OH)CO ₃
Cm	-11.6	-13.0	Cm not in thermodynamic database.		

- (a) Approximate steady-state concentrations (log M) for 0.4 μm and 18 $\text{\AA}$ filtered samples.
- (b) At oxygen fugacities $\log f(\text{O}_2) = -0.7$ (atmospheric) and $\log f(\text{O}_2) = -12.0$ where $f(\text{O}_2)$ is in bars.⁽¹¹⁾ Two values (i.e., -7.2/-7.0) indicate a concentration range.
- (c) Solubility-controlling phases (H = haiweeite, S = soddyite, Sch = schoepite), all phases are crystalline except Pu(OH)₄ which is amorphous.

The approximate steady-state uranium concentrations ($1.5 \mu\text{g/mL}$, $\log M = -5.2$) falls in the range calculated for precipitation of soddyite. No soddyite lines were found in an XRD pattern from a filter containing specimen rinse residues. However, an indication for haiweeite (a Ca-U-silicate phase) formation was provided by a single strong line in the XRD pattern from this rinse filter sample and is discussed in Section 3.7. Neptunium concentration is controlled by equilibrium with NpO_2 in the EQ3/6 simulations, and the calculated Np concentration is highly dependent on solution Eh and pH. Changing the O_2 fugacity [$f(\text{O}_2)$] from $10^{-0.7}$ bars to 10^{-12} bars resulted in improved agreement between the measured and calculated Np concentrations. EQ3/6 results at $f(\text{O}_2) = 10^{-12}$ bars were originally calculated because this $f(\text{O}_2)$ value resulted in good agreement with the Np results measured at 25°C in the Series 3 tests. Although the solution was in contact with air, redox equilibria probably were not well established among the various phases during these tests.

Approximate steady-state Pu concentrations measured in the HBR and TP tests ($10^{-8.4}$ and $10^{-9.1}$ M respectively) are much greater than the EQ3/6 values calculated for solubility control by crystalline PuO_2 , and much lower than the concentrations calculated for solubility control by amorphous $\text{Pu}(\text{OH})_4$. However, the measured Pu concentrations are in fair agreement with those reported by Rai and Ryan,⁽¹⁴⁾ who measured the solubility of PuO_2 and hydrous $\text{PuO}_2 \cdot x\text{H}_2\text{O}$ in water at 25°C over time periods up to 1300 days. At a pH of 8, which was the extrapolated lower limit of their data, and the approximate pH of the HBR and TP tests, they reported that Pu concentration ranged from $10^{-7.4}$ M, where amorphous $\text{PuO}_2 \cdot x\text{H}_2\text{O}$ was thought to control concentration, down to about 10^{-9} M, where aging of the amorphous material produced a more (but incompletely) crystalline PuO_2 that was thought to control concentration.

The measured Am concentrations were lower than predicted in the EQ3/6 simulations based on precipitation of $\text{Am}(\text{OH})\text{CO}_3$ at 25°C or $\text{Am}(\text{OH})_3$ at 90°C . A possible explanation for this difference is that Am, and likely Cm, may have precipitated from solution with the lanthanides. The chemistry of trivalent Am and Cm can be expected to be very similar to that of light lanthanide fission product elements, which are present in spent fuel at much greater concentrations than are Am and Cm and have similar ionic radii in the

trivalent state. The transuranic actinides may also be precipitating at low concentrations in the uranium-bearing precipitates. Sorption of actinides on colloids or other surfaces such as the fuel or test hardware may have also controlled some aspects of solution concentration not considered in the geochemical simulations.

3.3 FISSION PRODUCTS

Dissolution behavior of soluble fission product radionuclides from spent fuel differs from that of actinide radionuclides in two important ways. First, some important fission product radionuclides tend to partially segregate from the UO_2 fuel matrix phase during irradiation, and they are not necessarily congruently released with the actinides as the matrix phase dissolves. The second difference is that many of the important fission product nuclides tend to be relatively soluble, and their release probably will not be limited by achieving a maximum solubility limited concentration in a limited amount of water flow.

Mobile fission products such as cesium and iodine concentrate in the fuel-cladding gap (and in cracks and open porosity) from which they are rapidly released with initial water contact. The quantities of various nuclides that are rapidly and preferentially released with initial water contact have been referred to as "gap inventory." A continuous preferential release of fission product elements, Cs, Tc, I, and possibly Sr, appears to occur for an indefinite period after the gap inventory pulse is released. A primary source for the continuous preferential release is thought to be preferential release from grain boundaries⁽¹⁵⁾ where mobile fission products are thought to concentrate during irradiation. This type of release is referred to as "preferential" release rather than "grain boundary" release in this report since the actual locations and state of fission product concentrations in light water reactor (LWR) fuel are not well characterized. Quantitative measurement of the preferential component of continuously released soluble nuclides has been limited to date because of difficulties in determining release contributions for these nuclides originating from simultaneous congruent dissolution of the fuel matrix phase. Eventually, the continuous preferential release component should decrease and may ultimately disappear as

the inventory of radionuclides concentrated at locations such as exposed grain boundaries is depleted. At such time, soluble nuclide release would result primarily from congruent dissolution of the fuel matrix phase and be indicative of the matrix dissolution rate.

Four soluble fission products measured at detectable levels in the Series 2 tests were ^{137}Cs , ^{90}Sr , ^{99}Tc , and ^{129}I . There was no evidence that concentrations of these fission products were limited by secondary phase formation during the Series 2 tests.^(a) Measurements were also made for ^{126}Sn and ^{79}Se , but activities of these two nuclides were either below or near detection limits.

3.3.1 Cesium

The activities of ^{134}Cs (2.06-yr half-life) plus ^{137}Cs (30.2-yr half-life) plus $^{137\text{m}}\text{Ba}$ (short-lived ^{137}Cs daughter) account for about 38% of the total activity in the fuel tested. The only significant long-lived Cs isotope, ^{135}Cs (2,300,000-yr half-life), has a low inventory of ~350 Ci/1000 MTHM, which is equivalent to about 0.02% of the total activity in 1000-yr-old spent fuel. The EPA 10,000-yr cumulative release limit given in Table 1 of 40 CFR 191⁽¹⁶⁾ for ^{135}Cs or ^{137}Cs is 1,000 Ci/1,000 MTHM.

The ^{137}Cs inventory fraction measured in solution versus time is plotted for the five test cycles in Figure 3.6. A gap inventory release of about 0.7% in the HBR test and about 0.23% in the TP test occurred at the beginning of Cycle 1. Each plotted data point includes the inventory fraction measured in solution on the sampling day plus the inventory fractions removed in prior samples during the test cycle. With the exception of a few data points, ^{137}Cs inventory fraction measured in solution during Cycle 1 generally increased with time after the initial gap inventory release.

The ratio of the ^{137}Cs inventory fraction to that of uranium measured in the first solution sample from each cycle is given in Table 3.7. The average of this ratio for Cycles 3, 4, and 5 is about 2.5. Considering that cesium is

(a) Solubility limits may have been reached for Sr, and possibly Cs, in the 85°C Series 3 tests.⁽³⁾

FIGURE 3.6. Inventory Fraction of ^{137}Cs Measured in Solution

much more soluble than uranium, and assuming that matrix dissolution and uranium-bearing secondary phase formation occur continuously, these ratios may represent a maximum value for the degree of preferential cesium dissolution. In Cycle 2, after a fast fractional release of $\sim 7.5 \times 10^{-5}$ in the HBR test and $\sim 2.5 \times 10^{-5}$ in the TP test, both tests exhibited continuous ^{137}Cs release at a rate of about 6×10^{-7} of specimen inventory per day. During Cycles 3 and 4 the average release rate between the first and final solution samples was about 3×10^{-7} of inventory per day for both tests. During Cycle 5 the average continuous release rate was $\sim 2.6 \times 10^{-7}$ per day for both tests. These data suggest that the matrix dissolution rate was between about 1×10^{-7} and 3×10^{-7} per day during Cycles 3, 4, and 5.

The quantities of ^{137}Cs measured in the different sample types are given in Table 3.8. These data indicate that most of the ^{137}Cs was measured in solution. The total ^{137}Cs inventory fractions measured in all sample types, 0.818% for the HBR test and 0.347% for the TP test, which are heavily weighted by the Cycle 1 gap release, are greater than those measured for any other nuclide.

3.3.2 Strontium

Strontium-90 is the only significant radioactive Sr isotope in spent fuel. ^{90}Sr beta decays (28.6-yr half-life) to ^{90}Y , which then beta decays (64-h half-life) to stable ^{90}Zr . ^{90}Sr plus ^{90}Y account for about 28% of total

TABLE 3.7. ^{137}Cs to Uranium Fractional Inventory Ratios in First Solution Samples^(a)

Test	Cycle 1 ^(b)	Cycle 2	Cycle 3	Cycle 4	Cycle 5
HBR	489	9.77	2.29	2.93	4.53
TP	50	2.26	1.49	1.90	2.08

(a) ^{137}Cs unfiltered, uranium 0.4- μm filtered.

(b) Uranium unfiltered.

TABLE 3.8. ^{137}Cs Quantities Measured (μCi)

	Cycle 1		Cycle 2		Cycle 3		Cycle 4		Cycle 5	
	HBR	TP	HBR	TP	HBR	TP	HBR	TP	HBR	TP
Periodic samples	12600	1010	151	30	53.7	11.9	49.2	16.4	18.8	3.04
Final solution	28200	3080	840	204	377	111	350	122	200	55.1
[Cs(ppb)]	(3277)	(357)	(98)	(24)	(44)	(13)	(41)	(14)	(23)	(6.4)
Rod samples	28	2.6	2.0	0.3	0.5	0.4	--	--	--	--
Rinse	1560	786	50.5	14.2	40.8	9.38	30.0	10.1	29.2	5.81
Acid strip	612	138	24.9	13.4	17.4	12.1	13.2	3.74	18.2	3.08
Cycle total	43000	5017	1070	262	489	145	442	152	266	67
$\div 10^{-5}$ Inv.	776	308	19.6	16.1	9.0	9.1	8.2	9.6	4.95	4.24
% in Solution	94.88	81.52	92.80	89.31	88.01	84.86	90.23	90.91	82.19	86.74

Summary of Cycles

Σ Cycle totals	43000	5017	44070	5279	44559	5424	45001	5576	45267	5643
$\div 10^{-5}$ Inv.	776	308	796	324	805	333	813	343	818	347

activity in the spent fuel specimens tested, and together with ^{137}Cs and $^{137\text{m}}\text{Ba}$ account for a substantial portion of the decay heat during the repository thermal period. However, ^{90}Sr and ^{137}Cs , along with their short-lived daughter products ^{90}Y and $^{137\text{m}}\text{Ba}$, will have essentially decayed out before the end of the minimum 300-yr NRC required containment period.

Regular analysis for ^{90}Sr did not begin until Cycle 4. Inventory fractions measured in solution during Cycles 4 and 5 are plotted in Figure 3.7. Continuous ^{90}Sr release was observed during Cycles 4 and 5 following initial fractional releases that were similar to those measured for ^{137}Cs at the beginning of the cycles. The average continuous release rates between the first and final samples of Cycles 4 were $\sim 1.5 \times 10^{-7}$ per day for the HBR test and 2.2×10^{-7} per day for the TP test assuming that released strontium remained in solution. Average continuous release rates of $\sim 1.5 \times 10^{-7}$ per day in the HBR test and $\sim 1.35 \times 10^{-7}$ per day in the TP test were measured during Cycle 5.

FIGURE 3.7. Inventory Fraction of ^{90}Sr Measured in Solution During Cycles 4 and 5

Quantities of ^{90}Sr measured in different sample types are given in Table 3.9. Strontium-90 was not measured during Cycle 1; was measured on the 154-day, final-solution, rinse, and acid-strip samples in Cycle 2; and was measured on the final-solution, rinse, and acid-strip samples in Cycle 3. Most of the ^{90}Sr measured appears to have been in solution. The amount of ^{90}Sr fractional release measured in Cycles 3 and 4 was marginally less than that measured for ^{137}Cs .

3.3.3 Technetium

Technetium-99 is the only significant radioactive Tc isotope in spent fuel. ^{99}Tc beta decays (213,000-yr half-life) to stable ^{99}Ru . The ^{99}Tc inventory in a 33,000 MWd/MTHM burnup fuel is about 13,000 Ci/1000 MTHM, which is equivalent to about 0.75% of the 1000-yr total radioactive inventory. The ^{99}Tc 10,000-yr EPA cumulative release limit (40 CFR 191, Table 1) is 10,000 Ci/1000 MTHM.⁽¹⁶⁾

Inventory fraction of ^{99}Tc measured in solution is plotted in Figure 3.8. Initial fast releases (determined by extrapolation to Day 0) of about 1.4×10^{-4} and 1.9×10^{-4} of inventory in HBR and TP tests, respectively, occurred at the beginning of Cycle 1, followed by continuous dissolution at the rates of 2.0×10^{-7} and 3.6×10^{-7} per day in these two tests, respectively. Initial fast releases on the order of those observed for ^{137}Cs , and generally a little greater than those observed for uranium, were observed for ^{99}Tc in the later test cycles. The later cycle ^{99}Tc data showed more scatter than the ^{137}Cs data because the ^{99}Tc activities in solution in these samples were, in general, only about an order of magnitude above the detection limit. During Cycle 2 the ^{99}Tc average continuous release rate was about half that observed for ^{137}Cs and was about equal to that of ^{137}Cs ($\sim 3 \times 10^{-7}$ per day) during Cycles 3, 4, and 5.

The ^{99}Tc quantities measured in the different sample types are given in Table 3.10. Since the rinse and acid strip values were often less than detectable, the cycle total values contained more than 5% "less than" values and are, therefore, indicated as "less than" values. Where significant data were available, the data indicated that most of the ^{99}Tc measured was in solution.

TABLE 3.9. ^{90}Sr Quantities Measured (μCi)

	Cycle 1		Cycle 2		Cycle 3		Cycle 4		Cycle 5	
	HBR	TP	HBR	TP	HBR	TP	HBR	TP	HBR	TP
Periodic samples	NM	NM	155 ^(a)	22.2 ^(a)	NM	NM	27.3	6.3	7.2	1.45
Final solution	NM	NM	630	89	227	39.4	134	54.4	82.2	20.4
[Sr(ppb)]	--	--	(32.6)	(4.6)	(11.8)	(2.0)	(6.9)	(2.8)	(4.3)	(1.1)
Rod samples	NM	NM	1	0.3	NM	NM	--	--	--	--
Rinse	NM	NM	71	5.4	31.1	4.9	15.2	3.5	11.9	1.27
Acid strip	NM	NM	30	15.7	3.2	2.6	7.4	2.5	10.5	2.03
Cycle total	--	--	887	132.6	--	--	184	66.7	112	25.15
$\div 10^{-5}$ Inv.	--	--	24.8	12.2	--	--	5.2	6.3	3.2	2.4
% in Solution	--	--	88.5	83.9	--	--	87.7	91	80	86.0

(a) Based on 154-day sample.

NM = not measured.

FIGURE 3.8. Inventory Fraction of ⁹⁹Tc Measured in Solution

TABLE 3.10. ^{99}Tc Quantities Measured (nCi)

	Cycle 1		Cycle 2		Cycle 3		Cycle 4		Cycle 5	
	HBR	TP	HBR	TP	HBR	TP	HBR	TP	HBR	TP
Periodic samples	43 ^(a)	15 ^(b)	8.0	2.4	6.9	<1.1	10.4	3.5	2.0	0.59
Final solution	113	53	56	11.3	55.2	6.3	57.4	13.5	37.2	6.42
[^{99}Tc (ppb)]	(26)	(12.4)	(13)	(2.6)	(13)	(2.5)	(13.5)	(3.2)	(8.7)	(1.5)
Rod samples	<0.3	--	<0.1	<0.1	<0.3	<0.3	--	--	--	--
Rinse	18.1	11.6	<5.4	<5.4	7.0	<5.4	<5.4	<5.4	3.2	<2.7
Acid strip	28.4	6.1	4.0	<2.7	<2.7	4.5	<2.7	<2.7	2.4	<1.35
Cycle total	~203	~86	<74	<22	72.1	<17.6	<75.9	<25.1	44.8	<11.1
$\div 10^{-5}$ Inv.	~23	~32	<8.6	<8.3	8.4	<6.9	<8.9	<9.9	5.3	<4.4
% in Solution	~77	~79	~87	--	86	--	--	--	87.5	--

Summary of Cycles

Σ Cycle totals	~203	~86	<277	<108	<349	<126	<425	<151	<470	<162
$\div 10^{-5}$ Inv.	~23	~32	<32	<41	<40	<48	<49	<58	<54	<62

(a) Assumes 0.45 nCi/mL in all solution samples.

(b) Assumes 0.2 nCi/mL in all solution samples.

3.3.4 Iodine

Iodine-129 beta-decays with a 17,000,000-yr half-life to stable ^{129}Xe and is the only significant radioactive iodine isotope remaining in spent fuel a few years after reactor discharge. With an inventory of about 30 Ci/1000 MTHM in the tested specimens, ^{129}I had the lowest activity of any nuclide measured and required neutron activation analysis for its detection. The EPA 10,000-yr cumulative release limit for ^{129}I is 100 Ci/1000 MTHM, which allows for eventual release of the total ^{129}I inventory. Although ^{129}I has a low inventory in spent fuel, it is relatively soluble, may possibly be mobile in the vapor phase as I_2 , and has a potential for incorporation into the biosphere.

Inventory fraction of ^{129}I measured in solution is plotted in Figure 3.9. The TP test showed a significantly higher fractional ^{129}I release than the HBR test during Cycles 1, 2, and 4, and approximately equal fractional release during Cycles 3 and 5. By extrapolation, the initial rapid release in Cycle 1 of the HBR test was about 5×10^{-5} of inventory. In Cycle 5, following initial releases comparable to those observed for the other fission products, both tests exhibited a uniform continuous release of about 1.5×10^{-7} per day averaged between the first and final solution samples.

The quantities of ^{129}I measured in the different sample types are given in Table 3.11. As with the other fission products, most of the ^{129}I measured was in solution. In contrast to data reported for CANDU fuel,⁽¹⁵⁾ where ^{129}I fractional release from the gap inventory was near that for ^{137}Cs , ^{129}I fractional release in the Series 2 bare fuel tests was two orders of magnitude less than that of ^{137}Cs . Similar ^{129}I release was observed in the 25°C Series 3 test where about 7.7×10^{-5} of the HBR specimen inventory was estimated to have been released to solution during Cycle 1⁽³⁾ compared to about 1×10^{-4} of inventory in the Cycle 1 Series 2 HBR test. However, ^{129}I release exhibited a strong temperature dependence in the Series 3 tests where Cycle 1 ^{129}I fractional release was 30% to 40% of the ^{137}Cs fractional release at 85°C.

FIGURE 3.9. Inventory Fraction of ¹²⁹I Measured in Solution

TABLE 3.11. ^{129}I Quantities Measured (pCi)

	Cycle 1		Cycle 2		Cycle 3		Cycle 4		Cycle 5	
	HBR	TP	HBR	TP	HBR	TP	HBR	TP	HBR	TP
Periodic samples	50 ^(a)	44 ^(b)	28 ^(a)	12 ^(a)	24	6.6	18	5.9	4.2	1.2
Final solution	180	148	117	55	116	35	69	29.5	54.8	16
[I(ppb)]	(5.8)	(4.8)	(3.8)	(1.8)	(3.8)	(1.1)	(2.2)	(1.0)	(1.8)	(0.5)
Rinse	NM	NM	8.5	4.4	6.2	2.3	4.1	1.4	5.4	1.2
Acid strip	NM	NM	8.9	5.7	6.9	4.8	NM	NM	NM	NM
Cycle total	230	192	162	77	153	48.7	91	36.8	64.4	18.4
$\div 10^{-5}$ Inv.	10.5	29.2	7.5	12.0	7.1	7.7	4.2	5.8	3.0	2.9
% in Solution	--	--	89.5	87.0	91.5	85.4	--	--	--	--

Summary of Cycles

Σ Cycle totals	230	192	392	269	545	318	636	355	700	373
$\div 10^{-5}$ Inv.	10.5	29.2	18.0	41.2	25.1	48.9	29.3	54.7	32.3	57.6

(a) Activity measured in a single periodic sample assumed for all periodic samples.

(b) Final solution activity assumed for all periodic solution samples.

NM = not measured.

3.3.5 Fission Product Summary and Matrix Dissolution Rate

The fractional inventories of ^{137}Cs , ^{90}Sr , ^{99}Tc , and ^{129}I measured in solution in the HBR test are compared in Figure 3.10. Gap inventories varied significantly between the fission products, as indicated by the Cycle 1 releases. Only ^{99}Tc and ^{129}I are shown for Cycle 1 since ^{137}Cs release was off scale and ^{90}Sr was not measured during Cycle 1. Based on results from the 25°C Series 3 test, ⁽³⁾ ^{90}Sr would likely have fallen between the ^{137}Cs and ^{99}Tc data in Cycle 1 had it been measured, resulting in a Cycle 1 release order of $^{137}\text{Cs} > ^{90}\text{Sr} > ^{99}\text{Tc} > ^{129}\text{I}$. Different degrees of preferential release for these fission products continued through Cycle 2 as shown in Figure 3.10. Strontium-90 release was actually greater than that for ^{137}Cs in Cycle 2 of the HBR test. However, ^{137}Cs fractional release was slightly greater than that of ^{90}Sr in Cycle 2 of the TP test.

A much more congruent release of fission products was observed starting with Cycle 3. The range of initial fractional release values continued to decrease with each cycle of the HBR test. With the exception of an initially high value for ^{99}Tc in Cycle 5, initial fission product releases during the TP test also approached congruent behavior in the later test cycles. All four fission products exhibited an initial release of about 7×10^{-6} of inventory at the start of Cycle 5 of the HBR test, which is about four times the uranium inventory measured in solution in the first Cycle 5 sample. The fission product-to-uranium fractional inventory ratios in the first Cycle 5 sample from the TP test were 2.1, 1.7, and 2.5 for ^{137}Cs , ^{99}Tc , and ^{129}I , respectively. During Cycle 5, ^{90}Sr and ^{129}I continuous release ranged from 1.35×10^{-7} to 1.65×10^{-7} per day averaged between the first and final sample in both tests. Average continuous release rates for ^{137}Cs in both tests, and ^{99}Tc in the HBR test, during Cycle 5 were in the range of 2.5×10^{-7} to 2.9×10^{-7} per day.

The degree of preferential fission product release presumably decreases with each cycle as the inventory of concentrated fission products is depleted. Assuming that the degrees to which ^{90}Sr and ^{129}I were preferentially released during Cycle 5 were small, a matrix dissolution rate of $\sim 1 \times 10^{-7}$ per day would be indicated. Based on a comparison of fission product and uranium fractional inventories present in initial solution samples (see Table 3.7 for

FIGURE 3.10. Comparison of ^{137}Cs , ^{90}Sr , ^{99}Tc and ^{129}I Inventory Fractions Measured in Solution During the HBR Test

$^{137}\text{Cs}/\text{U}$ ratios) in later test cycles, the amount of ^{90}Sr and ^{129}I preferentially released in the later cycles is probably not greater than twice the amount congruently released with the actinides as the fuel matrix dissolves. Therefore, 1×10^{-7} per day ($\sim 4 \times 10^{-5}$ per year) appears to be a reasonable estimate of the fuel matrix dissolution rate in the later cycles of the Series 2 bare fuel tests. This compares to an estimated matrix dissolution rate of $\sim 10^{-7}$ per day at 25°C for CANDU spent fuel particles.⁽¹⁵⁾ However, such estimates of matrix dissolution rates from static or semi-static test results are somewhat uncertain. Such estimates could be made with more confidence if the degree of preferential dissolution of soluble nuclides could be measured in a "flow-through" test where all dissolved uranium remains in solution.

The $\sim 1 \times 10^{-7}$ per day matrix dissolution rate is not normalized for surface area and would presumably be surface area dependent. The matrix dissolution rate will likely increase with time as a result of fuel degradation. One form of degradation would be an increase in surface area as a result of preferential dissolution of grain boundaries. Significant quantities of fuel grains were released from the fuel particle surfaces in the Series 2 and 3 bare fuel tests and collected in the specimen rinse residues after each test cycle. Another form of degradation likely to effect matrix dissolution and soluble nuclide release rates is oxidation of the fuel. The two effects are probably related in that oxidation may significantly enhance preferential grain boundary dissolution. The average initial particle size in the test specimens was about 2 to 3 mm, which is representative of particle sizes normally formed in the fuel by thermal cracking during irradiation. No attempt was made to normalize the data to surface area, since no reliable method was available to measure the wettable surface area of the fuel specimens.^(a) The use of a surface area-normalized dissolution rate to predict long-term release implies the need to develop a model for the time-dependent surface area and state of the fuel in the repository. Such a model will

(a) Geometric surface areas were calculated to range from 2.1 to 2.6 cm^2/g for HBR and TP bare fuel specimens tested in the Series 3 tests. Geometric surface area determination for these fuels is discussed in Appendix E of PNL-7170 (Ref. 3).

probably be difficult to develop and validate. The NRC and EPA (40 CFR 191, Table 1) release limits are fractional release limits based on nuclide inventories in the repository regardless of the waste form state. Therefore, the most productive approach initially may be to test fuels that have undergone various degrees of degradation to establish bounding values for inventory-normalized dissolution rates for various potential fuel states regardless of the actual wettable surface area of the fuel states.

3.4 ACTIVATION PRODUCTS

Four significant activation products associated with LWR spent fuel are ^{59}Ni , ^{63}Ni , ^{60}Co , and ^{14}C . ^{59}Ni and ^{63}Ni result primarily from activation of natural nickel contained in assembly structural hardware. Except for possible incorporation into cladding "crud" deposits, the Ni activation products were assumed not to be present at significant activity levels in the fuel specimens tested and were not measured. Activities of ^{60}Co and ^{14}C were measured during the tests.

3.4.1 Cobalt-60

Cobalt-60 is an activation product produced primarily by neutron activation of ^{59}Co . ^{60}Co is thought to be produced primarily in fuel assembly structural hardware. With a half-life of 5.26 yr, ^{60}Co is not a concern for long-term containment. However, its beta decay to stable ^{60}Ni is accompanied by two relatively hard gamma emissions (~1.1 and 1.3 MeV). ^{60}Co plus ^{137}Cs and ^{134}Cs account for a major portion of the hard gamma activity that requires heavy shielding during shipping and emplacement in the repository.

Cobalt-60 appeared along with ^{134}Cs and ^{137}Cs during gamma spectrometry analyses of solution samples. Quantities of ^{60}Co measured in the different sample types are given in Table 3.12. Most of the ^{60}Co measured was in solution, indicating that it was soluble. There was a much greater release of ^{60}Co from the TP fuel. ^{60}Co inventory was not determined for either fuel, but the data suggest a significantly greater inventory for the TP fuel. The combined Cycle 1 plus Cycle 2 ^{60}Co releases measured from the TP slit-defected, TP holes-defected, and TP undefected cladding test specimens were 20.9 μCi , 2.88 μCi , and 0.37 μCi , respectively,⁽²⁾ compared to 287 μCi for

TABLE 3.12. ^{60}Co Quantities Measured (μCi)

	Cycle 1		Cycle 2		Cycle 3		Cycle 4		Cycle 5	
	HBR	TP	HBR	TP	HBR	TP	HBR	TP	HBR	TP
Periodic samples	BD	34.7	0.21	9.1	0.31	4.43	0.08	2.08	0.02	0.36
Final solution	BD	154	1.16	32.9	0.94	15.8	0.38	9.44	0.17	4.14
$^{60}\text{Co}(\text{pg/mL})$	--	(544)	(4.1)	(116)	(3.3)	(56)	(1.3)	(33)	(0.6)	(15)
Rod samples	0.3	0.4	BD	0.6	0.005	0.21	--	--	--	--
Rinse	BD	47.6	BD	1.6	BD	0.52	BD	0.46	BD	0.22
Acid strip	BD	5.0	BD	1.1	BD	0.89	BD	0.86	BD	0.15
Cycle total	0.3	242	1.37	45	1.26	21.8	0.46	12.8	0.19	4.87
% in Solution	--	78	--	93	--	93	--	90	--	92

Summary of Cycles

Σ Cycle totals	0.3	242	1.67	287	2.93	309	3.39	322	3.58	326
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BD = below detection.

the TP bare fuel specimen. (^{60}Co was generally below detection in the slit-defected, holes-defected and undefected HBR fuel tests.) These data indicate that the ^{60}Co was primarily released from the TP fuel rather than from exterior cladding crud deposits as had been expected. Relatively rapid release at the beginning of Cycle 1 in the TP test, followed by continuous slower release, and progressively lower releases in subsequent cycles, is similar to the release behavior of the soluble fission products.

3.4.2 Carbon-14

Carbon-14 (5730-yr half-life) is an activation product formed during irradiation by the (n,p) reaction on nitrogen impurities, and from the (n, α) reaction on ^{17}O .⁽¹⁷⁾ ORIGEN calculations for ^{14}C inventories in spent fuel depend on assumed values for initial ^{14}N impurity levels in the fuel and cladding, which are not generally well known and may vary significantly between individual fuel samples. Also, current ORIGEN predictions do not include estimates of ^{14}C produced from ^{17}O in the reactor primary coolant that may become incorporated on the cladding and assembly surfaces. Carbon-14 was radiochemically measured on two fuel and cladding samples taken from the HBR ATM-101 C5 rod that was used to prepare specimens for the Series 2 and Series 3 tests. The average of the two ^{14}C analyses gave 0.49 $\mu\text{Ci/g}$ for fuel and 0.53 $\mu\text{Ci/g}$ for cladding. An additional ^{14}C analysis on a fuel sample from the HBR ATM-101 N9 rod gave 0.33 $\mu\text{Ci/g}$. Carbon-14 is of particular concern because it is mobile in the vapor phase as CO_2 and in groundwater as HCO_3^- , and has a high potential for incorporation in the biosphere.

The quantities of ^{14}C activity measured in different sample types in the Series 2 bare fuel tests are given in Table 3.13. Results from the Series 3 tests in sealed vessels indicated that most of the ^{14}C released in the unsealed Series 2 tests was probably lost to the atmosphere as CO_2 and not measured.^(3,13) Therefore, significant ^{14}C results from the Series 3 tests using HBR and TP fuel from the same assemblies will be summarized here. ^{14}C activities measured in solution samples from the Series 3 TP test were almost an order of magnitude greater than measured in the Series 3 HBR tests. In the Series 3 HBR tests, ^{14}C fractional release ranged from about 0.5% to 1.2% of inventory, which was on the order of that measured for ^{137}Cs and greater than measured for any other nuclide. Comparatively little ^{14}C was measured from

TABLE 3.13. ^{14}C Quantities Measured^(a) (nCi)

	Cycle 1		Cycle 2		Cycle 3		Cycle 4		Cycle 5	
	HBR	TP	HBR	TP	HBR	TP	HBR	TP	HBR	TP
Periodic samples	1.9 ^(b)	NM	1.5 ^(b)	3.5 ^(b)	2.6	3.2	3.8	4.0	0.7	0.7
Final solution	6.4	13.2	6.1	11.3	6.3	6.1	6.8	4.3	5.3	6.0
$[^{14}\text{C}(\text{pg/mL})]$	(5.8)	(11.8)	(5.4)	(10.0)	(5.6)	(5.4)	(6.0)	(3.8)	(4.7)	(5.4)
Rinse	NM	NM	3.2	3.5	3.0	25.4	<2.7	<2.7	3.0	<2.7
Cycle total	8.3	13.2	10.8	18.3	11.9	34.7	<13.3	<11.0	9.0	<9.4
$\div 10^{-5}$ Inv.	16.8	--	22.1	--	24.5	--	<27.4	--	18.7	--
<u>Summary of Cycles</u>										
Σ Cycle totals	8.3	13.2	19.1	31.5	31.0	66.2	<44.3	77.2	<53.3	<86.6
$\div 10^{-5}$ Inv.	16.8	--	38.9	--	63.4	--	<90.8	--	<109.5	--

(a) Most of the ^{14}C released was lost to the atmosphere as CO_2 and was not measured.

(b) ^{14}C measured in 63-day Cycle 1 sample and 154-day Cycle 2 sample was assumed for all periodic solution samples for those cycles.

NM = not measured.

the undefected specimen indicating that release was primarily from the fuel or gap inventory. The measured ^{14}C release showed little dependence on temperature or bare fuel versus fuel in defected cladding, suggesting that most of the measured release originated from gap or gap plus grain boundary inventory.

The ^{14}C activity range for all of the Series 2 periodic and final solution samples was about 10 to 70 pCi/mL. As $\text{H}^{14}\text{CO}_3^-$, 10 pCi/mL would be equivalent to about 1.1×10^{-5} ppm compared to a HCO_3^- concentration in the J-13 water that remained at about 120 ppm during the Series 2 tests. At steady state, approximately 10^{-6} to 10^{-7} of the bicarbonate contained ^{14}C . In the Series 3 HBR tests, ^{14}C activities measured in solution were generally in the 100 to 1000 pCi/mL range, and up to 6000 pCi/mL was measured in the Series 3 TP test.

3.5 RINSE AND ACID STRIP SUMMARY

The primary purposes for the cycle termination rinse procedure were to remove precipitated material from the specimen surfaces before starting the next cycle and to remove undissolved fuel fines from vessel components before acid stripping. The fuel was rinsed by rocking side-to-side in a beaker with J-13 water so that the fuel particles tumbled from side-to-side across the bottom of the beaker. The first time this procedure was performed, the J-13 water became dark and turbid with suspended particles. After decanting off the dark turbid rinse water, the procedure was repeated until the decanted J-13 water came off clear. Five rinses were required before the decanted rinse water was clear, and five rinse cycles were used thereafter in the bare fuel rinsing procedure. Later examination of rinse solution residues indicated that most of the particles removed from the specimen were loose fuel grains apparently released from the fuel surface as a result of preferential grain boundary dissolution. Phases containing actinide elements that precipitated out of solution or remained at the fuel surface as undissolved secondary phases were also presumed to be removed by the rinse procedure.

Specimen and vessel rinse solutions were collected in a single beaker, the volume made up to 600 mL, and the rinse solution allowed to settle overnight before taking a sample from the top of the solution for analysis. Specimen inventory fractions of several nuclides measured in the 600 mL of

TABLE 3.14. Inventory Fractions Measured in Rinse Solutions^(a) ($\times 10^6$)

Nuclide	Cycle 1		Cycle 2		Cycle 3		Cycle 4		Cycle 5	
	HBR	TP	HBR	TP	HBR	TP	HBR	TP	HBR	TP
U	9.46	15.9	1.48	1.73	0.97	1.13	0.63	0.59	1.68	1.00
²³⁹⁺²⁴⁰ Pu	4.11	3.27	4.16	1.42	0.96	1.10	0.67	0.84	1.23	0.87
²⁴¹ Am	4.10	2.81	1.09	1.30	0.93	1.11	0.83	1.06	1.39	1.06
²⁴⁴ Cm	4.93	3.79	6.01	1.37	0.95	1.01	0.76	0.92	1.30	0.92
¹³⁷ Cs	281.5	482.5	9.25	8.73	7.51	5.89	5.57	6.38	5.43	3.68
⁹⁰ Sr	NM	NM	20.8	5.03	9.16	4.62	4.50	3.34	3.53	1.21
⁹⁹ Tc ^(b)	20.5	43.2	<6.3	<21	8.2	<21	<6.4	<21	3.8	<10.6

(a) 0.4- μ m filtered.(b) ⁹⁹Tc activities were near detection limits.

NM = not measured.

rinse solution are given in Table 3.14. Actinide concentrations in the Cycle 1 rinse solutions were similar to those observed in the test solutions suggesting that approximately the same steady state was achieved with fuel grains and secondary phases removed with the rinse water. The preferential ^{137}Cs content of the Cycle 1 rinse solutions is probably caused primarily by residual test solution not removed by decanting that contained excess ^{137}Cs from the gap inventory. In later test cycles, the actinides appear to be congruently dissolved in the rinse solutions, which may be a result of dissolution from previously undissolved fuel grains. The more soluble fission product nuclides (^{137}Cs , ^{90}Sr and ^{99}Tc) appear to have been preferentially dissolved in the rinse solution samples.

Nuclide inventory fractions measured in the acid strip solutions are given in Table 3.15. The transuranic actinides plutonium, americium, and curium were contained in the acid strip samples in near-direct proportion to their test inventories. The lesser inventory fractions for uranium (and fission products in later cycles) suggest that, although part of the acid strip inventories may have originated from previously undissolved fuel fines, the transuranic actinides may have congruently plated-out in proportion to their rates of release during fuel dissolution. The transuranic actinides are preferentially formed in the outer circumferential region of the fuel during irradiation, and preferential plate-out originating from fuel near the outer pellet surfaces could also explain the preferential occurrence of the transuranic nuclides relative to uranium in the acid strip samples.

3.6 SOLUTION CHEMISTRY

Inductively coupled plasma (ICP) emission spectrometry analyses for cations, ion chromatography (IC) analyses for anions, and inorganic carbon analyses for bicarbonate were conducted on cycle starting J-13 well water samples at the beginning of each test cycle, on final solution samples, and on selected periodic solution samples. Results from these analyses are tabulated in Appendix B. A substantial drop in the concentration of any of these solution species would indicate precipitation of solid phases, which in some cases may incorporate radionuclides. No consistent decreases in concentrations of solution species contained in the starting J-13 well water were

TABLE 3.15. Inventory Fractions Measured in Acid Strip Solutions ($\times 10^6$)

Nuclide	Cycle 1		Cycle 2		Cycle 3		Cycle 4		Cycle 5	
	HBR	TP	HBR	TP	HBR	TP	HBR	TP	HBR	TP
U	37.7	41.6	4.35	6.91	1.36	7.16	1.09	1.31	2.69	1.92
$^{239+240}\text{Pu}$	65.6	58.0	5.57	10.9	2.71	10.5	2.87	2.33	5.04	2.78
^{241}Am	74.0	55.3	6.07	10.3	3.13	10.3	3.51	2.76	6.15	3.59
^{244}Cm	78.2	58.6	6.49	10.6	3.14	11.3	3.34	2.71	5.35	3.38
^{137}Cs	110.0	84.7	4.56	8.23	3.20	7.59	2.45	2.36	3.39	1.95
^{90}Sr	NM	NM	8.78	14.6	0.94	2.47	2.09	2.38	3.00	1.94
$^{99}\text{Tc}^{(a)}$	32.5	23.0	4.65	<10.4	<3.2	17.7	<3.2	<10.6	2.87	<5.3

(a) ^{99}Tc activities were near detection limits.
 NM = not measured.

observed during the Series 2 tests. These data are consistent with data from the 25°C Series 3 test, where stable concentrations of species contained in the starting J-13 well water were also observed.⁽³⁾ Sample-to-sample variations in these data were generally small and attributed to analytical limitations. A few results, such as the low potassium value for the Cycle 2 starting J-13 water, appear to be flyer data points and are most likely the result of analytical error.

The solution chemistry results did not indicate the occurrence of any significant contamination or other significant chemistry variables during the five testing cycles. A slight alkaline pH shift was observed during most test cycles. In the pH range covered by the tests (7.2 to 8.6), pH would be buffered by dissolved HCO_3^- . The values given for HCO_3^- in Appendix B were calculated from "inorganic carbon" concentrations corrected for molecular weight by multiplying times a factor of 5.0833.

3.7 SOLIDS CHARACTERIZATION

Precipitation of phases that showed Si as the major elemental constituent detectable by EDS microanalysis in the SEM was documented in Reference 2 for Cycles 1 and 2 of the Series 2 bare fuel tests. These phases were thought to be formed as colloidal silica or silica gel. Apparent silica flocs on Cycle 3 solution sample filters are shown in Figure 3.11. Fuel particles that were presumably located on top of the bare fuel particle specimens during the test were also found to be coated with a phase assumed to be precipitated silica gel (Figure 3.12). Amorphous-appearing deposits observed in rinse solution filter residues showed Si, or Si together with varying proportions of U, as the only elements detected by EDS microanalysis in the SEM (Figure 3.13). However, the rinse filter residues also contained significant quantities of undissolved fuel grains, and the U lines observed with Si lines during EDS analyses of these phases may have originated from fuel particles under the low-density silica deposits examined in the SEM. The apparent quantities of Si-containing phases observed in these tests seem to be significant compared to the 7.5 mg of Si present in the starting J-13 well water. Since the concentration of Si did not drop, Si dissolution from the fused silica test vessels during the tests may be indicated.

HBR Cycle 3 224 Day Sample

2 μm

TP Cycle 3 224 Day Sample

2 μm

FIGURE 3.11. Floccules Retained on 0.4-μm Filters Used to Filter Solution Samples. Only the element Si was detected by EDS microanalysis of the floccule phase.

FIGURE 3.12. Fuel Particle (A) and Scale Particles (B) from HBR Cycle 1 Coarse Rinse Sediment with EDS Microanalysis Results Given for Selected Spots

FIGURE 3.13. Fuel Particles and Amorphous-Appearing Deposit on Rinse Solution Filter from Cycle 5 of the TP Test. [EDS spectrum (top) is shown for indicated spots. Uranium shown in the EDS spectrum may be from fuel particles under the amorphous-appearing phase.]

Quantities of solids residues present as coarse sediments and collected on rinse filters at the ends of test cycles were given in Table 2.4. The SEM examination of several samples from these residues indicated that the most abundant phase present in these samples was particles or individual grains of fuel. The second most abundant phase was the amorphous-appearing Si containing phases discussed above. Particles with a faceted, plate-like appearance showing only calcium in their EDS spectra were observed in several samples and were presumed to be calcite (Figure 3.12). However, calcite precipitation was probably limited since solution Ca concentration did not drop substantially and no other sources for Ca are identified. A few particles showing only Zr or Zr plus U were assumed to be cladding scale. A single particle having a Mg-Si composition was also observed. A few agglomerates observed on fuel particle surfaces showed Si, Ca, and U lines in their EDS spectra, which may result from co-precipitated silica and calcite on a fuel substrate or from a Ca-U-Si secondary phase.

X-ray diffraction examination was performed on a section of the Cycle 3 rinse solution filter from the HBR test. The pattern obtained is shown in Figure 3.14 along with JCPDS reference "stick patterns" for UO_2 (JCPDS File No. 5-550), calcite (24-27), and haiweeite (13-118). Peaks in the sample pattern that are matched by lines in the reference patterns are identified as U, C, and H, respectively. Indexing information for 19 peaks resolved in the sample pattern is given in Table 3.16. The haiweeite identification is tentative since only one peak at $d = 9.247\text{\AA}$ is matched and other major lines in reference patterns are missing. Preferred crystalline orientation on the filters is a possible explanation for absence of other haiweeite lines in the sample pattern. The same apparent line at about $d = 9.30\text{\AA}$ was observed in XRD patterns of three rinse filters from 85°C Series 3 tests with only questionable weak indications for a few other lines from the reference patterns. Three haiweeite patterns that provide a good match for this line are contained in the JCPDS files. These files, along with the lattice parameter of the most intense line, are 13-118, $d = 9.30\text{\AA}$; 12-721, $d = 9.26\text{\AA}$; and 22-160, $d = 9.16\text{\AA}$. Although the line in the 12-721 pattern best matches the line in the Series 2 Cycle 3 HBR rinse filter pattern, this pattern contains extra, relatively intense lines not in the 13-118 pattern, and the 13-118 pattern best matches

FIGURE 3.14. X-Ray Diffraction Pattern from Cycle 3 HBR Test Rinse Filter and Reference JCPDS Patterns for UO_2 (U), Haiweeite (H), and Calcite (C)

the patterns from the 85°C Series 3 rinse filters.^(a) A search was made of reference patterns for other compounds containing uranium and elements in J-13 water, and the haiweeite patterns were the only ones found with a strong line that closely matched the $d = 9.247\text{\AA}$ line in the sample pattern. Five lines of the sample pattern listed in Table 3.16 were not matched.

Secondary phase formation appears to be temperature dependent. In the 85°C Series 3 tests in stainless steel vessels, drops in Ca and Si concentrations correspond to the precipitation of the calcium-uranium-silicate secondary phases uranophane and haiweeite.⁽³⁾ A drop in solution Ca, Mg and HCO_3^- concentrations correlated with the appearance of acid-soluble white scale presumed to be calcite at the water-line in the 85°C test vessels. No significant drops in the concentrations of J-13 water species were observed in the

(a) A more extensive discussion of JCPDS reference data and evaluation of XRD patterns from rinse filters is provided in reference 3.

TABLE 3.16. Indexing for HBR Cycle 3 Rinse Filter XRD Pattern

Deg. 2θ	d (Å)	I (%)	Matched Line ^(a)		
			Phase	d (Å)	I (%)
5.40	16.057	0.5	--	--	--
9.45	9.247	3.7	H	9.26	100
28.16	3.154	100.0	U	3.157	100
28.58	3.109	7.8	--	--	--
29.40	3.025	41.7	C	3.030	100
32.60	2.738	24.0	U	2.735	48
36.07	2.482	0.7	C	2.495	7
39.45	2.276	1.3	C	2.284	18
43.20	2.088	1.9	C	2.094	27
46.84	1.934	38.9	U	1.934	49
48.57	1.869	2.2	C	1.873	34
55.58	1.649	31.6	U	1.649	47
58.30	1.580	8.9	U	1.579	13
60.99	1.516	2.1	--	--	--
64.68	1.438	2.6	--	--	--
68.40	1.368	4.9	U	1.368	9
70.24	1.334	0.6	--	--	--
75.57	1.255	10.2	U	1.255	18
77.86	1.225	6.8	U	1.223	15

- (a) U = UO₂, JCPDS File No. 5-550.
 C = calcite, JCPDS File No. 24-27.
 H = haiweeite, JCPDS File No. 12-721.

25°C Series 2 or Series 3 tests. Crystalline appearing secondary phases were evident to a much greater extent in residues from the 85°C tests than from the 25°C test. In particular, extensive quantities of uranophane crystals observed in the 85°C residues were not present in the 25°C residues.

4.0 SUMMARY AND CONCLUSIONS

Radionuclide releases were measured from PWR spent fuel specimens tested in NNWSI J-13 well water in unsealed fused silica vessels under ambient hot cell air conditions (25°C). Two bare fuel specimens were tested, one prepared from a rod irradiated in the H. B. Robinson (HBR) Unit 2 reactor and the other from a rod irradiated in the Turkey Point (TP) Unit 3 reactor. Both fuels were low-gas release and moderate burnup. The specimen particle size range (2 to 3 mm) was that which occurs in the fuel as a result of thermal cracking. A semi-static test method was used in which the specimens were tested for multiple cycles starting in fresh J-13 water. Periodic water samples were taken during each cycle with the sample volume (~10% of test solution) being replenished with fresh J-13 water. The specimens were tested for five cycles for a total time of 34 months.

4.1 PRINCIPAL OBSERVATIONS AND CONCLUSIONS

1. Actinide concentrations appeared to rapidly reach steady-state levels during each test cycle. Concentrations of Pu, Am, and Cm were dependent on filtration, with Am and Cm concentrations being affected the most by filtration, suggesting that these elements may have formed colloids. Approximate steady-state concentrations of actinide elements indicated in 0.4- μ m filtered solution samples are given below.

U -- 4×10^{-6} to 8×10^{-6} M (1 to 2 ppm)

Pu -- 8.8×10^{-10} to 4.4×10^{-9} M (20 to 100 pCi/mL $^{239+240}\text{Pu}$)

Am -- $\sim 1.5 \times 10^{-10}$ M (~100 pCi/mL ^{241}Am)

Cm -- $\sim 2.6 \times 10^{-12}$ M (~50 pCi/mL ^{244}Cm)

Np -- 2.4×10^{-9} M (0.4 pCi/mL ^{237}Np).

2. Actinide releases as a result of water transport should be several orders of magnitude lower than the NRC 10 CFR 60.113 release limits (10^{-5} of 1000-yr inventory per year) if actinide concentrations (true solution plus colloids) in the repository do not greatly

exceed the steady-state concentrations measured in 0.4- μ m filtered samples. Assuming a water flux through the repository of 20 L per year per waste package containing 3140 kg of spent fuel saturates at the actinide elemental concentrations given above, the following annual fractional releases are calculated based on 1000-yr inventories for 33,000 MWD/MTHM burnup PWR fuel:

U (8×10^{-6} M), 1.4×10^{-8} per year

Pu (4×10^{-9} M), $\sim 1 \times 10^{-9}$ per year

Am, $\sim 8 \times 10^{-10}$ per year

Cm, $\sim 1 \times 10^{-8}$ per year

Np, $\sim 3 \times 10^{-9}$ per year.

3. Gap inventory ^{137}Cs releases of about 0.7% of inventory in the HBR test and about 0.2% of inventory in the TP test were measured at the start of Cycle 1. Smaller initial Cycle 1 releases on the order of 10^{-4} of inventory were measured for ^{129}I and ^{99}Tc .
4. Fission product nuclides ^{137}Cs , ^{90}Sr , ^{99}Tc , and ^{129}I were continuously released with time and did not reach saturation in solution. The continuous release rates of these soluble nuclides were relatively constant during Cycles 3, 4, and 5. During Cycle 5, the release rate for both ^{90}Sr and ^{129}I was about 5.5×10^{-5} of inventory per year in both tests. Marginally higher continuous release rates on the order of 1×10^{-4} of inventory per year were measured for ^{137}Cs and ^{99}Tc .
5. The degree to which the soluble nuclides (^{137}Cs , ^{90}Sr , ^{99}Tc , and ^{129}I) were preferentially released relative to the amount of congruent dissolution of the UO_2 matrix phase was not quantitatively measured. However, the near-congruent release of soluble nuclides in later test cycles, and the inventory ratios of these nuclides to that of uranium in initial solution samples from the later cycles (a ratio of about 2.5 for ^{137}Cs), suggest that the fractional release rates for these nuclides may not have greatly exceeded the matrix dissolution rate. A matrix dissolution rate of about

4×10^{-5} per year appears to be a reasonable estimate for the 2- to 3-mm size fuel particles tested based on these data.

6. The present data suggesting fuel matrix dissolution rates greater than 10^{-5} per year imply that demonstrating 10 CFR 60.113 compliance for soluble nuclides will involve considerations other than the durability of the spent fuel waste form, such as scenarios for low-probability water contact, a distribution of cladding/container failures over time, or very low migration rates. In time, fuel degradation resulting from oxidation and grain boundary dissolution (increasing surface area) may increase the matrix dissolution rate. Upper limits for degraded fuel matrix dissolution rates are yet to be determined.
7. Comparison to the Series 3 tests (sealed vessels) indicated that most of the ^{14}C released in the Series 2 tests was lost to the atmosphere as CO_2 and not measured. The ^{14}C was preferentially released in the Series 3 tests at about 1% of its inventory measured in HBR fuel samples. As an activation product derived partially from nitrogen impurities, evaluation of ^{14}C release relative to 10 CFR 60.113 is complicated because its inventory and distribution in fuel is not well characterized.
8. The quantities of precipitated secondary phase material observed in filter residues were significantly less than observed in the 85°C Series 3 tests. UO_2 and calcite were the only phases confirmed by XRD examination of a cycle termination rinse filter, with a tentative indication of haiweeite based on a single line in the XRD pattern. Amorphous-appearing, silicon-containing phases were also observed by SEM on the rinse filters, and silicon-containing flocs were observed on filters used to filter solution samples. With the possible exception of haiweeite for uranium, phases controlling the solubility of actinide nuclides were not identified.

4.2 ADDITIONAL DATA NEEDS AND RECOMMENDATIONS

Probably the most immediate fuel dissolution data need is to better determine long-term matrix dissolution rates and the dependent release rates for soluble nuclides. As a start, a better determination of the actual rate of matrix dissolution in the current NNWSI semi-static tests would be helpful. Flow-through dissolution tests, in which all dissolved uranium remains in solution to be measured along with more soluble radionuclides, should allow determination of the degree to which soluble radionuclides are preferentially dissolved, and provide a means for estimating matrix dissolution rates in static or semi-static dissolution tests.

Defining potential fuel degradation states that may occur in the post-containment repository environment, and conducting dissolution tests with fuel specimens representative of such degradation states, are recommended to provide data for estimating bounding values for soluble nuclide release rates. One form of fuel degradation likely to effect matrix dissolution and soluble nuclide release rates is oxidation of the fuel. Another form of degradation would be increased surface area as a result of preferential grain boundary dissolution. The two effects are related in that oxidation is likely to enhance preferential grain boundary dissolution and increase wettable surface area. Oxidized fuel specimens from low-temperature spent fuel oxidation studies are currently available for dissolution testing.

Estimation of wettable surface areas for tested specimens, if possible, may allow determination of surface area normalized dissolution rates that correlate more directly to chemical models. Factors such as grain boundary exposure and fuel porosity make it difficult to estimate effective surface areas for fuel specimens such as those in the Series 2 bare fuel tests. One approach to measurement of area normalized dissolution rates is to use fuel specimens that have been crushed to individual grains. Geometric surface areas were determined for HBR and TP bare fuel specimens tested in the Series 3 tests. Geometric surface areas ranged from 2.1 to 2.6 g/cm² depending on fuel type and particle shape assumptions. Determination of geometric surface area for these two fuels is discussed in Appendix E of the Series 3 report.⁽³⁾

Identification of secondary phases controlling actinide concentrations in the laboratory dissolution tests is important if these data are to be used with confidence for validation of geochemical modeling codes. Development of methods for characterization of precipitated residues from the dissolution tests is recommended. Also, the potential for transport of sparingly soluble nuclides in the colloid state is not well understood.

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APPENDIX A

RADIONUCLIDE INVENTORY AND RADIOCHEMISTRY DATA

APPENDIX A

RADIONUCLIDE INVENTORY AND RADIOCHEMICAL DATA

A.1 RADIONUCLIDE INVENTORY DATA

Specimen radionuclide inventories used for most calculations in this report were calculated from ORIGEN-2 data given in PNL-5109^(A1) for the ATM-101 H. B. Robinson Unit 2 PWR fuel 12 years after reactor discharge. Since the Turkey Point fuel was similar (same vendor, same design, similar vintage and same 2.55% ²³⁵U initial enrichment), these ORIGEN-2 data were considered appropriate for both fuels. An age of 10.5 years from discharge was used for the Turkey Point fuel. Linear interpolation was applied to correct the tabulated ORIGEN-2 data for age and burnup. A factor of 0.8815 was then used to convert the inventories from a "per gram metal" basis to a "per gram fuel" basis. The resulting per gram fuel radionuclide inventories are given in Table 2.3 of the text. Specimen weights required for calculating per specimen radionuclide inventories are contained in Table 2.2 of the text.

A.2 RADIOCHEMICAL DATA

Results from uranium analyses were generally reported in $\mu\text{g/mL}$ (ppm) units. Results for other radiochemical analyses were generally reported as disintegrations per minute (dpm) per mL of solution. Rod sample results were reported as dpm/rod ($\mu\text{g/rod}$ for uranium). Data were converted from dpm to pCi units using the conversion factor of 1 pCi = 2.2 dpm. Concentrations were calculated from the pCi/mL data using Equations (A.1) and (A.2). The $\mu\text{g/pCi}$ and isotope/element conversion factors for Equation (A.1) are contained in Table A.1. The radiochemical results for all sample analyses in pCi units (μg units for uranium) are given for each cycle of both tests in Tables A.2 through A.5 of this appendix.

TABLE A.1. Activity-Concentration Conversion Factors

<u>Radionuclide</u>	<u>($\mu\text{g/pCi}$)</u>	<u>Turkey Point^(a)</u>	<u>H. B. Robinson^(a)</u>
^{14}C	2.24 E-7	--	--
^{60}Co	8.82 E-10	--	--
^{90}Sr	7.07 E-9	0.546	0.546
^{99}Tc	5.87 E-5	1.000	1.000
^{126}Sn	3.52 E-7	0.30	0.30
^{129}I	6.13 E-3	0.76	0.76
^{137}Cs	1.16 E-8	0.40	0.40
^{237}Np	1.42 E-3	0.999	0.999
^{238}Pu	5.72 E-8	0.012	0.014
^{239}Pu	1.63 E-5	0.595	0.577
^{240}Pu	4.41 E-6	0.255	0.263
^{241}Am	3.09 E-7	0.863	0.840
^{244}Cm	1.20 E-8	0.930	0.930

(a) Isotope-to-element mass ratio based on ORIGEN-2 data in PNL-5109^(A1) interpolated to 27.7 MWd/kgM for Turkey Point and 30.2 MWd/kgM for H. B. Robinson burnup, at 10.5 and 12 years after discharge, respectively.

$$\text{Elemental Concentration } (\mu\text{g/mL}) = \frac{\text{Activity (pCi/mL)} \times (\mu\text{g/pCi})}{\text{isotope/element}} \quad (\text{A.1})$$

For conversion to molarity:

$$\text{Molarity (mole/L)} = \frac{\mu\text{g/mL}}{1000 \times \text{atomic mass}} \quad (\text{A.2})$$

For calculation of plutonium concentration from $^{239+240}\text{Pu}$ pCi/mL data using Equation (A.1), the ^{239}Pu (or ^{240}Pu) pCi/mL value is needed. The $^{239}\text{Pu}/^{239+240}\text{Pu}$ activity ratio should be 0.374 for the HBR fuel and 0.389 for the TP fuel based on the PNL-5109^(A1) ORIGEN-2 data.

TABLE A.2. Radiochemical

DAY	SAMPLE TYPE	VOLUME ml	pH	U			Pu-239-Pu-240			Am-241	
				UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A	UNFILTER	0.4
1	Sol	5	7.742	3.30E-00			8.60E-02			3.97E-03	
6	Sol	10	7.958	4.50E-00	3.80E-00	3.70E-00	2.13E-03	1.78E-02	9.82E-01	8.29E-03	1.03
6	Rod			5.20E-00			8.15E-03			3.64E-04	
6	Rinse	**		1.70E-00			2.62E-03			1.21E-04	
20	Sol	5	8.262	4.00E-00			6.40E-02			3.04E-03	
30	Sol	15	8.207	3.70E-00	3.50E-00	3.40E-00	1.97E-02	1.94E-02	1.14E-02	1.03E-03	9.50
30	Rod			4.90E-00			1.02E-04			4.30E-04	
63	Sol	25	8.221	2.50E-00	2.40E-00	2.30E-00	1.78E-02	1.20E-02	4.80E-01	9.32E-02	6.31
63	Rod			4.60E-00			8.15E-03			3.63E-04	
120	Sol	15	8.542	1.70E-00	1.60E-00	1.70E-00	1.49E-02	1.22E-02	2.40E-01	7.21E-02	5.81
120	Rod			4.60E-00			6.85E-03			3.05E-04	
181	Sol	20	8.458	1.40E-00	1.40E-00	1.30E-00	1.40E-02	9.64E-01	2.12E-01	6.58E-02	4.82
181	Rod			7.90E-00			1.35E-04			5.80E-04	
223	Sol	250	8.495	1.20E-00	1.20E-00	1.20E-00	1.12E-02	7.62E-01	2.61E-01	1.11E-03	2.91
223	Rod			6.90E-00			9.41E-03			3.99E-04	
223	Rinse	600			1.10E-00			4.23E-02			1.82
223	Strip	300		9.00E-00			1.35E-04			6.22E-04	
CYCLE 2											
20	Sol	20	8.427	2.40E-00	2.40E-00	2.40E-00	1.47E-02	1.40E-02	6.94E-01	4.50E-02	3.93
20	Rod			7.50E-01			3.00E-03			1.51E-04	
62	Sol	20	8.460	2.20E-00	2.10E-00	2.00E-00	9.80E-02	7.75E-02	3.00E-02	2.40E-03	2.09
62	Rod			1.40E-01			1.68E-05			7.70E-05	
154	Sol	25	8.200	2.00E-00	2.00E-00	2.00E-00	4.41E-01	3.20E-01	1.94E-01	1.31E-02	9.19
202	Sol	250	8.560	2.00E-00	2.00E-00	2.00E-00	3.33E-01	2.60E-01	1.62E-01	1.00E-02	6.94
202	Rod			3.50E-00			1.00E-03			4.82E-03	
202	Rinse	600			1.70E-01			3.92E-02			1.94
202	Strip	300		1.00E-00			1.13E-03			5.32E-03	
CYCLE 2											
20	Sol	20	8.427	2.40E-00	2.40E-00	2.40E-00	1.47E-02	1.40E-02	6.94E-01	4.50E-02	3.93
20	Rod			7.50E-01			3.00E-03			1.51E-04	
62	Sol	20	8.460	2.20E-00	2.10E-00	2.00E-00	9.80E-02	7.75E-02	3.00E-02	2.40E-03	2.09
62	Rod			1.40E-01			1.68E-05			7.70E-05	
154	Sol	25	8.200	2.00E-00	2.00E-00	2.00E-00	4.41E-01	3.20E-01	1.94E-01	1.31E-02	9.19
202	Sol	250	8.560	2.00E-00	2.00E-00	2.00E-00	3.33E-01	2.60E-01	1.62E-01	1.00E-02	6.94
202	Rod			3.50E-00			1.00E-03			4.82E-03	
202	Rinse	600			1.70E-01			3.92E-02			1.94
202	Strip	300		1.00E-00			1.13E-03			5.32E-03	

DAY	SAMPLE TYPE	VOLUME ml	pH	Co-60			SrY-90			Np-239	
				UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A	UNFILTER	0.4
1	Sol	5	7.742								
6	Sol	10	7.958								
6	Rod										
6	Rinse	**									
20	Sol	5	8.262								
30	Sol	15	8.207								
30	Rod										
63	Sol	25	8.221	<1.80E-04	<1.80E-04	<1.80E-04				6.31E-01	3.60
63	Rod			1.40E-03						2.70E-00	
120	Sol	15	8.542	<3.30E-04	<2.93E-04	<2.93E-04					
120	Rod			<1.00E-04						<4.50E-00	
181	Sol	20	8.458							<4.50E-01	<4.50
181	Rod			1.07E-04						2.25E-00	
223	Sol	250	8.495	<2.94E-04	<3.35E-04	<2.94E-04				<4.50E-01	<4.50
223	Rod			1.45E-04							
223	Rinse	600			<6.22E-03					<4.50	
223	Strip	300		<2.64E-03						3.15E-00	
CYCLE 2											
20	Sol	20	8.427	2.60E-03	2.80E-03	2.57E-03				<5.41E-01	4.50
20	Rod			<1.04E-04							
62	Sol	20	8.460	3.50E-03	3.94E-03	3.20E-03				4.50E-01	<4.50
62	Rod										
154	Sol	25	8.200	3.44E-03	4.41E-03	2.65E-03	2.30E-06				
202	Sol	250	8.560	4.64E-03	3.81E-03	3.07E-03	2.62E-06			3.60E-01	4.05
202	Rod						9.01E-05			2.21E-00	
202	Rinse	600						1.18E-05			<2.25
202	Strip	300					9.91E-04			2.25E-01	

UNITS: Solution (Sol), Rinse and Strip samples in pCi/ml for all but Uranium, ug/ml for Uranium.

Rod samples in pCi/rod for all but Uranium, ug/rod for Uranium.

* Am-241 values through 181-day rod sample calculated from Pu-239-Pu-240 and Am-241-Pu-238 values using

** Rod rinse sample reported in pCi/rod (ug/rod for Uranium).

[illegible]

A.3

TABLE A.3. Radiochemical Data

DAY	SAMPLE TYPE	VOLUME ml	pH	U			Pu-239-Pu-240			Am-241-Pu-238		
				UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A
4	Sol	20	8.32	2.98E-00	2.58E-00	2.68E-00	2.91E-02	2.09E-02	1.65E-02	1.63E-03	7.78E-02	4.8E-03
4	Rod			2.88E-00			3.48E-03			2.88E-04		
14	Sol	20	8.33	2.40E-00	2.40E-00	2.40E-00	1.72E-02	1.21E-02	6.22E-01	1.88E-03	4.88E-02	1.8E-03
63	Sol	20	8.44	2.12E-00	2.12E-00	2.12E-00	1.13E-02	7.48E-01	2.61E-01	6.31E-02	3.13E-02	7.8E-03
63	Rod			1.48E-00			1.64E-03			7.93E-03		
112	Sol	20	8.38	1.74E-00	1.74E-00	1.85E-00	1.05E-02	8.15E-01	2.95E-01	5.54E-02	3.45E-02	6.8E-03
112	Rod			4.88E-01			6.94E-02			3.74E-03		
224	Sol	250	8.54	1.40E-00	1.70E-00	1.50E-00	9.41E-01	5.95E-01	2.39E-01	4.88E-02	2.55E-02	6.5E-03
224	Rod			9.38E-01			1.65E-03			8.74E-03		
224	Rinse	600			1.10E-01			9.77E-01			3.87E-02	
224	Strip	300		3.10E-01			5.58E-02			2.58E-03		
CYCLE 4												
7	Sol	30	8.51	6.58E-01	7.78E-01	6.88E-01	2.19E-02	6.49E-01	4.41E-01	9.28E-02	2.88E-02	1.8E-03
63	Sol	30	8.44	1.28E-00	1.38E-00	1.48E-00	3.08E-01	2.78E-01	2.07E-01	8.58E-01	7.79E-01	4.8E-03
148	Sol	30	8.34	1.13E-00	1.13E-00	9.58E-01	9.28E-01	2.07E-01	1.67E-01	3.98E-02	5.63E-01	3.8E-03
240	Sol	250	8.21	1.28E-00	1.38E-00		1.94E-01	1.88E-01		5.89E-01	4.73E-01	
240	Rinse	600			7.28E-02			6.76E-01			3.23E-02	
240	Strip	300		2.58E-01			5.77E-02			2.78E-03		
CYCLE 5												
5	Sol	20	8.04	6.48E-01	4.78E-01		9.18E-01	4.59E-01		3.59E-02	1.38E-02	
61	Sol	20	8.45	9.28E-01	9.38E-01		2.52E-01	2.21E-01		7.25E-01	5.77E-01	
132	Sol	250	8.41	1.78E-00	1.68E-00		1.89E-01	1.85E-01		4.95E-01	4.77E-01	
132	Rinse	600			1.98E-01			1.22E-02			5.68E-02	
132	Strip	300		6.18E-01			1.88E-03			4.55E-03		
DAY	SAMPLE TYPE	VOLUME ml	pH	Co-60			SrY-90			Np-237		
				UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A
4	Sol	20	8.32	4.23E-03	3.68E-03	3.25E-03				3.88E-01	7.21E-01	9.4E-01
4	Rod									<9.81E-01		
14	Sol	20	8.33	3.58E-03	3.97E-03	2.38E-03				7.21E-01	6.76E-01	1.8E-01
63	Sol	20	8.44	4.55E-03	3.73E-03	2.89E-03				4.85E-01	6.76E-01	2.7E-01
63	Rod			2.97E-03						8.58E-01		
112	Sol	20	8.38	3.83E-03	3.79E-03	3.23E-03				5.41E-01	2.25E-01	1.1E-01
112	Rod			2.38E-03						4.85E-01		
224	Sol	250	8.54	3.74E-03	4.89E-03	2.72E-03	9.18E-05			3.88E-01	3.88E-01	4.8E-01
224	Rod						8.11E-04			<2.25E-01		
224	Rinse	600						5.18E-04		<2.25E-01		
224	Strip	300					1.88E-04			<2.25E-01		
CYCLE 4												
7	Sol	30	8.51		8.65E-02		1.62E-05			2.25E-01	2.78E-01	<2.2E-01
63	Sol	30	8.44	1.45E-03	1.65E-03	1.89E-03	3.89E-05			4.85E-01	3.88E-01	1.8E-01
148	Sol	30	8.34	1.21E-03	1.12E-03	1.28E-03	4.38E-05			3.88E-01	3.88E-01	3.1E-01
240	Sol	250	8.21	1.58E-03	1.82E-03		5.38E-05			4.85E-01	4.85E-01	
240	Rinse	600						2.53E-04			<1.35E-01	
240	Strip	300					2.45E-04			<1.35E-01		
CYCLE 5												
5	Sol	20	8.04	2.88E-02	2.88E-02		1.17E-05			5.88E-01	4.95E-01	
61	Sol	20	8.45	6.31E-02	6.48E-02		2.43E-05			4.58E-01	4.58E-01	
132	Sol	250	8.41	6.67E-02	4.95E-02		3.29E-05	3.23E-05		4.95E-01	3.88E-01	
132	Rinse	600						1.98E-04			5.41E-01	
132	Strip	300					3.51E-04			3.88E-01		

UNITS: Solution (Sol), Rinse and Strip samples in pCi/ml for all but Uranium, ug/ml for Uranium.
 Rod samples in pCi/rod for all but Uranium, ug/rod for Uranium.
 NC Not counted because unfiltered fraction was less than detectable.

for the HBR Test Cycles 3, 4, and 5

Am-241			Cm-244			Cs-137			Cs-134		
UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A
9.37E-02	3.16E-02	3.66E-01	1.09E-03	3.41E-02	4.01E-01	4.35E-05	4.32E-05	4.18E-05	1.58E-04	1.55E-04	1.38E-04
9.32E-03			8.56E-03			1.87E-05					
6.08E-02	2.02E-02	9.91E-00	6.93E-02	2.31E-02	9.46E-00	6.09E-05	6.14E-05	4.59E-05	1.83E-04	1.86E-04	1.52E-04
3.73E-02	1.46E-02	7.66E-00	4.73E-02	1.79E-02	6.76E-00	8.51E-05	8.60E-05	7.79E-05	3.04E-04	3.16E-04	2.82E-04
4.32E-03			4.22E-03			1.22E-05			4.12E-03		
3.13E-02	1.61E-02	8.56E-00	3.88E-02	2.08E-02	7.84E-00	8.92E-05	9.01E-05	8.34E-05	3.17E-04	3.02E-04	2.95E-04
4.24E-03			2.48E-03			6.08E-04			2.03E-03		
2.55E-02	1.20E-02	9.46E-00	3.38E-02	1.58E-02	6.76E-00	1.51E-05	1.47E-05	1.5E-06	4.73E-04	4.59E-04	
6.05E-03			4.73E-03			1.47E-05					
	1.97E-02			1.77E-02			6.60E-04			2.16E-03	
1.32E-03			1.16E-03			5.81E-04			1.97E-03		
7.25E-02	3.65E-01	3.16E-00	4.59E-02	5.72E-01	3.16E-00	1.72E-05	1.66E-05	1.63E-05	5.18E-03	5.36E-03	4.44E-03
2.43E-01	1.67E-01	2.12E-00	2.79E-01	1.94E-01	1.88E-00	6.18E-05	6.14E-05	5.14E-05	1.52E-04	1.49E-04	1.54E-04
2.32E-02	9.46E-00	2.39E-00	1.71E-02	9.91E-00	1.88E-00	9.50E-05	9.73E-05	9.41E-05	2.68E-04	2.72E-04	2.64E-04
9.46E-00	8.56E-00		1.17E-01	8.56E-00		1.40E-05	1.42E-05		3.59E-04	3.73E-04	
	1.77E-02			1.41E-02			5.66E-04			1.01E-03	
1.49E-03			1.25E-03			4.39E-04			1.19E-03		
1.46E-02	1.55E-01		1.11E-02	1.71E-01		1.62E-05	1.61E-05		4.69E-03	3.93E-03	
2.25E-01	9.46E-00		1.85E-01	1.04E-01		4.64E-05	4.55E-05		1.66E-04	1.13E-04	
8.11E-00	7.21E-00		1.04E-01	8.56E-00		8.02E-05	7.93E-05		1.85E-04	1.81E-04	
	2.92E-02			2.41E-02			4.86E-04			9.82E-02	
2.58E-03			1.98E-03			6.08E-04			1.34E-03		
Tc-99			Se-79 (Sn-126)			C-14			I-129		
UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A
4.95E-01	4.50E-01	3.69E-01	<9.01E-00			1.62E-01			1.01E-01		
<7.21E-01			<6.76E-01								
5.99E-01	5.99E-01	5.00E-01	<9.01E-00			2.01E-01			2.95E-01		
1.08E-02	1.26E-02	1.13E-02	<9.01E-00			4.01E-01			4.04E-01		
<9.01E-00			<9.01E-01								
1.26E-02	1.34E-02	1.21E-02	<4.50E-00			4.50E-01			3.81E-01		
<9.01E-01			<4.50E-00								
2.21E-02	2.16E-02	2.16E-02	4.95E-01	<2.25E-01	<2.25E-01	2.52E-01			4.65E-01		
<9.01E-01			<2.25E-01						3.63E-02		
	1.17E-01		<2.25E-01			4.95E-00				1.04E-02	
<9.01E-00			3.15E-01						2.30E-02		
1.89E-01	2.39E-01	2.39E-01	<2.25E-01	<2.25E-01	<2.25E-01	6.89E-01			1.24E-01		
1.44E-02	1.22E-02	9.46E-01	<2.25E-01	NC	NC	3.11E-01			2.12E-01		
1.85E-02	1.71E-02	1.53E-02	<2.25E-01	NC	NC	2.57E-01			2.62E-01		
2.38E-02	2.48E-02		<2.70E-01	NC	NC	2.70E-01			2.76E-01		
<9.01E-00			<2.70E-01			<4.50E-00				6.82E-03	
<9.01E-00			<2.25E-01								
2.97E-01	4.73E-01		<1.35E-01	NC		6.31E-00			6.74E-02		
7.21E-01	7.66E-01		<1.35E-01	NC		2.88E-01			1.42E-01		
1.49E-02	1.58E-02		<1.35E-01	NC		2.12E-01			2.19E-01		
	5.41E-00		<1.35E-01			4.95E-00				9.03E-03	
8.11E-00			3.15E-01								

TABLE A.4. Radiochemical

DAY	SAMPLE TYPE	VOLUME ml	pH	U			Pu-239-Pu-240			Am-241-Pu-238	
				UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A	UNFILTER	0.4 um
1	Sol	5	7.926	3.10E-00			1.59E-03			7.30E-03	
6	Sol	10	8.162	4.50E-00	4.40E-00	4.20E-00	5.50E-02	4.85E-02	2.64E-02	2.45E-03	2.04E-03
6	Rod			8.30E-00			9.14E-03			3.29E-04	
15	Sol	5	8.144	4.80E-00			4.32E-03			1.51E-04	
30	Sol	15	8.320	4.80E-00	4.80E-00	5.40E-00	4.85E-02	5.00E-02	1.82E-02	2.11E-03	1.98E-03
30	Rod			2.40E-00			4.14E-03			1.65E-04	
62	Sol	20	8.288	4.90E-00	4.60E-00	4.60E-00	5.27E-02	4.55E-02	1.80E-02	2.17E-03	1.73E-03
62	Rod			1.60E-00			2.91E-03			1.65E-04	
120	Sol	20	8.429	4.80E-00	4.60E-00	4.20E-00	4.86E-02	4.59E-02	2.07E-02	1.93E-03	1.70E-03
120	Rod			1.10E-00			2.06E-03			8.15E-03	
181	Sol	250	8.458	4.00E-00	4.00E-00	4.00E-00	4.55E-02	4.11E-02	2.17E-02	1.77E-03	1.57E-03
181	Rod			2.00E-00			3.32E-03			1.30E-04	
181	Rinse	600			6.10E-01			1.05E-02			3.79E-03
181	Strip	300		3.20E-00			3.79E-03			1.44E-04	
CYCLE 2											
20	Sol	20	8.462	1.40E-00	1.40E-00	1.40E-00	2.48E-02	2.35E-02	1.94E-02	5.18E-02	5.00E-02
20	Rod			1.50E-00			1.95E-03			1.32E-04	
71	Sol	20	8.485	2.10E-00	2.00E-00	2.00E-00	2.07E-02	1.89E-02	1.46E-02	4.28E-02	3.92E-02
71	Rod			3.90E-01			1.50E-03			1.16E-04	
154	Sol	25	8.490	2.60E-00	2.60E-00	2.60E-00	1.73E-02	1.73E-02	1.34E-02	3.65E-02	3.40E-02
195	Sol	250	8.490	2.40E-00	2.20E-00	2.10E-00	1.80E-02	1.71E-02	1.44E-02	3.78E-02	3.38E-02
195	Rod			1.10E-00			1.95E-03			1.47E-04	
195	Rinse	600			6.50E-02			4.41E-01			1.57E-03
195	Strip	300		5.20E-01			6.80E-02			2.49E-03	
DAY	SAMPLE TYPE	VOLUME ml	pH	Co-60			SrY-90			Np-237	
				UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A	UNFILTER	0.4 um
1	Sol	5	7.926	3.41E-04							
6	Sol	10	8.162	1.83E-05	1.82E-05	1.42E-05					
6	Rod			6.76E-02							
15	Sol	5	8.144	3.53E-05							
30	Sol	15	8.320	4.95E-05	4.86E-05	2.91E-05					
30	Rod			5.41E-04							
62	Sol	20	8.288	5.59E-05	5.50E-05	4.28E-05				1.80E-00	
62	Rod			2.95E-04							
120	Sol	20	8.429	6.17E-05	6.26E-05	5.32E-05				9.46E-01	1.04E-01
120	Rod			9.05E-04							
181	Sol	250	8.458	6.17E-05	6.04E-05	5.54E-05				4.50E-01	4.50E-01
181	Rod			2.30E-05							
181	Rinse	600			7.93E-04						2.25E-01
181	Strip	300		1.66E-04						4.50E-01	
CYCLE 2											
20	Sol	20	8.462	1.21E-05	1.22E-05	1.07E-05				2.25E-01	2.25E-01
20	Rod			6.26E-04							
71	Sol	20	8.485	1.50E-05	1.55E-05	1.22E-05				4.50E-01	4.50E-01
71	Rod			9.82E-04							
154	Sol	25	8.490	1.45E-05	1.47E-05	1.05E-05	3.42E-05				
195	Sol	250	8.490	1.32E-05	1.36E-05	1.13E-05	3.56E-05			3.60E-01	4.95E-01
195	Rod			4.73E-05			2.61E-05			1.80E-00	
195	Rinse	600			2.69E-03			9.01E-03			2.25E-01
195	Strip	300		3.52E-03			5.23E-04			2.25E-01	

UNITS: Solution (Sol), Rinse and strip samples in pCi/ml for all but Uranium, ug/ml for Uranium.
Rod samples in pCi/rod for all but Uranium, ug/rod for Uranium.

• Am-241 values through 120-day rod sample calculated from Pu-239-Pu-240 and Am-241-Pu-238 values using Pu-

ta for the TP Test Cycles 1 and 2

Am-241				Cm-244				Cs-137				Cs-134			
10 A	UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A			
	4.44E-03			4.44E-03			1.12E-07			9.32E-05					
1.36E-02	1.47E-03	1.16E-03	6.36E-01	1.58E-03	1.06E-03	3.06E-01	1.38E-07	1.32E-07	1.25E-07	1.09E-06	1.09E-06	1.05E-06			
	1.65E-04			1.53E-04			6.35E-05			4.45E-04					
	7.34E-03			7.52E-03			1.42E-07			1.16E-06					
0.89E-02	1.24E-03	1.08E-03	1.34E-01	1.28E-03	1.07E-03	1.08E-00	1.42E-07	1.41E-07	1.38E-07	1.13E-06	1.12E-06	1.08E-06			
	9.06E-03			7.84E-03			3.97E-05			3.23E-04					
1.46E-02	1.23E-03	9.14E-02	9.34E-00	1.22E-03	9.10E-02	1.44E-01	1.42E-07	1.40E-07	1.34E-07	1.13E-06	1.09E-06	1.05E-06			
	5.33E-03			4.91E-03			3.86E-05			2.48E-04					
1.00E-02	1.06E-03	8.79E-02	8.88E-00	1.06E-03	8.87E-02	2.39E-01	1.28E-07	1.32E-07	1.30E-07	9.58E-05	9.73E-05	9.55E-05			
	4.46E-03			4.04E-03			5.27E-05			3.84E-04					
1.64E-02	9.73E-02	8.24E-02	7.61E-01	1.00E-03	8.51E-02	7.84E-01	1.23E-07	1.24E-07	1.25E-07	8.42E-05	8.56E-05	8.78E-05			
	6.80E-03			6.17E-03			7.07E-05			4.91E-04					
		1.85E-02			1.73E-02			1.31E-05			9.05E-04				
	7.25E-03			5.36E-03			4.59E-05			3.17E-04					
1.05E-02	8.15E-01	5.86E-01	4.50E-00	7.21E-01	5.77E-01	3.68E-00	2.26E-05	2.28E-05	2.17E-05	1.46E-04	1.26E-04	1.34E-04			
	1.36E-04			1.18E-04			1.14E-05			<1.02E-04					
1.49E-02	8.11E-01	7.21E-01	5.41E-00	9.01E-01	7.66E-01	4.05E-00	4.23E-05	4.27E-05	3.99E-05	2.82E-04	2.75E-04	2.48E-04			
	1.04E-04			1.09E-04			3.79E-04			<1.24E-04					
1.18E-02	8.51E-01	6.49E-01	3.33E-00	8.47E-01	7.03E-01	2.25E-00	6.94E-05	6.76E-05	6.40E-05	4.18E-04	4.32E-04	3.70E-04			
1.39E-02	8.42E-01	6.17E-01	7.21E-00	8.11E-01	5.41E-01	3.68E-00	8.15E-05	8.24E-05	7.93E-05	4.64E-04	5.14E-04	4.73E-04			
	1.45E-04			1.26E-04			1.24E-05								
		8.42E-01			6.13E-01			2.37E-04			1.69E-03				
	1.33E-03			9.58E-02			4.45E-04			2.88E-03					
Tc-99				Se-79				C-14				I-129			
8 A	UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A			
	1.98E-02														
17E-00	2.03E-02	2.25E-02	2.03E-02												
50E-01	2.12E-02	1.89E-02	1.89E-02	<9.01E-00			5.27E-01			5.98E-01					
		1.94E-01													
	2.03E-01														
50E-01	2.38E-01	1.89E-01	2.34E-01												
50E-01	2.79E-01	2.03E-01	4.19E-01												
	5.41E-01						5.41E-01			1.86E-01					
50E-01	4.58E-01	7.21E-01	5.86E-01	<9.01E-00			4.58E-01			2.19E-01					
	<9.01E-01			<9.01E-00											
		<9.01E-00			<9.01E-00	<9.01E-00	5.88E-00			7.31E-03					
	<9.01E-00			<9.01E-00						1.91E-02					

[Pu-239-Pu-240] activity ratio of 1.792.

TABLE A.5. Radiochemical Data

DAY	SAMPLE TYPE	VOLUME ml	pH	U			Pu-239-Pu-240			Am-241-Pu-239	
				UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A	UNFILTER	0.4 um
4	Sol	20	8.41	5.90E-01	5.30E-01	5.10E-01	1.55E-02	1.45E-02	1.20E-02	3.50E-02	3.01E-02
4	Rod			8.80E-01			1.59E-03			6.44E-03	
14	Sol	20	8.38	7.50E-01	7.50E-01	7.50E-01	2.16E-02	1.91E-02	1.35E-02	4.41E-02	3.80E-02
63	Sol	20	8.29	1.12E-00	1.18E-00	1.16E-00	1.98E-02	1.89E-02	1.40E-02	4.07E-02	3.63E-02
63	Rod			1.10E-00			1.03E-03			4.05E-03	
112	Sol	20	8.47	1.17E-00	1.39E-00	1.22E-00	1.93E-02	1.89E-02	1.50E-02	3.88E-02	3.85E-02
112	Rod			4.80E-01			8.92E-02			3.70E-03	
224	Sol	250	8.61	1.20E-00	1.20E-00	1.30E-00	1.97E-02	1.73E-02	1.57E-02	4.01E-02	3.50E-02
224	Rod			8.80E-01			1.27E-03			5.32E-03	
224	Rinse	600			4.20E-02			3.30E-01			1.39E-02
224	Strip	300		5.30E-01			6.44E-02			2.64E-03	
CYCLE 4											
7	Sol	30	8.50	3.70E-01	3.80E-01	3.40E-01	1.39E-02	1.27E-02	9.91E-01	3.12E-02	2.59E-02
63	Sol	30	8.64	7.10E-01	8.10E-01	8.10E-01	2.25E-02	1.57E-02	1.22E-02	2.60E-02	2.80E-02
148	Sol	30	8.16	8.50E-01	8.30E-01	8.40E-01	1.50E-02	1.49E-02	1.12E-02	2.98E-02	2.50E-02
240	Sol	250	7.30	1.10E-00	9.70E-01		1.63E-02	1.28E-02		2.94E-02	2.04E-02
240	Rinse	600			2.20E-02			2.57E-01			1.00E-02
240	Strip	300		9.60E-02			1.42E-02			6.04E-02	
CYCLE 5											
5	Sol	20	8.22	2.40E-01	2.30E-01		9.41E-01	6.70E-01		2.35E-02	1.32E-02
61	Sol	20	8.47	4.80E-01	5.00E-01		1.00E-02	1.04E-02		2.60E-02	1.92E-02
132	Sol	250	8.44	8.70E-01	9.60E-01		1.10E-02	1.06E-02		2.80E-02	2.03E-02
132	Rinse	600			3.60E-02			2.61E-01			1.00E-02
132	Strip	300		1.40E-01			1.67E-02			7.39E-02	
Co-60											
DAY	SAMPLE TYPE	VOLUME ml	pH	UNFILTER	0.4 um	18 A	SrY-90			Np-237	
4	Sol	20	8.41	4.73E-04	4.60E-04	3.55E-04				<2.25E-01	<2.25E-01
4	Rod									<4.50E-01	
14	Sol	20	8.38	4.73E-04	4.82E-04	3.29E-04				1.80E-01	1.80E-01
63	Sol	20	8.29	6.80E-04	5.80E-04	3.67E-04				4.50E-01	4.50E-01
63	Rod			3.64E-04						4.50E-01	
112	Sol	20	8.47	6.62E-04	6.50E-04	5.00E-04				2.70E-01	1.35E-01
112	Rod			6.04E-04						3.15E-01	
224	Sol	250	8.61	6.31E-04	6.44E-04	5.54E-04	1.50E-05			4.50E-01	4.50E-01
224	Rod			1.14E-05			1.80E-05			<2.25E-01	
224	Rinse	600			8.65E-02			8.11E-03		<2.25E-01	
224	Strip	300		2.95E-03			8.50E-03			2.70E-01	
CYCLE 4											
7	Sol	30	8.50	1.40E-04	1.44E-04	1.23E-04	2.70E-04			1.35E-01	<2.25E-01
63	Sol	30	8.64	2.32E-04	2.00E-04	1.82E-04	6.70E-04			1.80E-01	<2.25E-01
148	Sol	30	8.16	3.16E-04	3.00E-04	2.44E-04	1.15E-05			2.70E-01	2.25E-01
240	Sol	250	7.30	3.77E-04	3.61E-04		2.18E-05			5.41E-01	3.60E-01
240	Rinse	600			7.61E-02			5.81E-03		<2.25E-01	
240	Strip	300		2.80E-03			8.20E-03			1.35E-00	
CYCLE 5											
5	Sol	20	8.22	5.45E-03	5.30E-03		1.85E-04			3.15E-01	7.21E-01
61	Sol	20	8.47	1.23E-04	1.22E-04		5.41E-04			2.25E-01	<1.35E-01
132	Sol	250	8.44	1.60E-04	1.60E-04		8.15E-04	8.47E-04		2.70E-01	2.25E-01
132	Rinse	600			3.72E-02			2.12E-03			9.01E-02
132	Strip	300		5.05E-02			6.70E-03				

UNITS: Solution (Sol), Rinse and Strip samples in pCi/ml for all but Uranium, ug/ml for Uranium.

Rod samples in pCi/rod for all but Uranium, ug/rod for Uranium.

NC: Not counted because unfiltered fraction was less than detectable.

a) for the TP Test Cycles 3, 4, and 5

Am-241				Cm-244				Cs-137				Cs-134			
18 A	UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A			
11E-02	8.58E-01 3.89E-03	4.95E-01	5.41E-00	8.29E-01 2.99E-03	5.89E-01	3.08E-00	5.83E-04 8.87E-04	5.81E-04	5.18E-04	3.83E-03	3.34E-03	3.35E-03			
25E-02	9.91E-01	5.45E-01	2.70E-00	8.28E-01	5.59E-01	1.88E-00	8.88E-04	7.58E-04	7.88E-04	5.14E-03	3.79E-03	5.88E-03			
33E-02	8.65E-01 2.39E-03	5.77E-01	1.80E-00	8.33E-01 2.82E-03	5.98E-01	9.01E-01	2.85E-05 5.63E-04	2.88E-05	1.88E-05	1.87E-04	1.18E-04	1.88E-04			
41E-02	7.39E-01 1.58E-03	5.14E-01	2.70E-00	8.82E-01 1.78E-03	5.88E-01	3.98E-00	2.54E-05 3.17E-04	2.53E-05	2.32E-05	1.36E-04	1.37E-04	1.27E-04			
48E-02	8.87E-01 2.82E-03	4.95E-01	4.95E-00	7.39E-01 2.44E-03	4.77E-01	4.58E-00	4.43E-05 2.82E-05	4.59E-05	4.55E-05	1.86E-04	2.19E-04	2.36E-04			
		7.03E-01			4.41E-01			1.56E-04							
	1.31E-03			9.91E-02			4.84E-04			2.27E-03					
52E-02	7.52E-01	7.16E-01	2.70E-00	5.95E-01	3.38E-01	9.01E-01	5.14E-04	4.95E-04	5.18E-04	2.55E-03	3.16E-03	2.82E-03			
59E-02	4.41E-01	3.29E-01	2.61E-00	3.78E-01	2.88E-01	1.88E-00	1.78E-05	1.78E-05	1.75E-05	6.53E-03	7.75E-03	7.38E-03			
78E-02	6.31E-01 6.49E-01	3.33E-01	3.24E-00	4.73E-01 6.28E-01	2.84E-01	1.49E-00	3.19E-05 4.88E-05	3.14E-05	3.02E-05	1.34E-04	1.32E-04	1.32E-04			
		4.58E-00 6.58E-01			3.68E-00 4.05E-01			4.82E-05 1.68E-04		1.75E-04	1.74E-04	5.14E-02			
	3.44E-02			2.38E-02			1.25E-04			4.82E-02					
	6.58E-01 3.78E-01 7.38E-01	7.48E-00 2.76E-01 3.68E-01		4.82E-01 3.29E-01 3.47E-01	5.88E-00 2.61E-01 2.93E-01		3.48E-04 1.18E-05 2.28E-05	3.33E-04 1.17E-05 2.24E-05		1.29E-03 4.39E-03 7.79E-03	1.33E-03 4.47E-03 7.78E-03				
	4.48E-02	6.58E-01		2.97E-02	4.05E-01		1.83E-04	9.68E-03		3.54E-02	3.15E-02				
Tc-99				Se-79 (Sn-126#)				C-14				I-129			
18 A	UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A	UNFILTER	0.4 um	18 A			
5E-01	<9.01E-00 <7.21E-01	<9.01E-00	<9.01E-00	<9.01E-00 <6.78E-01			8.56E-00			4.82E-02					
5E-01	1.48E-01 <9.01E-00	1.48E-01	9.91E-00	<9.01E-00 <9.01E-00			1.71E-01 8.38E-01			5.67E-02 1.13E-01					
5E-01	<9.01E-00	2.25E-01	1.53E-01	<9.01E-01											
5E-01	2.18E-01 <9.01E-01	2.76E-01	1.62E-01	<4.58E-00 <9.01E-00			5.14E-01			1.18E-01					
5E-01	2.52E-01 <9.01E-01	3.88E-01	4.58E-01	<2.25E-01# <2.25E-00#	<2.25E-01#		2.43E-01			1.48E-01 2.36E-02					
		<9.01E-00		<2.25E-01#			4.23E-01			3.91E-03					
	1.49E-01			3.88E-01#						1.61E-02					
5E-01	1.78E-01	9.91E-00	<9.01E-00	3.88E-01#	4.58E-01#	<2.25E-01#	7.57E-01			3.31E-02					
5E-01	7.21E-01	3.88E-01	4.58E-01	<2.25E-01#	NC	NC	2.48E-01			5.81E-02					
5E-01	2.70E-01	2.83E-01	3.15E-01	<2.25E-01#	NC	NC	3.28E-01			9.25E-02					
	5.41E-01	5.41E-01		<2.70E-01#	NC		1.71E-01			1.18E-01					
		<9.01E-00			<2.25E-01#		<4.58E-00			2.27E-03					
	<9.01E-00			<2.25E-01#											
	2.93E-01	2.52E-01		<2.25E-01#	NC		8.11E-00			1.62E-02					
	<9.01E-00	<9.01E-00		<1.35E-01#	NC		2.57E-01			4.15E-02					
	2.57E-01	2.57E-01		<1.35E-01#	NC		2.39E-01			6.42E-02					
		<4.58E-00			<1.35E-01#		<4.58E-00			1.92E-03					
	<4.58E-00			<2.25E-01#											

A.3 RADIOCHEMISTRY ERROR ESTIMATES

The primary sources of error in the reported radiochemistry data are

- volume measurement errors incurred during sample aliquot preparations
- recovery errors involved in radiochemical separations
- counting statistics.

A summary of estimated error resulting from these factors is given below:

- $\frac{^{239+240}\text{Pu}, ^{238}\text{Pu} + ^{241}\text{Am}, ^{244}\text{Cm}}$

Method: Direct plate followed by total alpha counting and alpha spectrometry

Volume Errors: $\pm 2\%$

Recovery: 100% (no separation required)

Counting Statistics at $\pm 1\sigma$:

1 dpm/mL (0.45 pCi/mL)	= $\pm 60\%$
10 dpm/mL (4.5 pCi/mL)	= $\pm 8\%$
100 dpm/mL (45 pCi/mL)	= $\pm 2.5\%$
1,000 dpm/mL (450 pCi/mL)	= $\pm 1.5\%$

The counting statistics for 1, 10, and 100 dpm/mL are based on a 100- μL aliquot plate counted for 480 min with a background of 0.2 cpm. The 1,000 dpm/mL counting statistic is based on a 100- μL aliquot plate counted for 100 min with a background of 1 cpm. (Higher-activity plates are counted on higher-background counters, saving newer, lower-background counters for low-activity samples.)

- ^{241}Am

Method: Separation by anion exchange, plate, alpha count, and alpha spectrometry

Volume Errors: $\pm 3\%$

Recovery: $97 \pm 2\%$

Counting Statistics: Same as above for direct plate alpha, since the same volumes, counting times, and equipment are used

- ^{237}Np

Method: Separation by cation exchange and solvent extraction

Volume Errors: $\pm 2\%$

Recovery: $98 \pm 2\%$

Counting Statistics at $\pm 1\sigma$:

1 dpm/mL (0.45 pCi/mL)	= $\pm 30\%$
10 dpm/mL (4.5 pCi/mL)	= $\pm 6\%$
100 dpm/mL (45 pCi/mL)	= $\pm 4\%$
1,000 dpm/mL (450 pCi/mL)	= $\pm 1\%$

The ^{237}Np counting statistics for 1 and 10 dpm/mL are based on a 200- μL aliquot plate counted for 480 min with a background count of 0.2 cpm. Counting statistics for 100 and 1,000 dpm/mL are based on a 100-min count with a background of 1 cpm.

- ^{99}Tc

Method: Separation by cation exchange and solvent extraction followed by beta proportional counting

Volume Error: $\pm 4\%$

Recovery: 94 $\pm 2\%$

Counting Statistics:

20 dpm/mL (9 pCi/mL) = Lower limit at 2σ
100 dpm/mL (45 pCi/mL) = $\pm 11\%$ at 1σ
1,000 dpm/mL (450 pCi/mL) = $\pm 1.6\%$ at 1σ

The ^{99}Tc counting statistics are based on a 500- μL aliquot extracted into 5 mL with 2 mL plated for beta counting. Counting time is 100 min with a background of 30 cpm.

- ^{137}Cs , ^{134}Cs , ^{60}Co

Method: Gamma spectrometry

Volume Errors: $\pm 2\%$

Recovery: 100% (no separation required)

Counting Statistics at $\pm 1\sigma$:

1,000 dpm/mL (450 pCi/mL) = $\pm 20\%$
10,000 dpm/mL (4,500 pCi/mL) = $\pm 8\%$
100,000 dpm/mL (45,000 pCi/mL) = $\pm 2\%$
1,000,000 dpm/mL (450,000 pCi/mL) = $\pm 1\%$

(Based on 1-mL aliquot counted for 60 min.)

- Uranium

Method: Scintrex UA-3 uranium analyzer, laser-excited fluorescence

Overall error is estimated to be $\pm 10\%$ at 1σ when the instrument is operating in its optimal range. The lower limit is $0.001 \mu\text{g/mL}$ ($\pm 0.001 \mu\text{g/mL}$) using a $100\text{-}\mu\text{L}$ sample aliquot.

A.4 REFERENCE

- A1. Barner, J. O. 1984. Characterization of LWR Spent Fuel MCC-Approved Testing Material ATM-101. PNL-5109, Pacific Northwest Laboratory, Richland, Washington.

APPENDIX B

SOLUTION CHEMISTRY DATA

TABLE B.1. Solution Chemistry^(a) for the C5C-H HBR Test Cycles 1 and 2

	Cycle 1				Cycle 2		
	Start J-13 Water	30 Days	120 Days	223 Days	Start J-13 Water	154 Days	202 Days
pH	7.2	8.21	8.54	8.50	8.0	8.20	8.56
Al	0.11	0.09	0.10	<0.08	<0.08	<0.08	<0.08
B	<0.10	<0.01	0.09	0.26	0.21	0.21	0.23
Ca	15.0	12.7	12.1	12.3	11.2	12.6	12.4
Fe	--	0.21	0.15	0.08	<0.01	<0.01	<0.01
K	5.5	4.5	2.8	2.2	1.95 ^(b)	5.46	5.2
Mg	2.1	1.8	2.1	2.0	0.93	2.00	2.00
Mo	0.08	0.26	0.21	0.20	<0.02	0.08	0.08
Na	49.5	41.6	44.5	45.5	43.1	45.1	44.1
Sr	--	--	--	0.06	0.04	0.05	0.05
Si	31.9	24.5	26.2	32.7	30.6	36.4	36.2
Cl	7.3	7.8	7.3	7.6	7.4	7.7	7.5
F	2.7	2.4	2.1	2.2	2.3	2.4	2.1
PO ₄	2.8	--	--	--	--	--	--
NO ₂	--	~0.5	~0.5	~0.6	--	~1.4	~1.7
NO ₃	8.7	7.4	8.1	8.3	8.3	7.1	6.6
SO ₄	18.8	18.8	18.6	18.5	18.6	18.6	19.8
CO ₃	118.0	--	120.0	118.0	121.5	112.0	112.0

(a) Units in $\mu\text{g/mL}$, 0.4- μm filtered.

(b) Low value attributed to analytical error.

TABLE B.2. Solution Chemistry^(a) for the C5C-H HBR Test Cycles 3, 4, and 5

	Cycle 3		Cycle 4		Cycle 5	
	Start J-13 Water	224 Days	Start J-13 Water	240 Days	Start J-13 Water	132 Days
pH	7.69	8.54	8.00	8.21	7.73	8.41
Al	0.16	0.11	0.075	<0.08	0.08	0.16
B	0.18	0.21	0.29	0.3	0.17	0.15
Ca	12.72	10.94	15.04	12.6	12.2	14.6
Fe	<0.02	<0.01	<0.01	<0.01	<0.01	<0.01
K	6.25	8.14	3.66	6.10	5.7	7.68
Mg	1.85	1.90	2.83	2.01	2.0	2.33
Mo	0.23	0.067	0.04	0.12	0.03	0.09
Na	41.54	41.18	41.52	44.8	46.4	60.8
Si	29.76	35.22	37.78	32.4	36.0	35.2
Sr	--	0.052	0.046	0.04	0.05	0.05
Cl	7.2	7.2	7.0	7.57	6.69	8.37
F	2.19	2.11	2.1	2.42	2.12	2.37
PO ₄	--	--	--	--	--	1.18
NO ₂	--	--	--	--	--	--
NO ₃	7.92	9.7	7.9	6.29	7.55	6.27
SO ₄	18.2	18.9	18.0	18.8	17.04	19.7
CO ₃	131.1	115.4	125.6	126.6	124.5	123.5

(a) Units in $\mu\text{g/mL}$, 0.4- μm filtered.

TABLE B.3. Solution Chemistry^(a) for the I9-24 TP Test Cycles 1 and 2

	Cycle 1			Cycle 2		
	Start J-13 Water	30 Days	181 Days	Start J-13 Water	154 Days	195 Days
pH	7.2	8.32	8.46	8.0	8.49	8.49
Al	0.11	0.89	0.14	<0.08	<0.08	<0.08
B	<0.10	<0.01	0.20	0.21	0.23	0.17
Ca	15.0	12.3	13.1	11.2	12.6	11.6
Fe	--	0.11	0.14	<0.01	0.012	<0.01
K	5.5	1.3	3.5	1.95 ^(b)	4.7	4.8
Mg	2.1	2.0	2.0	0.93	2.0	2.0
Mo	0.08	<0.02	0.104	<0.02	0.036	0.042
Na	49.5	54.9	46.9	43.1	45.6	44.2
Si	31.9	31.4	31.8	30.0	33.1	30.6
Sr	--	--	0.049	0.04	0.043	0.042
Cl	7.3	6.2	7.6	7.4	7.7	7.3
F	2.7	2.4	2.2	2.3	2.4	2.3
PO ₄	2.8	--	--	--	--	--
NO ₂	--	--	~0.4	--	~0.4	~0.2
NO ₃	8.7	7.1	8.1	8.3	9.2	8.3
SO ₄	18.8	21.1	18.8	18.6	19.1	19.1
CO ₃	118.0	--	117	121.5	118	119

(a) Units in $\mu\text{g/mL}$, 0.4- μm filter error.

(b) Low value attributed to analytical error.

TABLE B.4. Solution Chemistry^(a) for the I9-24 TP Test Cycles 3, 4, and 5

	Cycle 3		Cycle 4		Cycle 5	
	Start J-13 Water	224 Days	Start J-13 Water	240 Days	Start J-13 Water	132 Days
pH	7.69	8.61	8.00	7.30	7.73	8.44
Al	0.16	0.22	0.075	<0.08	0.08	0.17
B	0.18	0.22	0.29	0.27	0.17	0.14
Ca	12.72	12.73	15.04	12.4	12.2	14.6
Fe	<0.02	<0.012	<0.01	<0.01	<0.01	<0.01
K	6.25	5.99	3.66	6.4	5.7	4.86
Mg	1.85	2.21	2.83	2.00	2.0	2.21
Mo	0.23	0.077	0.04	0.03	0.03	0.10
Na	41.54	46.68	41.52	44.4	46.4	60.1
Si	29.76	40.44	37.78	36.0	36.0	34.0
Sr	--	0.045	0.046	0.04	0.05	0.05
Cl	7.2	7.4	7.0	12.49	6.69	8.77
F	2.19	2.21	2.1	5.26	2.12	2.39
PO ₄	--	--	--	--	--	--
NO ₂	--	--	--	--	--	--
NO ₃	7.92	9.0	7.9	9.24	7.55	8.76
SO ₄	18.2	18.9	18.0	21.3	17.04	19.1
CO ₃	131.1	129.1	125.6	45.2	124.5	122.5

(a) Units in $\mu\text{g/mL}$, 0.4- μm filtered.

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